NONEQUILIBRIUM STEADY-STATE PHYSICS WITH QUANTUM MASTER EQUATIONS

VON DER FAKULTÄT MATHEMATIK UND PHYSIK DER UNIVERSITÄT STUTTGART
ZUR ERLANGUNG DER WÜRDE EINES DOKTORS DER NATURWISSENSCHAFTEN
(DR. RER. NAT.) GENEHMIGTE ABHANDLUNG

Vorgelegt von

Michael Konopik

aus Nürnberg

Hauptberichter: Prof. Dr. Eric Lutz
Mitberichter: Prof. Dr. Maria Daghofer
Prof. Dr. Tilman Pfau

Tag der mündlichen Prüfung: 24. Juni 2021

Universität Stuttgart

1. Institut für Theoretische Physik

2021

CONTENTS

EINLEITUNG

Egal welches System wir in der Natur betrachten, ultimativ sind alle Teil eines Grösseren, oder um es anders auszudrücken: Es gibt weder im klassischen noch im quantenmechanischen Fall ein komplett von seiner Umgebung isoliertes System. Dynamische Gleichungen wie die Newtonschen Bewegungsgleichungen im klassischen oder die Schrödingergleichung im quantenmechanischen Fall mögen zwar prinzipiell die gesamte Natur beschreiben, doch in der Praxis ist es selbst numerisch nur für relativ einfache Probleme möglich diese Gleichungen auch wirklich zu lösen. Insbesondere ist im Allgemeinen die genaue physikalische Umgebung, häufig auch als das Bad bezeichnet, des betrachteten Systems noch nicht einmal bekannt. Da diese jedoch nicht unbedingt von Interesse ist, ist es vorteilhaft approximative Methoden zu verwenden und die Dynamik des Systems zu modifizieren, wodurch thermodynamisch beobachtbare Verhaltensweisen reproduziert werden können. Es gibt eine Vielzahl solcher Näherungen. Eine Klasse von Näherungen ist durch die sogenannte quantenmechanische Markoff'sche Mastergleichung gegeben. Wird eine Mastergleichung von Grund auf aus einem geschlossenen Modell hergeleitet, so bezeichnet man dies auch als 'globale' Mastergleichung. Je nach Art der betrachteten Systeme ist es jedoch möglich, dass eine solche Herleitung nicht zwangsläufig möglich ist, oder zumindest sehr aufwendig sein kann. In solchen Fällen gibt es heuristische Herangehensweisen oder man kann versuchen, mittels Störungsrechnung eine approximative Mastergleichung herzuleiten. Das Problem dieser Gleichungen ist jedoch, dass deren Gültigkeit meistens nicht betrachtet wird, höchstens a posteriori gerechtfertigt. Diese Arbeit wird sich unter anderem mit eben dieser Gültigkeit beschäftigen. Zuerst betrachten wir den Fall der sogenannten 'lokalen' Mastergleichung, bei dem die System-Bad-Wechselwirkung heuristisch in die dynamische Gleichung des Systems eingepflegt wird. Mittlerweile wird diese Art auch dazu benutzt, Phasenübergangsverhalten von Quantensystemen zu beschreiben. Wir werden diese lokale Mastergleichung mit der Globalen vergleichen und untersuchen, wie sehr diese in der Lage sind, das kritische Verhalten eines Modells zweier gekoppelter harmonischer Oszillatoren wiederzugeben. Unter anderem werden wir für den thermischen Fall die Gibbsverteilung nutzen, um diese mit einem exakten Ergebnis vergleichen zu können. Im Nichtgleichgewicht werden wir die Quanten-Langevingleichung als Vergleichsgröße nutzen. Wir werden zeigen, dass der lokale Ansatz generell Schwierigkeiten hat das stationäre Verhalten quantitativ fehlerfrei zu reproduzieren. Je nach genauer Wechselwirkung kann es sogar sein, dass der lokale Ansatz völlig versagt und kein kritisches Verhalten aufzeigt. Danach betrachten wir einen Störungsrechnungsansatz in der Herleitung der Mastergleichung. Wir betrachten, ob die erste oder zweite Ordnung besser in der Lage ist das Verhalten der globalen Mastergleichung zu reproduzieren. Wir zeigen, dass höhere Ordnungen zwar durchaus verschiedene stationäre Aspekte niedrigerer Ordnungen zu korrigieren vermögen, dies für die betrachteten niedrigen Ordnungen, jedoch nicht für alle Systemeigenschaften gilt. Wir zeigen insbesondere, dass die Störungsrechnung

auch zu Mastergleichungen führen kann, die selbst für Parameter infinitesimal nahe des Entwicklungspunktes unphysikalisch werden. Die Störungs-Mastergleichungen müssen daher mit Vorsicht benutzt werden. Nach dieser Betrachtung verschiedener approximativer Methoden bezüglich der Herleitung einer Mastergleichung betrachten wir dynamische Eigenschaften von allgemeinen (physikalischen) Mastergleichungen. Das Verhalten eines Systems, auf das eine kleine Störung wirkt, wird von den Systemeigenschaften bestimmt, mittels der linearen Antworttheorie. Wir bestimmen die lineare Antwortfunktion des Systems, leiten verschiedene Äquivalenzklassen her und betrachten für diese verschiedene mathematische/ physikalische Eigenschaften. Ein Vorteil dieser verschiedenen Klassen ist, dass diese je nach Kontext vorteilhaft benutzt werden können. Zum Beispiel ist eine Klasse besser dazu geeignet Symmetrieeigenschaften für ein System, das "detailed balanceërfüllt, zu bestimmen und eine andere Klasse ist signifikant einfacher explizit zu berechnen. Wir betrachten verschiedene Beispiele um die Unterschiede in den physikalischen Eigenschaften hervorzuheben, die ein System mit oder ohne "detailed balance"besitzt, und das Versagen der linearen Antwort nahe eines kritischen Punktes. Zuletzt betrachten wir ausführlich Fluktuations-Dissipations Theoreme, die die lineare Antwortfunktion mit Korrelationsfunktionen verbindet, und leiten diese für die verschiedenen Klassen her.

ABSTRACT

No system in nature is completely isolated from its surroundings, regardless if classical or quantum. While equations like the Newton equations in the classical case or Schrödinger equations in the quantum case describe the dynamics of in principle all systems, one is usually only able to calculate the dynamics of small ones. The effects that these surroundings, or baths create generally have to be approximated. One such approximate treatment is given by the quantum Markovian master equation. Deriving a master equation from first principles is usually termed as a 'global' master equation. Depending on the system considered, such derivations are not necessarily easy to accomplish or even possible. In such cases, alternative approaches, or even perturbation theory may be attempted to derive a master equation. However, one has to be careful in using such approaches as their validity is generally not considered, but rather justified a posteriori. In this work we investigate the validity of such approximative master equations. We will first consider the case of 'local' master equations, in which system-bath interactions are heuristically added to the dynamics. This kind of master equation is lately being used to describe phase transition behavior of quantum systems. We will investigate and compare these local master equations with the global one in their ability to reproduce the steady-state behavior of a model of two coupled harmonic oscillators, which expresses a critical behavior at a specific coupling strength. We will compare them with the Gibbs state properties in the equilibrium and the solution of the quantum Langevin equation in the nonequilibrium context. We show that the local approach is generally not able to reproduce the steady-state behavior quantitatively and depending on small changes in the system Hamiltonian, it can also be the case that it fails to even reproduce qualitative effects like the critical behavior itself. Then we consider a perturbative approach for deriving the master equation and investigate how well the first and second order perturbations are able to reproduce the steady-state properties of the global approach. We show that while higher orders improve the description of some aspects, this is not necessarily the case for all. In fact, low orders can create unphysical states even for infinitesimally small deviations about the expansion point. Thus low orders have to be used with caution. After these investigations of approximative approaches, we then consider some dynamical properties of quantum master equations. We treat the dynamical properties of such equations and consider how a system reacts to a small perturbation, by employing linear response theory. We derive different classes of the response functions and consider what properties they possess. The advantage of the different classes is that while one might be better to e.g. investigate mathematical properties like symmetries under quantum detailed balance, while others are easier to calculate. These will further be used to consider different examples emphasizing the differences between detailed balance fulfilling and breaking steady-states or the breakdown of linear response approaching a critical point. Lastly we will investigate fluctuation-dissipation theorems for these response classes, connecting response and fluctuations.

1

INTRODUCTION

All models are approximations. Essentially, all models are wrong, but some are useful. However, the approximate nature of the model must always be borne in mind.

George E. P. Box

1.1. THE DESCRIPTION OF OPEN QUANTUM SYSTEMS

THE development of standard quantum mechanics in the 20th century enabled physicists to calculate physical problems in various new fields, in particular the consistent description of microscopic systems. In this regime one can find that the structure of nature is fundamentally quantized, described by e.g. the emergence of Planck's elementary quantum of action $\hbar$ [1]. With this theory, it was first possible to calculate consistently e.g. the experimentally observed quantizated energy levels of atoms [2], or the quantization of light as described by the photoelectric effect [3] and solve mathematical problems like the quantized harmonic oscillator [1, 4, 5]. While it was and still is successful in describing these and many more problems, the standard quantum mechanics assumes a fundamentally *closed* or isolated system, whose dynamical properties are fully governed by the Schrödinger or von Neumann equation [4, 6], which are fundamentally time-reversal symmetric. These properties are consistent with the fact that the standard quantum mechanics is the generalization of classical, isolated dynamics described by Newtonian or Hamilton mechanics.

However, a (quantum) system can never truly be completely isolated from all of its surroundings. In classical cases there are effects like friction or the description of various chemical reactions [7, 8]. For quantum systems one can observe effects like the decay of an excited atom to its ground state [6, 9], the often experimentally observed thermalization of a system to a Gibbs thermal state [6], or decoherence effects of a state, which cannot be described by standard quantum mechanics. Further, one is often interested in describing equilibrium or nonequilibrium dynamics of a smaller system of interest that is coupled to a single larger system or even multiple ones. If these larger systems

are sufficiently high-dimensional, then the dynamics may not be possible to be solved, even numerically. For special cases, such problems may be solved by interpreting the larger systems as infinitely large baths that are unchanged by the interaction with the small system. Then one can evaluate the effects of these baths in a thermodynamically consistent way.

For being able to dynamically describe such problems, one can either attempt to find a completely new description, or extend, possibly heuristically or approximatively, the closed description by adding additional terms into the dynamical equations [5, 10]. The usual approach in (quantum) mechanics is starting from a closed system model that contains both the system of interest and the additional larger ones. The dynamical equations of these will then be approximated, resulting in effective dynamics. There exists a wide variety of these effective dynamics. Classically, such effective dynamics are described by the concepts of Langevin equations [8, 11, 12], master equations [8, 11] or the Boltzmann equation [11]. For quantum dynamics these effective dynamics are given by e.g. Feynman-Vernon influence functionals [6, 13, 14], stochastic differential equations [6, 8], Redfield equations [6, 8], quantum Langevin equations [15, 16], quantum master equations [6, 14], or numerical quantum simulation techniques like the hierarchical equations of motion [17] or Monte Carlo simulations [6, 18]. Their differing level of applied approximations is often accompanied with a highly different level of complexity for being able to apply their methods.

One widely used approach is given by quantum Markovian master equations. Quantum Markovian master equations have been instrumental in the study of open quantum systems since their introduction by Wolfgang Pauli in 1928 [19]. They offer powerful, yet approximate, means to describe the time evolution of the reduced density operator of quantum systems coupled to external environments [6, 15, 20–23]. They allow the analysis of the dynamics of both diagonal density matrix elements (populations), involved in thermalization processes, and of nondiagonal density matrix elements (coherences), associated with dephasing phenomena. As a consequence, they have found widespread application in many different areas, ranging from quantum optics [24] and condensed matter physics [14] to nonequilibrium statistical mechanics [25] and quantum information theory [26].

More recently, these quantum Markovian master equations started to being used to describe nonequilibrium phenomena of systems that are weakly coupled to multiple baths [10, 27–32]. Further, they were used to describe the behavior of more complex systems that show quantum critical behavior or that posses phase transition characteristics like the description of the open Dicke model [33–35]. Due to their large number of successful applications, there are also many attempts to find general (thermodynamic) properties of these [6, 20, 23, 36]. One such is the field of linear response. Linear response theory describes the dynamical behavior of a system which is subject to a small perturbation and its interesting feature is that the response function of the systems can be obtained from its unperturbed dynamical properties [25, 37, 38], independent of the exact nature of the perturbation. For the nonequilibrium quantum case, some forms of response functions were derived by various authors [39–42]. Linear response is further very fruitful, as one is able to find fluctuation-dissipation theorems. Fluctuation-dissipation theorems connect these linear response functions with fluctuation proper-

ties of the system [38, 39, 41, 42] laying bare thermodynamic properties of them.

The more popular these quantum master equations became, the more they were being used less rigorously. Ideally one derives these master equations properly from first principles [6], which is also usually called the 'global' master equation approach [10, 43]. However, often these derivations are difficult or even maybe impossible to do for general systems and instead heuristic approaches are used. One such is given by the so called 'local' master equations [10], where one simply adds an open dynamics part from a less complicated system, for which the master equation can be derived, to the dynamical equation of a more complex system, without necessarily any derivation. While obtaining such approximative approaches is significantly easier, it was recently was shown that these may cause unphysical behavior [10, 43]. One such example is the emergence of unphysical heat currents [10]. Interestingly, there are also works about a model for which assumptions made in the derivation of the global master equation are wrong and in this case the local approach may even sometimes result in dynamics that are able to more accurately describe the correct system properties [43].

The aim of this thesis is to investigate the validity of such heuristic, or approximative, approaches to quantum Markovian master equations applied to quantum critical phenomena, for both equilibrium and nonequilibrium models. These models are further used to consider the physicality of these approaches. For quantum Markovian master equations that correctly describe the system's dynamics, general physical properties are then considered by extending linear response theory to these general nonequilibrium situations and fluctuation dissipation theorems are being derived.

1.2. OUTLINE OF THE THESIS

Chapter 2 will focus on the differences between local and global master equations concerning steady-state quantum critical behavior of two coupled harmonic oscillators, individually coupled to different baths with potentially unequal temperatures. These approaches are compared with each other using Gibbs states in the equilibrium case and quantum Langevin equations for nonequilibrium situations as benchmarks. Both local properties in the form of local occupation numbers and nonlocal ones like quantum mutual information or negativity will be investigated. The differences between these forms for finite and vanishing temperature will be further explored. It will be shown that while the local approaches is able in same cases to at least qualitatively reproduce the global behavior, it generally fails to properly describe the steady-state behavior of the considered critical system.

Chapter 3 will then consider the derivation of the quantum master equation perturbatively. in this perturbative context, the local approach may be regarded as a 0-th order perturbation and the next order, the first order is being investigated in its possibly improved ability of describing the coupled harmonic oscillator model's steady-state behavior for equilibrium and nonequilibrium situations. Also there the local property of the occupation number and nonlocal ones like quantum mutual information will be compared. Further, thermodynamic properties like heat currents and the uncertainty principle will be compared. It will be shown that while the higher order perturbation does improve the steady-state properties of the description in some aspects, it does not do so across the board, even for infinitesimally small perturbations. It further also pos-

sibly creates unphysical states. Lastly, the second order perturbation will be treated and shown that this order improves the results of the small coupling regime significantly

Chapter 4 will then consider general (dynamical) properties of quantum Markovian master equations by treating linear response theory and deriving open quantum generalizations of classical response function classes found in [37]. For these response function classes symmetries are then investigated, when they fulfill quantum detailed balance. These classes will then further used to calculate examples using the coupled oscillator model where nonequilibrium properties are investigated as well as the breakdown of linear response approaching a critical point, emphasizing the differences different equilibrium and nonequilibrium regimes.

The last chapter 5 will build upon these response function classes and extend the closed dynamics' Callen Welton formulation of a fluctuation-dissipation theorem to the open quantum context. For being able to do so, first the closed dynamics' fluctuation-dissipation theorem will be rederived and the mathematical differences between open and closed quantum dynamics emphasized.

2

GLOBAL AND LOCAL MARKOVIAN MASTER EQUATIONS FOR CRITICAL PHENOMENA

2.1. INTRODUCTION

In the past decade, quantum Markovian master equations have become a popular tool to investigate nonequilibrium phase transitions that occur between (detailed-balance breaking) steady-states [35, 45–62]. Special attention has been given to two broad classes of out-of-equilibrium phase transitions: (i) those induced by external driving fields in systems interacting with a single bath (driven-dissipative processes) [45–51] and (ii) those generated by the coupling of a system to several baths (boundary-driven processes) [35, 52–62]. Remarkably, nontrivial exact analytic steady-state solutions of so called local quantum master equations of the Lindblad type have been obtained for various many-body spin-chain models [35, 52, 55–58, 60, 62].

However, the form of these quantum master equations employed in these studies is often postulated. Their validity is thus not completely clear a priori. This is especially true for boundary-driven processes where the system of interest is coupled to several reservoirs. In this case, it has recently been shown that local master equations, that are commonly used to examine nonequilibrium phase transitions [35, 52–62], may violate the second law of thermodynamics [10] and give rise to nonphysical results, even in the limit of small bath couplings, such as incorrect steady-state distributions or nonzero currents for vanishing bath interactions [43, 63–72]. These inconsistencies are related to the fact that local quantum master equations, whose total dissipator is simply the sum of the local system-bath dissipators derived by neglecting the intra-system coupling, incorrectly neglect subsystems-bath correlations induced by the coupling to the full system Hamiltonian, in contrast to the proper derivation of a quantum master equations,

Parts of this chapter have been published in [44]

termed global master equations [10, 43, 63–72]. Interestingly, the local approach has been shown to provide a better description of quantum heat engines than the global approach in some parameter regimes [67]. The validity of Lindblad master equations has, for example, been discussed in the context of quantum transport [73, 74], quantum relaxation [27, 75], and entanglement generation [76]. But the considered models cannot be used to investigate nonequilibrium phase transition behavior, as they do not possess them.

In this chapter, we examine the accuracy of a quantum-master-equation description of dissipative critical phenomena by analyzing an exemplary system consisting of two interacting harmonic oscillators, each weakly coupled to a thermal reservoir. This system naturally appears in many areas, most notably in cavity optomechanics [77]. Superradiant phase-transition models, such as the Dicke model [78] and the Tavis-Cummings model [79], can also be mapped onto such a system after a Holstein-Primakoff transformation [80, 81] as we will briefly consider in the next part. We concretely compare local and global quantum master equations, with and without rotating-wave approximation for the oscillator-oscillator interaction. We furthermore compare these equations in the thermal case to the expectation of the corresponding Gibbs state. We also will use the results of the quantum Langevin equation [15] as a further control solution in the nonequilibrium case. While a quantum Langevin equation is also only a Markovian approximation [15, 16], a coincidence between the Langevin equation and the (global) master equation may be regarded as an internal consistency check of the Markovian approximation. We explicitly evaluate the stationary mean occupation number of one of the quantum oscillators for various equilibrium and nonequilibrium temperature differences.

We find that the local master equation generally fails to qualitatively reproduce the results of the Gibbs state for equilibrium situations, and the quantum Langevin description in the nonequilibrium case, especially at high temperatures, while the global approach exhibits complete agreement for equilibrium situations and a close behavior in nonequilibrium cases. We show that this feature is directly related to the inability of the local description to correctly capture intersystem correlations, which we quantify with the help of the quantum mutual information and negativity [26].

2.2. THE MODEL AND ITS DYNAMICAL DESCRIPTION

As mentioned above, one field of application of quantum master equations is in the treatment of phase transitions. For these systems it is often the case that a relevant coupling constant has to be sufficiently finite to cause a phase transition. One such examples is given by the so called Dicke model [80, 82–87]. The Dicke model describes the interaction between an ensemble of N atoms, which can be approximated as a collection of identical two-level systems with gap ω_1 that are put in a vacuum cavity, described by the Hamiltonian

$$H_{Dicke} = \omega_1 J_z + \omega_2 a_2^\dagger a_2 + \frac{\lambda}{\sqrt{2j}}(a_2^\dagger + a_2)(J_+ + J_-), \tag{2.1}$$

with J_i the collective spin-algebra of the pseudo-spin of length $j = N/2$. The cavity (light) eigenmode is described by a single mode of frequency ω_2 described by a bosonic

Hilbert space with ladder operators $a_2, a_2^\dagger$. A rotating-wave version is given by neglecting the non quantum-number conserving terms $a_2^\dagger J_+$, $a_2 J_-$. While the rotating-wave approximation is usually associated with a weak-coupling condition, $\lambda/\omega_i \ll 1$, it has recently been shown that counter-rotating terms may be effectively suppressed in modulated systems, even in the ultrastrong regime [88, 89]. The coupling between the atoms and the light-mode is realized by shining laser light perpendicular to the mirrors into the cavity [33, 47, 90]. This laser causes excitations in the atoms and decoherent emissions. Increasing the laser strength, the decoherent emission of the atoms increases and more light is emitted into the cavity-mode. The cavity light intensity causes then stimulated absorption and emission in the atomic gas ensemble, creating a coupling between the atoms and the cavity-mode described by the coupling constant λ. Once the intensity is sufficiently strong, for a critical coupling strength λ_c, this results in a positive feedback loop, creating a coherent state constituted of the atoms in the cavity and the mode field. This sudden change in behavior can be considered as a quantum phase transition [80, 82, 85–87]. Considering purely the Hamiltonian properties of the corresponding Gibbs state for both the Dicke and the rotating-wave version [80, 85, 86], the critical coupling strength is given by $\lambda_{cPP} = \sqrt{\omega_1\omega_2}/2$ for the full Dicke Hamiltonian and $\lambda_{cR} = \sqrt{\omega_1\omega_2}$ for the rotating-wave version at zero temperature. For finite temperatures T and the rotating-wave case, [82, 85, 86] showed that the critical point increases exponentially with the inverse of the temperature, $\beta_c = 1/(kT_c) = 2\omega_2 \operatorname{artanh}(\omega_1\omega_2/\lambda^2)/\omega_1$ for $\lambda > \lambda_{cR}$.

Kopylov et al. [34] considered the effects of open quantum mechanics for the phase transition behavior of the Dicke Hamiltonian. In particular they used the local approach to model the coupling of the cavity mode to the vacuum field, being described by a $T = 0$ bath. Using the Holstein Primakoff transformation and mean-field theory, they found that the critical coupling also depends on the cavity mode-vacuum field coupling γ, $\lambda_{cDo}^2 = (\gamma^2 + 4\omega_1^2)\omega_2/(16\omega_1)$. A similar result was obtained by Dimer et al. [33] who found $\lambda_{cDo}^2 = \omega_2(\gamma^2 + \omega_1^2)/(4\omega_1)$. Their small difference stems from the fact, that Dimer et al. [33] use the local master equation on the level of the collective spins, while Kopylov et al. [34] use the local master equation for the fluctuation terms of a two-harmonic oscillator system. They furthermore considered the fluctuations about the mean-field solution which express a similar behavior. The fluctuations can be described by a Hamiltonian of two coupled Harmonic oscillators [33, 34]. The resulting Hamiltonian from (2.1) is then given by

$$H_{\mathrm{DHO}} = \sum_i \omega_i a_i^\dagger a_i + \lambda(a_1^\dagger a_2 + a_1 a_2^\dagger) + \kappa(a_1 a_2 + a_1^\dagger a_2^\dagger), \tag{2.2}$$

where $\kappa = \lambda$ for the full Dicke Hamiltonian, which will be termed the position-position coupling Hamiltonian in the following, as in this case the intra-system coupling is of the form $x_1 x_2 = (a_1 + a_1^\dagger)(a_2 + a_2^\dagger)$ and $\kappa = 0$ for the rotating-wave Hamiltonian.

This Hamiltonian describes the full Dicke system below the critical points, where the mean-field vanishes [33]. Therefore we will consider its critical behavior approaching these critical coupling strengths for the open quantum case of two coupled harmonic oscillators each locally interacting with a bath, using the local and global quantum master equation and consider how various aspects of their steady-state behavior differ from each other. We will compare these equations with the Gibbs state and the quantum

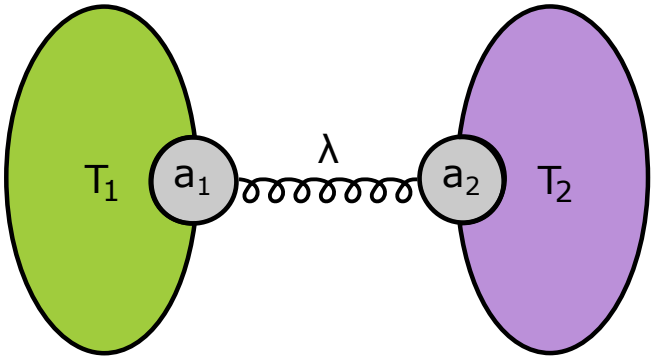

Figure 2.1: Coupled harmonic oscillators. Two quantum harmonic oscillators interact with each other with interaction strength λ, either strongly or weakly $\kappa = \lambda, 0$. Each of them is weakly coupled to a heat bath with respective temperature T_j, $(j = 1,2)$.

Langevin equation. For being able to do so, let's consider first the properties of the Hamiltonian (2.2) and the form of the master equations and Langevin equation.

2.2.1. COUPLED HARMONIC OSCILLATOR PROPERTIES

The toy-model of two coupled harmonic oscillators (2.2), which will be used throughout this work, is a quadratic (bosonic) Hamiltonian. Quadratic Hamiltonians are generally possible to be diagonalized by using an appropriate transformation of the operators [91, 92]. Diagonalizing the Hamiltonian (2.2), one receives a transformed Hamiltonian that has the form of two uncoupled harmonic oscillators $H = \omega_+ d_+^\dagger d_+ + \omega_- d_-^\dagger d_-$, where the coupling is encoded in the diagonal eigenfrequencies $\omega_\pm$.

In the case of the rotating-wave interaction [10], or quantum number conserving interaction $\kappa = 0$, the diagonalized Hamiltonian of H_{DHO} (2.2) has the following form of its eigenfrequencies

$$\omega_\pm^{\mathrm{rw}} = \left(\omega_1 + \omega_2 \pm \sqrt{(\omega_1 - \omega_2)^2/4 + \lambda^2}\right)/2. \tag{2.3}$$

The operator transform needed to receive this diagonalized Hamiltonian is given by use of the rotated operators $d_- = a_2 \cos\theta - a_1 \sin\theta$ and $d_+ = a_1 \cos\theta + a_2 \sin\theta$, where the angle θ satisfies $\cos^2\theta = (\omega_1 - \omega_-^{\mathrm{rw}})/(\omega_+^{\mathrm{rw}} - \omega_-^{\mathrm{rw}})$ [10]. The ω_-^{rw} frequency is positive only for $\lambda < \lambda_{cR}$ and vanishes at the critical point. For coupling strengths λ larger than the critical one $\lambda > \lambda_{cR}$, the frequency becomes negative and thus the Hamiltonian is no longer bound from below, thus the Hamiltonian (2.2) becomes unphysical. The positive eigenfrequency ω_+^{rw} always stays positive.

For the position-position case $\kappa = \lambda$ on the other hand, the eigenfrequencies are given by [82]

$$\omega_\pm^{\mathrm{pp}} = \left[\left(\omega_1^2 + \omega_2^2 \pm \sqrt{(\omega_1^2 - \omega_2^2)^2 + 16\lambda^2\omega_1\omega_2}\right)/2\right]^{\frac{1}{2}}. \tag{2.4}$$

The diagonalization of the position-position Hamiltonian (2.2) is more involved as it couples all four ladder operators with each other, $(a_1, a_2, a_1^\dagger, a_2^\dagger) = S(c_1, c_2, c_1^\dagger, c_2^\dagger)$ [82].

The 4×4 diagonalization matrix S is partitioned into four blocks with the 2×2 matrix A on the diagonal blocks and 2×2 matrix B matrix on the off-diagonal blocks:

$$A=\begin{pmatrix}\frac{(\omega_+^{\mathrm{pp}}+\omega_1)\cos\theta}{2\sqrt{\omega_+^{\mathrm{pp}}\omega_1}} & \frac{-(\omega_-^{\mathrm{pp}}+\omega_1)\sin\theta}{2\sqrt{\omega_-^{\mathrm{pp}}\omega_1}}\\ \frac{(\omega_+^{\mathrm{pp}}+\omega_2)\sin\theta}{2\sqrt{\omega_+^{\mathrm{pp}}\omega_2}} & \frac{-(\omega_-^{\mathrm{pp}}+\omega_2)\sin\theta}{2\sqrt{\omega_-^{\mathrm{pp}}\omega_2}}\end{pmatrix}\ B=\begin{pmatrix}\frac{(-\omega_+^{\mathrm{pp}}+\omega_1)\cos\theta}{2\sqrt{\omega_+^{\mathrm{pp}}\omega_1}} & \frac{(\omega_-^{\mathrm{pp}}-\omega_1)\sin\theta}{2\sqrt{\omega_-^{\mathrm{pp}}\omega_1}}\\ \frac{(-\omega_+^{\mathrm{pp}}+\omega_2)\sin\theta}{2\sqrt{\omega_+^{\mathrm{pp}}\omega_2}} & \frac{(\omega_-^{\mathrm{pp}}-\omega_2)\sin\theta}{2\sqrt{\omega_-^{\mathrm{pp}}\omega_2}}\end{pmatrix}. \tag{2.5}$$

Similar to the rotating-wave case, the eigenenergy ω_+^{pp} stays positive for all λ, ω_-^{pp} is only positive for $\lambda<\lambda_{cPP}$ and vanishes at the critical point. However, for larger coupling $\lambda>\lambda_{cPP}$ the eigenfrequency becomes imaginary $\omega_-^{\mathrm{pp}}\in i\mathbb{R}$. Thus the Hamiltonian (2.2) becomes always unphysical above the critical point, regardless of the type of coupling. The vanishing eigenfrequency also causes that, at the critical point, the ground state becomes infinitely degenerate.

It should be stressed, that this critical behavior of the coupled oscillator Hamiltonian (2.2) is in contrast to the commonly treated (classical) Hookian interaction $(x_1-x_2)^2$ [27, 73–76], which does not have such a critical point. This is the case, because the additional quadratic terms x_i^2 also modifies the local frequencies of the harmonic oscillators, preventing the intra-system coupling terms to reach the critical point.

With the help of these operator transformations, it is straightforward to calculate any moment of the thermal Gibbs state. Any observable $\langle A_iA_j\rangle=\mathrm{Tr}\{A_iA_j\rho_{eq}\}$ for arbitrary operators $A_i,A_j=a_1,a_2,a_1^\dagger,a_2^\dagger$ can be directly calculated by evaluating the Gibbs state using the diagonal form of (2.2), for which any expectation value can be directly calculated.

2.2.2. GLOBAL AND LOCAL MASTER EQUATIONS

One of the most well studied cases of a Lindblad master equation is the evolution of a single harmonic oscillator subject to interactions with its surroundings [6, 15, 93, 94]. The bath is modeled to be comprised of a large number of uncoupled oscillators. Its master equation is of the form (neglecting lamb shift terms)

$$\begin{aligned}\dot\rho=&-i[H,\rho]+\mathscr{D}(\rho)=-i[H,\rho]+\gamma N(\beta,\omega)(a^\dagger\rho a-\{aa^\dagger,\rho\}/2)\\&+\gamma(N(\beta,\omega)+1)(a\rho a^\dagger-\{a^\dagger a,\rho\}/2),\end{aligned} \tag{2.6}$$

with the friction coefficient of the system-bath interaction γ and the Bose-Einstein distribution $N(\beta,\omega)=1/(e^{\beta\omega}-1)$. The master equation (2.6) describes the open dynamics of the single oscillator well, however, the considered general setup in Fig. 2.1 is comprised of two oscillators that each are exclusively coupled to a different bath, usually necessitating a rederivation of the master equation.

Nevertheless, the local approach utilizes the dissipator of (2.6) by heuristically adding this dissipator for each bath interaction with an oscillator. Thus, the local master equation can be written in the form

$$\dot\rho=-i[H,\rho]+\mathscr{D}_1^{\mathrm{lc}}(\rho)+\mathscr{D}_2^{\mathrm{lc}}(\rho) \tag{2.7}$$

with the dissipator for each one of the baths given as in (2.6)

$$\mathscr{D}_i^{\mathrm{lc}}(\rho)=\gamma_i(\omega_i)N(\beta_i,\omega_i)(a_i^\dagger\rho a_i-\{a_ia_i^\dagger,\rho\}/2)+\gamma_i(\omega_i)(N(\beta_i,\omega_i)+1)(a_i\rho a_i^\dagger-\{a_i^\dagger a_i,\rho\}/2). \tag{2.8}$$

Considering Eq. (2.7) and (2.8), it is interesting to note that the local dissipators try to thermalize the mode i at $\beta_i\omega_i$.

The global approach on the other hand is properly derived from first principles using a number of approximations, which will be sketched in the following and the dissipators given. Starting from first principles [6, 27], the complete system consists of the Hamiltonian

$$H = H_S + H_{B1} + H_{B2} + H_{I1} + H_{I2}, \tag{2.9}$$

where the $H_{Bi} = \sum_k \omega_{ik} b_{ik}^\dagger b_{ik}$ is the Hamiltonian for the i-bath of an ensemble of harmonic oscillators with operators $b_{ik}, b_{ik}^\dagger$. The interaction between a bath and its corresponding coupled subsystem is given by $H_{Ii} = \sum_k \varepsilon_{ik}(a_i b_{ik}^\dagger + a_i^\dagger b_{ik})$. The dynamics for the density operator ρ of the full system + baths is given by the von Neumann equation. We assume that the two baths are completely uncoupled $\langle B_{ik} B_{jk}\rangle = 0$ for all $B_{ik} = b_{ik}, b_{ik}^\dagger$, $i \neq j$ and both of them are thermal baths $\left\langle b_{ik}^\dagger(t) b_{ik'}(s)\right\rangle = N(\beta_i,\omega_i)\delta_{kk'}\delta(t-s)$. Using the interaction picture form of the density operator $\rho^I(t) = U_0^\dagger(t)\rho(t)U_0(t)$, with $U_0(t) = \exp(-i(H_S + H_{B1} + H_{B2})t)$, the interaction picture version of the von Neumann equation is given by $\mathrm{d}_t\rho^I(t) = -i\sum_j [H_{Ij}^I(t), \rho^I(t)]$. This operator differential equation can be formally integrated. Reinserting the formal solution into the interaction picture dynamics and tracing over all bath degrees of freedom, one receives

$$\dot\rho^I(t) = -\sum_{ij}\int_0^t \mathrm{Tr}_B\left\{\left[H_{Ii}^I(t), \left[H_{Ij}^I(s), \rho^I(s)\right]\right]\right\}\mathrm{d}s \tag{2.10}$$

where Tr_B is the trace over all bath degrees of freedom. Due to the fact, that the two baths are completely uncorrelated, the system-bath coupling terms in (2.10) of the form $H_{Ij}H_{I\ell}$ will result in expectation values of the form $\langle B_{jk}^I(t) B_{\ell k}^I(s)\rangle = 0$, which vanish and thus the double commutators in (2.10) completely decouple into two double commutators that only contain interaction-terms of one of the baths. Therefore, the following derivation will be done using only one of the baths, whose derivation can be analogously applied to the second one. In the following, for the sake of notational simplicity, the bath density operator for the first bath ρ_{B1} will be replaced by ρ_B and the interaction Hamiltonian H_{I1} by H_I. The system and baths are also assumed to be initially completely uncoupled.

To simplify the differential equation (2.10), three approximations are done. The first one is the so called Born approximation, which assumes that the system and bath are only weakly coupled. Due to this weak coupling, it is assumed that the density matrix can be approximately described by a product state $\rho(t) = \rho_S(t)\otimes\rho_B$. This can be understood as follows: the weak coupling causes only very small correlations being created between the system and the bath. Ignoring these small correlations will result only in negligible errors. The second is the Markovian approximation, replacing $\rho_S^I(s) = \rho_S^I(t)$. Further time coarse-graining is done by removing the short time behavior by substituting s with $t-s$ and taking the upper time integral limit to infinity. This procedure results in the Redfield equation [6] in the interaction picture

$$\dot\rho_S^I(t) = -\int_0^\infty \mathrm{Tr}_B\left\{\left[H_I^I(t), \left[H_I^I(t-s), \rho_S^I(t)\otimes\rho_B\right]\right]\right\}\mathrm{d}s. \tag{2.11}$$

Equation (2.11) does no longer depend on the past of ρ_S^I, but it still a integro-differential equation. To remove this integral, the interaction Hamiltonian has to be explicitly inserted. The interaction Hamiltonian can be written in the general quadratic form $H_I = \sum_\ell A_\ell B_\ell$, where $\vec{A} = (a_1, a_1^\dagger)$ and $\vec{B} = (\sum_k \varepsilon_k^* b_k^\dagger, \sum_k \varepsilon_k b_k)$. The interaction picture bath operators are given straightforwardly by $b_\ell^{\dagger I} = e^{i\omega_k t} b_\ell^\dagger$ and $b_\ell^I = e^{-i\omega_k t} b_\ell$ due to the bath being comprised of an ensemble of uncoupled harmonic oscillators, $i[H_B, b_k] = -i\omega_k b_k$. The interaction picture time evolution of the system-operators is more complex to treat, since the system Hamiltonian (2.2) contains the additional intra-system coupling. Using the diagonalization transformations considered in chapter 2.2.1, any linear operator can be written as a linear combination of the operators in the basis where the Hamiltonian (2.2) is diagonal, $(a_1, a_2, a_1^\dagger, a_2^\dagger) = M(d_+, d_-, d_+^\dagger, d_-^\dagger)$. Thus, the system-operators can be written as a linear combination of the four diagonal operators, with the set $S = \{\omega_+, -\omega_+, \omega_-, -\omega_-\}$

$$a_1^I(t) = \sum_{\Omega \in S} L_1^I(\Omega, t), \tag{2.12}$$

where $L_1^I(\Omega, t) = e^{i\Omega t} L_1(\Omega)$, $\Omega = \pm\omega_\pm$ and the $L_1(\Omega)$ corresponds to the matrix elements of a_1, $L_2(\Omega)$ of $a_1^\dagger$, L_3 of a_2 and L_4 of $a_2^\dagger$. The master equation then takes the form

$$\begin{aligned}\dot{\rho}_S^I(t) = &-\mathrm{Tr}\left\{\int_0^\infty \sum_{\Omega_{1,2}\in S} \left[L_1^I(\Omega_1, t)B_1(t), \left[L_2^I(\Omega_2, t-s)B_2(t-s), \rho_S^I(t)\otimes\rho_B\right]\right]\right\} \\ &-\mathrm{Tr}\left\{\int_0^\infty \sum_{\Omega_{1,2}\in S} \left[L_2^I(\Omega_2, t)B_1(t), \left[L_1^I(\Omega_1, t-s)B_2(t-s), \rho_S^I(t)\otimes\rho_B\right]\right]\right\}.\end{aligned} \tag{2.13}$$

Terms of the form $L_i^I L_i^I B_i^I B_i^I$ will vanish because the baths are thermal $\langle B_i^I B_i^I\rangle = 0$. The time-evolution is thus given by eight individual components stemming from the two double-commutators in (2.13). As an example, we will consider one of these terms and consider how they have to be treated. The other terms then can be computed analogously. Considering only the first term in (2.13), one receives

$$\begin{aligned}\dot{\rho}_{S1}^I(t) &= \sum_{\Omega_{1,2}\in S} \int_0^\infty L_1^I(\Omega_1, t) L_2^I(\Omega_2, t-s)\rho(t)\langle B_1^I(t) B_2^I(t-s)\rangle \mathrm{d}s \\ &= \sum_{\Omega_{1,2}\in S} \int_0^\infty L_1(\Omega_1) L_2(\Omega_2) e^{i(\Omega_1+\Omega_2)t}\rho(t) \sum_k |\varepsilon_k|^2 N_k e^{i(\nu_k+\Omega_2)s}\mathrm{d}s.\end{aligned} \tag{2.14}$$

Taking a continuous limit of the number of bath modes, the bath contribution can then be rewritten as

$$\int_0^\infty \sum_k |\varepsilon_k|^2 N_k e^{i(\nu_k+\Omega_2)s}\mathrm{d}s = \int_0^\infty \int_0^\infty |\varepsilon(\nu)|^2 N(\beta_1, \nu) J(\nu) e^{i(\nu+\Omega_2)s}\mathrm{d}s\mathrm{d}\nu \tag{2.15}$$

with $J(\nu)$ the bath's density of state. The time-integral over the exponential function results in two contributions; a principal value part which causes a modification of the Hamiltonian part of the dynamics and the delta distribution part that causes the dissipative behavior [6]. Throughout this work, we assume that the principal value part can

be neglected. The bath contribution then becomes (for $\Omega_2 < 0$)

$$\int_0^\infty \int_0^\infty |\varepsilon(\nu)|^2 J(\nu) N(\beta_1, \nu) e^{i(\nu+\Omega_2)s} \mathrm{d}s\mathrm{d}\nu = \Gamma(-\Omega_2) N(\beta_1, -\Omega_2) \tag{2.16}$$

or vanishes if $\Omega_2 > 0$. Here $\Gamma(\Omega) = |\varepsilon(\omega)|^2 J(\omega)$ is a friction coefficient which is in general dependent on the frequency Ω of the corresponding mode. Inserting Eq. (2.16) into (2.14) one receives

$$\dot{\rho}^I_{S1}(t) = -\sum_{\Omega_{1,2}\in S} L_1(\Omega_1) L_2(\Omega_2) e^{i(\Omega_1+\Omega_2)t} \Gamma(-\Omega_2) N(\beta_1, -\Omega_2). \tag{2.17}$$

Now a third approximation has to be done. Assuming that one is interested only in long-time behavior, one can note that many contributions in (2.17) contain oscillatory factors $\exp(i(\Omega_1+\Omega_2)t)$. In the case that the frequencies do not cancel each other, the oscillations will be averaged out over long times. Thus, ignoring short time behavior, one can drop all contributions that have non vanishing exponentials, in the case (2.17) this means that only the $\Omega_2 = -\Omega_1 < 0$ contributions survive. Such an approximation is generally termed the secular approximation. Reiterating this procedure for the seven other contributions in (2.14) as well as for the second bath, one can derive the global master equation for the coupled harmonic oscillator problem. The resulting master equation will have the general form in the Schrödinger picture and for the operator basis $a_i, a_i^\dagger$,

$$\dot{\rho}_S(t) = -i[H_S, \rho_S] + \sum_{\alpha,\beta=1}^{4} \Gamma(C_\alpha, C_\beta) \left(C_\alpha \rho_S C_\beta - \frac{1}{2} \{C_\beta C_\alpha, \rho_S\} \right), \tag{2.18}$$

for system-bath coefficients $\Gamma(C_\alpha, C_\beta)$ and the vector of operators $\vec{C} = (a_1, a_2, a_1^\dagger, a_2^\dagger)$.

The global dissipators for the weak coupling case $\kappa = 0$ are then given as [10],

$$\begin{aligned}
\Gamma(a_1, a_1^\dagger) &= \gamma_1^+ c^4 + \gamma_1^- s^4 + (\gamma_2^+ + \gamma_2^-) c^2 s^2 \\
\Gamma(a_1^\dagger, a_1) &= \gamma_1^{+\prime} c^4 + \gamma_1^{-\prime} s^4 + (\gamma_2^{+\prime} + \gamma_2^{-\prime}) c^2 s^2 \\
\Gamma(a_2, a_2^\dagger) &= \gamma_2^- c^4 + \gamma_2^+ s^4 + (\gamma_1^+ + \gamma_1^-) c^2 s^2 \\
\Gamma(a_2^\dagger, a_2) &= \gamma_2^{+\prime} c^4 + \gamma_2^{-\prime} s^4 + (\gamma_1^{+\prime} + \gamma_1^{-\prime}) c^2 s^2 \\
\Gamma(a_1, a_2^\dagger) &= \gamma_1^+ c^3 s - \gamma_1^- s^3 c + \gamma_2^+ c s^3 - \gamma_2^- c^3 s \\
\Gamma(a_1^\dagger, a_2) &= \gamma_1^{+\prime} c^3 s - \gamma_1^{-\prime} s^3 c + \gamma_2^{+\prime} c s^3 - \gamma_2^{-\prime} c^3 s,
\end{aligned} \tag{2.19}$$

with $c = \cos\theta$, $s = \sin\theta$, $\gamma_i^\pm = \gamma_i(\omega_\pm)$, $\gamma_i^{\pm\prime} = \gamma_i^\pm e^{-\beta_i \omega_\pm}$ and $\Gamma(a_1^\dagger, a_2) = \Gamma(a_2^\dagger, a_1)$, $\Gamma(a_1, a_2^\dagger) = \Gamma(a_2, a_1^\dagger)$. All other terms vanish. The factors $\gamma_i^\pm$ correspond to the system-bath couplings and have to be chosen sufficiently small, such that the weak coupling approximation holds, $\gamma_i/\omega_i \ll 1$, but otherwise are in principle free parameter unless the bath is exactly known. In this chapter we assume that the baths are one-dimensional, thus $\gamma_i^\pm = \gamma_i$. Also other bath choices are possible, e.g. a three-dimensional bath [10] $\gamma_i^\pm = \gamma_i \omega_\pm^3$, as will be also treated in the next chapter 3.

For the position-position coupling case, the global Lindblad dissipators are more complex. For each individual contribution one has $\Gamma(C_i, C_j) = \Gamma^{ij}_{1,13} + \Gamma^{ij}_{1,31} + \Gamma^{ij}_{2,24} + \Gamma^{ij}_{2,42}$,

with

$$\begin{aligned}\Gamma^{ij}_{1,kl} =&\gamma_1(\omega_+)N(\beta_1,\omega_+)S^k_1S^l_3W^1_iW^3_j+\gamma_1(\omega_-)N(\beta_1,\omega_-)S^k_2S^l_4W^2_iW^4_j\\ &+\gamma_1(\omega_+)[N(\beta_1,\omega_+)+1]S^k_1S^l_3W^3_iW^1_j+\gamma_1(\omega_-)[N(\beta_1,\omega_-)+1]S^k_2S^l_4W^4_iW^2_j\end{aligned} \tag{2.20}$$

for the quantum oscillator 1 coupled to bath 1 at inverse temperature β_1 and with $W = S^{-1}$, where S is the transformation matrix of the Hamiltonian (see chapter 2.2.1). Here the indexes i, j run over 1-4, corresponding to the elements of $(a_1, a_2, a_1^\dagger, a_2^\dagger)$. The indexes k, l correspond to the initially chosen local coupling terms ($1 \to a_1$, $3 \to a_1^\dagger$) in the derivation of the master equation (2.11) before the diagonalization is applied, which can be ordered either as $a_i a_i^\dagger$ or $a_i^\dagger a_i$. Thus, there are 32 different terms corresponding to the 16 unique operator orderings C_iC_j in the dissipators. Expressions for second bath at inverse temperature β_2 are analogous with k, l now combinations of 2,4. As in the local case, the friction coefficients have to be sufficiently small $\gamma_i/\omega_\pm \ll 1$.

The friction coefficients γ_i have to be small as the master equation is derived under the weak coupling assumption.

2.2.3. LANGEVIN EQUATION

Quantum master equations describe the time-evolution of the density operator for an open system and can therefore be considered as a Schrödinger picture description of open dynamics. On the other hand, quantum Langevin equations determine the dynamics of the system operators and therefore may be identified as the Heisenberg picture of the open dynamics [6, 15]. A detailed treatment of various aspects of quantum Langevin equations can be found in the books by Gardiner [15] and Vogel, Gunnar [16]. The general form of a quantum Langevin equation in the bosonic mode description is, for the operators a_i

$$\dot{a}_i(t) = i[H, a_i(t)] - \gamma_i a_i + a_{i,in}(t) \tag{2.21}$$

and analogously for $a_i^\dagger$. Similarly as for the master equation, this equation contains two main contributions. On the one hand, the Hamiltonian part by the commutator and on the other the open part. $-\gamma_i a_i$ is the damping term. The input term $a_{i,in}(t)$ counteracts the damping term and is needed to conserve commutation relations, since $\mathrm{d}_t[a_i^\dagger(t), a_i(t)] = -2\gamma_i \neq 0$ would not vanish. These input terms furthermore enable the system to reach a (physical) steady-state. In general, both the damping term and the input noise are determined by the structure and state the bath is in. For a flat bath spectrum, which corresponds to quantum white noise [15], the input mean occupation number can be given in Fourier space as

$$\langle \tilde{a}_{i,in}(\nu)\tilde{a}^\dagger_{i,in}(\nu')\rangle = 2\gamma_i(1+N(\beta_i,\nu))\delta(\nu-\nu'), \tag{2.22}$$

where $\tilde{a}_{i,in}(\nu) = \int \exp(i\nu t)a_i(t)\mathrm{d}t$, $\tilde{a}^\dagger_{i,in}(\nu) = (\tilde{a}_{i,in}(\nu))^\dagger$ are the Fourier transformed operators.

Compared to classical Langevin equations, this equation is operator valued. Thus its treatment is generally more complex. For quadratic Hamiltonians H_{sys}, however, it is possible to solve for the first and second moments via Fourier transform, with which the operator differential equations 2.21 are transformed into operator identities. These can

be solved by finding a closed set $M\vec{A} = \vec{A}_{in}$, where $\vec{A}, \vec{A}_{in}$ are vectors consisting of the respectively needed system/input operators.

For the Hamiltonians of interest (2.2) with two locally coupled baths, the steady-state solution can be obtained by matrix inversion in Fourier space [47], $M(\nu)\vec{A}(\nu) + \vec{A}_{\text{in}}(\nu) = 0$, with the system vector $\vec{A}(\nu) = (\tilde{a}_1(\nu), \tilde{a}_1^\dagger(-\nu), \tilde{a}_2(\nu), \tilde{a}_2^\dagger(-\nu))$ and the input noise vector $\vec{A}_{\text{in}}(\nu) = (\sqrt{2\gamma_1}\tilde{a}_{1,\text{in}}(\nu)\sqrt{2\gamma_1}\tilde{a}_{1,\text{in}}^\dagger(-\nu), \sqrt{2\gamma_2}\tilde{a}_{2,\text{in}}(\nu), \sqrt{2\gamma_2}\tilde{a}_{2,\text{in}}^\dagger(-\nu))$. The matrix M is given by

$$M(\nu) = \begin{pmatrix} -i\nu + i\omega_1 & 0 & i\kappa & i\lambda \\ 0 & -i\nu - i\omega_1 & -i\lambda & -i\kappa \\ i\kappa & i\lambda & -i\nu + i\omega_2 & 0 \\ -i\lambda & -i\kappa & 0 & -i\nu - i\omega_2 \end{pmatrix} + \bar{\gamma} \tag{2.23}$$

with $\bar{\gamma} = \text{diag}(\gamma_1, \gamma_1, \gamma_2, \gamma_2)$. Inverting $M^{-1} = m$, the second moment $\langle a_1^\dagger a_1 \rangle$ in the algebraic space is given by

$$\begin{aligned} \langle a_1^\dagger a_1 \rangle &= \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \langle a_1^\dagger(\nu) a_1(\nu') \rangle e^{i(\nu-\nu')t}/2\pi \mathrm{d}\nu\mathrm{d}\nu' \\ &= (1/\pi)\int_{-\infty}^{\infty} \gamma_1(|m_{11}|^2 N(\beta_1,\nu) + |m_{12}|^2(N(\beta_1,-\nu)+1)) \\ &\quad + \gamma_2(|m_{13}|^2 N(\beta_2,\nu) + |m_{14}|^2(N(\beta_2,-\nu)+1))\mathrm{d}\nu \end{aligned}$$

and similar expressions for all the other second moments.

It should be noted, that Langevin equations are on a similar footing as master equations. In fact, Markovianity is generally also used to derive the quantum Langevin equation [16]. While they are similar in their approximations, they are still different approximative approaches, thus their results do not necessarily have to coincide. In the case that they do coincide, one might take this as an argument that the considered system is correctly described by such a Markovian description. If they differ, however, this is a possible argument that the description of the considered system may have 'fundamental' non-Markovian effects that should be taken into account for a consistent open dynamics' Heisenberg and Schrödinger picture.

2.3. THE COMPARISON OF GLOBAL AND LOCAL MASTER EQUATIONS

The two Hamiltonian forms of Eq. (2.2), given by $\kappa = 0, \lambda$, result in four different master equations, which will be compared in the following. Their steady-state behavior will be calculated by making use of the characteristic function and symplectic spaces (details can be seen in the appendix 7.1). We start by first considering the equilibrium case for them.

2.3.1. EQUILIBRIUM APPROACH TO THE CRITICAL POINT

The first quantity of interest is a local one; the mean occupation number of the first oscillator $\langle a_1^\dagger a_1 \rangle_{SS}$ in dependence of the reduced interaction strength between the two oscillators λ/λ_c. λ_c corresponds to the respective critical point of the model, $\lambda_c = \lambda_{cR}, \lambda_{cPP}$.

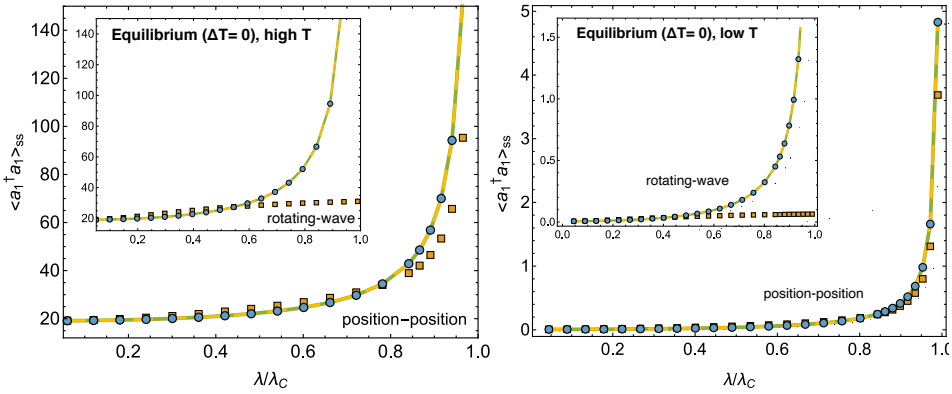

Figure 2.2: Steady-state mean occupation number $\langle a_1^\dagger a_1 \rangle_{\rm ss}$ of the first oscillator as a function of the dimensionless inter-oscillator interaction strength, $\lambda/\lambda_{\rm c}$, for the equilibrium (high-temperature) case $\Delta T = 0$. For the position-position interaction [see Eq. (2.2)], the results of the global quantum master equation (blue dots) perfectly agree with those of the quantum Langevin equation (green line) as well as those of the Gibbs state $\rho_{\rm eq} = \exp(-\beta H)/Z$ (yellow line), while those of the local quantum master equation deviate more and more as the critical point is approached. For the rotating-wave interaction (inset), the global approach still perfectly matches the predictions of the quantum Langevin equation, while the local scheme does not display any critical behavior. Parameters are $\gamma_1 = \gamma_2 = 1.5 \cdot 10^{-4}$, $\omega_1 = 5$, $\omega_2 = 2$ and $T_1 = T_2 = 98$.

Figure 2.2 considers the equilibrium case $\Delta T = T_2 - T_1 = 0$ for high and low temperatures and Fig. 2.3 shows the $T_i = 0$ equilibrium case. For finite temperatures, Fig. 2.2 it is observable there, that there is a perfect agreement between the global quantum master equation (blue dots), the quantum Langevin equation (green line) and the equilibrium (Gibbs) state $\rho_{\rm eq} = \exp(-\beta H_{DHO})/Z$ (yellow line) for all values of $\lambda/\lambda_{\rm c}$, for both the position-position and the rotating-wave (inset) interactions, and for both high as well as low absolute temperatures (left and right figure in 2.2). By contrast, the local quantum master equation (orange squares) deviates from these results in particular as the critical point is approached; noticeably, it does not exhibit any critical behavior at all for the (intra-system) rotating-wave interaction (inset), regardless of low or high temperatures. From the nature of the Hamiltonian, such a behavioral difference is expected for the local master equation. For the position-position coupling case, the unitary dynamics part of the local master equation (2.7) contains the non-quantum number conserving terms $(a_1 a_2 + a_1^\dagger a_2^\dagger)$. These enable the local dynamics to still create nontrivial behavior in the form of the divergence at the critical point. But since the dissipator (2.7) is in this case blind to the intra-system interaction, it is not able to completely replicate the correct behavior. In the rotating-wave case, however, this leads to the complete breakdown of the local approach. In this case, the whole dynamics is quantum number conserving, which renders the Hamiltonian part unable to force the local approach to experience the critical behavior.

Next, let us consider what kind of effect the case $T_i = 0$ creates. Compared to small temperatures, this case is purely quantum even when the critical point is approached $\lambda \to \lambda_c$. Thus, if critical behavior occurs, then one might call it full quantum critical behavior. In Fig. 2.3, the local occupation number of the position-position coupling

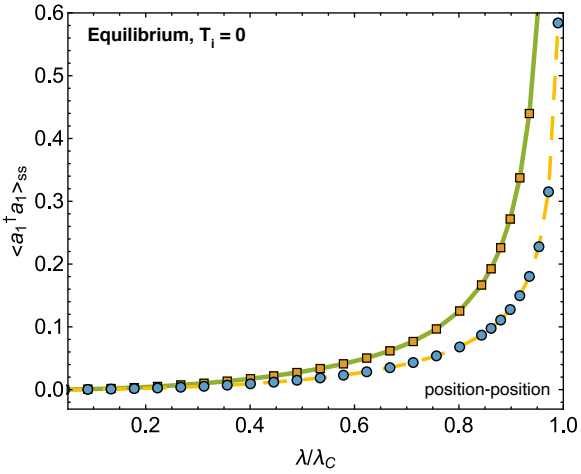

Figure 2.3: Behavior of the equations for $T_i = 0$ of the position-position coupling. In the rotating-wave case $\kappa = 0$, all are identical to 0 and thus not plotted. The global approach (blue dots) follows the (yellow dashed) analytic solution (Gibbs state at $T_i = 0$). Now the local (orange squares) and the quantum Langevin equation (green line) coincide, but do not reproduce the Gibbs state behavior. Parameter same as in Fig. 2.2.

case is plotted for the vanishing temperature case $T_i = 0$. It can be seen there, that at $T_i = 0$ the global master equation still completely follows the Gibbs distribution (yellow dashed). However, now the Langevin equation and the local approach coincide. But these do not coincide with the Gibbs distribution. This remarkable finding might be caused by two facts.

First, $T = 0$ is fundamentally quantum and it is quite questionable how valid the assumption of Markovianity is in this case. In fact, for the harmonic oscillator it was shown [95] that damped dynamics can be connected to weak coupling theory of the damped harmonic oscillator only in the case that $\hbar\gamma \ll kT$. On the other hand, the global approach results in quantum Markovian semigroups of a special kind. The dissipators in (2.19),(2.20) have, in the diagonal basis $d_{\pm}^{(\dagger)}$ of the Hamiltonians (2.2), the form of Davies generators [28], which have in the case $T_1 = T_2$ the Gibbs state as their faithful (steady-) state. The extensive approximations in the global master equation thus force the steady-state to be a Gibbs state even in the $T = 0$ case. Since such a property is not shared by the local as well as for the Langevin approach, these differ and surprisingly seem to coincide.

While the position-position coupling case shows such interesting behavior, the weak-coupling version $\kappa = 0$ is quite trivial. Since the Hamiltonian is completely quantum-number conserving, the ground state is still $|00\rangle$, the direct product of the ground state of the individual harmonic oscillators, regardless of coupling. Thus there does not exist a critical behavior for the weak coupling Hamiltonian. In fact, the steady-state of all the dynamical equations is exactly this ground-state for all coupling values λ/λ_c.

For a deeper understanding of the physical reason of the success or failure of the quantum-master-equation description of dissipative critical phenomena, we will first concentrate on the quantum mutual information between the two harmonic oscillators, $I(\rho) = S(\rho_1) + S(\rho_2) - S(\rho)$, where $S(\rho_i) = -\mathrm{tr}\{\rho_i \ln \rho_i\}$ is the von Neumann entropy and

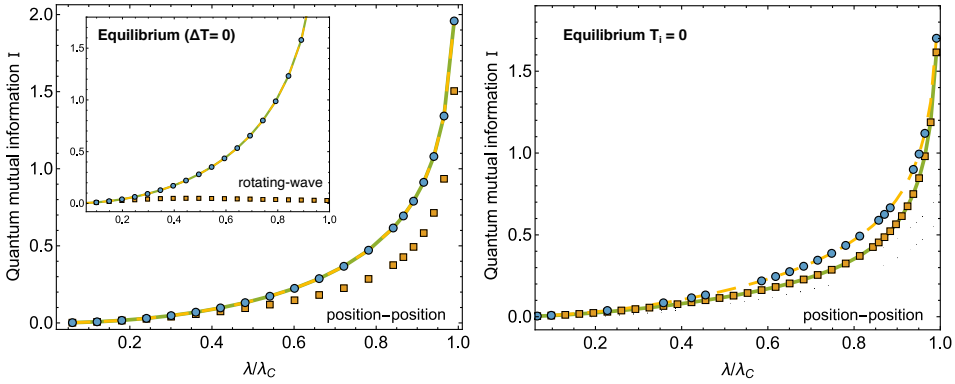

Figure 2.4: Steady-state quantum mutual information $I(\rho_{ss})$ of the two-oscillator system as a function of the dimensionless inter-oscillator interaction strength, λ/λ_c, for the equilibrium (high-temperature) case $\Delta T = 0$ (left) and the $T_i = 0$ case (right). (left): The mutual information displays an analogous dependence of the interaction strength as the steady-state mean occupation number $\langle a_1^\dagger a_1 \rangle_{ss}$ shown in Fig. 2.2. (right): For vanishing temperature, the Langevin equation coincides with the local approach completely, while the global approach coincides with the expected mutual information of the Gibbs ensemble, in accordance with the mean occupation number behavior in Fig. 2.3. Same parameters as in Fig. 2.2.

$\rho_i = \mathrm{tr}_i \rho$ are the reduced density operators of the respective harmonic oscillators and ρ is the density operator of the whole system [26] (see appendix 7.2 for details). The quantum mutual information is a measure of the total (classical and quantum) correlations between two subsystems and has been used broadly to characterise critical transitions [96–100].

Figure 2.4 shows the mutual information for the position-position coupling case at finite (left) and vanishing temperature (right). The inset contains the finite temperature rotating-wave interaction. The left figure in 2.4 shows that the stationary quantum mutual information $I(\rho_{ss})$ displays a very similar dependence on the interaction strength λ/λ_c as the average occupation number $\langle a_1^\dagger a_1 \rangle_{ss}$ represented in Fig. 2.2, both for the position-position and rotating-wave inter-oscillator interactions. The insufficiencies of especially the local quantum-master equation approach, may thus be caused by its inability to correctly reproduce the intersystem correlations, in particular close to the critical point. This feature can be confirmed mathematically by looking at the way the respective Lindblad quantum master equations are obtained [10]: the dissipators in the local master equation are indeed derived in the local eigenbasis of each separate harmonic oscillator, while those of the global master equation follow from a diagonalization of the interacting two-oscillator system (the unitary evolution given by the von Neumann term in Eq. 2.20 describes coupled dynamics in both cases). Therefore, the global scheme better accounts for intra-system correlations than the local one.

The right figure in 2.4 shows the same behavior as in the steady-state occupation number case in Fig. 2.3 for temperature $T_i = 0$. Only the global master equation (blue dots) is able to reproduce the mutual information of the Gibbs ensemble ground state (yellow dashed) correctly. Both the Langevin equation (green line) and local master equation (orange squares) are incorrect, but also agree with each other. In particular,

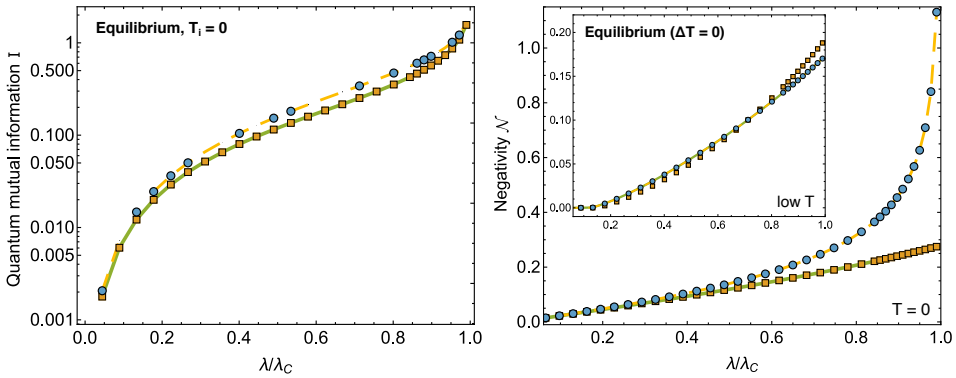

Figure 2.5: Steady-state negativity N of the two-oscillator system as a function of the dimensionless inter-oscillator interaction strength, λ/λ_c, for the equilibrium (low-temperature) case $\Delta T = 0$ (inset) and the $T = 0$ case on the right. On the left, the quantum mutual information is shown in a log plot at vanishing temperature. The low temperature case shows for the negativity, on the right (inset), mostly an agreement between the approaches, with small deviations for the local approach (orange squares). The $T_i = 0$ case shows that both the local and Langevin approach (green line) show a relatively weak entanglement creation approaching the critical point. The global approach (blue dots) and the Gibbs state (yellow line) shows a strong increase in negativity approaching the critical point. Compared with the log plot of the mutual information, this means that the global approach does not produce more correlations per se than the local one, but more coherences are being created. Same parameters as in Fig. 2.2.

both are not able to create sufficiently strong correlations. The quantum mutual information is dependent on the complete state of the dynamics' steady-state and thus the global approach does not just coincide with the Gibbs state for local observables, but completely coincide for equilibrium situations. The result at $T = 0$ further suggests, that the Langevin equation can potentially be described by the local master equation at $T = 0$.

The quantum mutual information considers general correlations between the subsystems, unable to distinguish whether these are classical or quantum. Coherences for two Gaussian systems can be described by the negativity [101]. The negativity is defined by the trace norm of the partial transpose of a state ρ, $||\rho^{T_A}||_1$ of a two-partite Hilbert space $\mathscr{H} = \mathscr{H}_1 \otimes \mathscr{H}_2$, with the trace norm of an operator being defined as $||\rho||_1 = \mathrm{Tr}\{(\rho\rho^\dagger)^{1/2}\}$, for a Hermitian operator ρ. The partial transpose of the A-subspace can be defined by $\langle i_A, j_B|\rho^{T_A}|k_A, \ell_B\rangle = \langle k_A, j_B|\rho|i_A, \ell_B\rangle$. While for density operators it is the case that $||\rho||_1 = 1$, where ρ only has positive eigenvalues, the partial transpose ρ^{T_A}, $\mathrm{Tr}\{\rho^{T_A}\} = 1$ contains also negative eigenvalues if ρ contains entanglement between the two subsystems and thus $||\rho^{T_A}||_1 \neq \mathrm{Tr}\{\rho^{T_A}\}$. Negativity is then defined as

$$\mathscr{N} = \frac{||\rho^{T_A}||_1 - 1}{2}. \tag{2.24}$$

While it is not an entanglement measure, the negativity is an entanglement monotone, i.e. if there exists entanglement, then the negativity becomes finite and qualitatively an increase in negativity means an increase in entanglement [101] (see appendix 7.2 for further details on how the negativity can calculated for Gaussian systems).

Entanglement exists in low temperature quantum situations. Therefore, only for low or vanishing temperatures a finite negativity is expected. Figure 2.5 considers the low

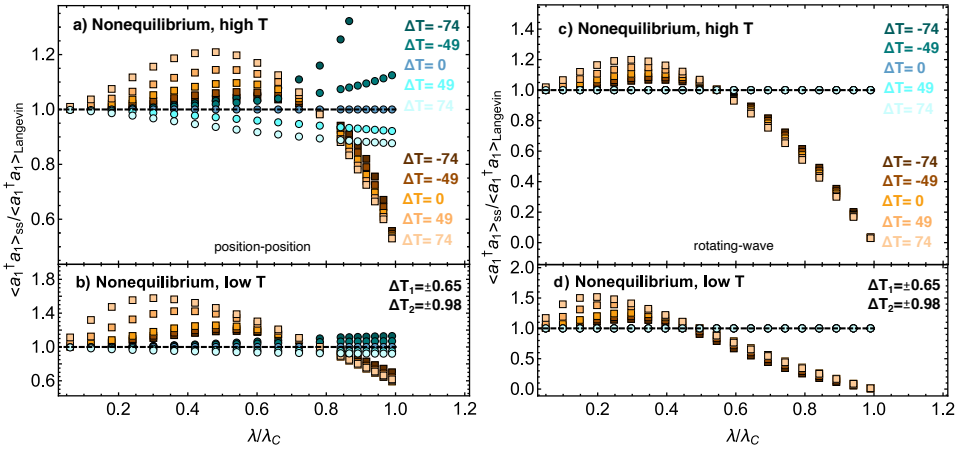

Figure 2.6: Ratio of the steady-state mean occupation numbers $\langle a_1^\dagger a_1\rangle_{\rm ss}/\langle a_1^\dagger a_1\rangle_{\rm Langevin}$ of the quantum master equation and the quantum Langevin equation as a function of λ/λ_c for various nonequilibrium temperature differences ΔT, for position-position interaction (left) and weak rotating-wave interaction (right). (left): In the high-temperature regime a) ($\beta_i\omega_i \ll 1$), The local (squares) approach generally shows larger differences to the Langevin solution than the global one (circles). Approaching the critical point, the local approach always similarly fails,while the differences for the global one depends on the sign of the temperature difference. For low temperatures b) the color coding is the same as for a) with the smaller temperature difference given by ΔT_1 and the larger with ΔT_2 the low temperature behavior is similar for the global approach, while the local one shows a strong erroneous behavior for intermediate coupling strengths. (right) for the rotating-wave Hamiltonian, both high and low temperaturs c),d) show complete coincidence of the global approach and the Langevin equation. The local approach completely fails approaching the critical point since it has no divergence behavior. Same parameters as in Fig. 2.2.

temperature (inset) and the vanishing temperature case $T_i = 0$ in the left figure. At low temperatures, the negativities of the global approach, the Gibbs state and the Langevin equation also coincide and the local approach slightly differs. In total, the negativity behaves linearly approaching the critical point. The $T_i = 0$ case on the other hand shows that the negativity of the global and the Gibbs state one rapidly increase approaching the critical point, whereas the local and the Langevin approach are not able to create such strong entanglement. Interestingly, considering the log plot on the left side in 2.5, one can see that the quantum mutual information between the approaches does not differ too strongly. This means that, the total amount of correlations being created is rather similar. But for the global approach, a larger part of these correlations are comprised of quantum correlations, in particular coherence. Therefore, the local approach creates more classical correlations. This might be caused due to the lack of the nonlocal dissipators of the form $\Gamma(a_1, a_2)$ etc.

2.3.2. NONEQUILIBRIUM APPROACH TO THE CRITICAL POINT

A nonequilibrium situation can be created by introducing two different temperatures for the baths $T_1 \neq T_2$. This will cause a heat current through the system that will drive it into a nonequilibrium steady-state. In the following it will be considered what effects this nonequilibrium situation causes, using the master equation description. For this

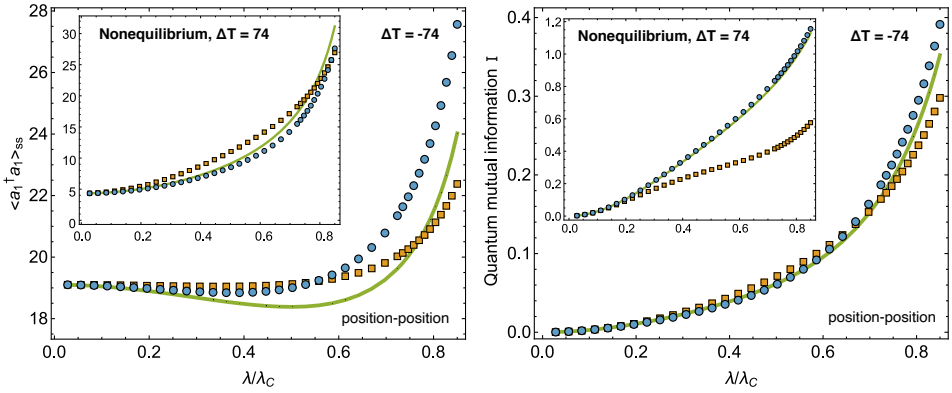

Figure 2.7: Example mean occupation number $\langle a_1^\dagger a_1\rangle_{\rm SS}$ (left) and quantum mutual information (right) in dependence of λ/λ_c for the largest temperature differences $\Delta T = \pm 74$ for the position-position coupling case, explaning the differences between sign changes in the temperature difference . The mean occupation number (left) shows that if the second bath is cooler $\Delta T = -74$, then the Langevin equation (green line) shows an initial cooling of the local occupation number as energy flows to the cooler bath. This is only compensated approaching the critical point. The global approach (blue dots) also resembles this behavior, but weaker. The local approach (orange squares) does not show this behavior. For a colder first bath $\Delta T = 74$, the three approaches are relatively close to each other. Quantum mutual information on the other hand (right figure) shows an interesting inversion of this behavior. While for a cooler second bath $\Delta T = -74$, all approaches have a rather similar mutual information in the considered λ/λ_c interval, the colder first bath $\Delta T = 74$ now shows a strong difference between the local approach and the global/Langevin approaches. These differences are caused by the nonresonant oscillators $\omega_1 \neq \omega_2$. Same parameters as in Fig. 2.2.

nonequilibrium situation, the Gibbs state can no longer be used as an exact benchmark. But, disregarding the vanishing temperature case $T_i = 0$, the quantum Langevin equation showed complete coincidence with the global approach and the Gibbs state in the thermal case. Therefore, in order to gain deeper insight on the nonequilibrium properties, the ratio of the steady-state mean occupation numbers of the master equations and the corresponding quantum Langevin equation expressions, $\langle a_1^\dagger a_1\rangle_{\rm ss}/\langle a_1^\dagger a_1\rangle_{\rm Langevin}$, are examined. This ratio is plotted against the strength of the nonequilibrium ΔT. The temperature differences are accomplished for $\Delta T < 0 : T_2 = T + \Delta T$, and $\Delta T > 0 : T_1 = T - \Delta T$, with the high temperature case $T = 98$ and low temperature case $T = 1.96$.

In the high-temperature regime ($\beta_i\omega_i \ll 1$), the left Fig. 2.6a shows that, for the position-position interaction, the local approach becomes worse as the system moves further away from equilibrium and that even the global approach now departs from the solutions of the quantum Langevin equation for large ΔT. A similar behavior is seen in Fig. 2.6b when the two temperatures are low ($\beta_i\omega_i \gg 1$). Similar results are displayed for the rotating-wave interaction in Figs. 2.6cd on the right side : remarkably, the global quantum master equation here always perfectly matches the quantum Langevin equation, for all λ and all ΔT, while the local quantum master equation always fails to describe critical behavior. That the behavior for high and low temperatures is similar, is somewhat expected approaching the critical point. Since one of the eigenenergies of the system (2.4) vanishes at the critical point, any finite temperature will lead to a diverging occupation number, since $\lim_{\lambda\to\lambda_C} N(\beta_i,\omega_-) \to \infty$. However, for smaller coupling

strengths it is visible that the general behavior differs for the local approach. Lower temperatures cause the difference between Langevin and local master equation to be maximal at lower coupling strengths, for both interaction cases in Fig. 2.6. Further, one can see that the sign of the temperature difference ΔT causes qualitative differences between the global and the Langevin approach. The magnitude of their difference is in part due to the different frequencies of the two harmonic oscillators ω_i which has a nonlinear effect because $N(\beta_i, \omega_i)$ is highly nonlinear.

An example explaining this qualitatively different behavior for the temperature differences of the local observable $\langle a_1^\dagger a_1 \rangle$ is also shown in Fig. 2.7, by considering the high temperature cases $\Delta T = \pm 74$ for the position-position coupling and the mean occupation number (left) as well as the quantum mutual information (right). The case that the second bath is cooler $\Delta T = -74$ shows that the Langevin equation can reproduce a behavior that one would expect in this nonequilibrium situation. If the subsystems are uncoupled $\lambda \approx 0$, then the respective oscillators are thermalized at the corresponding bath temperature T_i. But if the coupling is switched on, then the local mean occupation number (left inset in Fig. 2.7) should decrease as energy is drained from the cooler second oscillator, in dependence of the coupling strength. Only for sufficiently strong coupling strengths, approaching the critical point λ_c, this cooling effect is offset by the vanishing of the eigenfrequency ω_-. To a lesser extend this is also accomplished by the global master equation. The local approach, however, is not able to reproduce this behavior at all. Comparing the mutual information in this case (right figure in Fig. 2.7), one sees that, interestingly, the mutual information of all three approaches is rather similar, until the critical point is approached. On the other hand, for the case that the first bath is colder $\Delta T = 74$, one has a rather similar behavior for all three mean occupation numbers (left Fig. in 2.7). There is no counteracting effect like in the $\Delta T = -74$ case and therefore even the local approach seems to be able to follow the behavior of the global/Langevin equation relatively well until the critical point is being approached. However, the mutual information now has more distinct characteristics for this case (right inset). The local approach has now a larger difficulty reproducing the correlations in the system, while the global and Langevin approaches are quite similar. Of course, this behavior holds true for both local occupation numbers. But since the oscillator frequencies $\omega_1 \neq \omega_2$ are off-resonant, these different signs will be asymmetric about temperature.

2.3.3. SYSTEM-BATH COUPLING DEPENDENCE OF THE CRITICAL POINT

Let us finally consider what effect it has if the system-bath coupling is modified, using the master equation approaches. The Lindblad master equations are derived under the assumption of weak system-bath coupling. Therefore, the validity of these equations can become rather questionable for too large values γ_i. Recalling that, nevertheless, these equations are used to describe critical behavior (see chapter 2.2) for both master and Langevin equations, it is valid to ask how the critical point behaves in dependence of the coupling strength $\gamma = \gamma_1 = \gamma_2$. A log-plot is given in Fig. 2.8 for the critical point of the Langevin equation and the two master equations. The global master equation (blue circles) has a fixed critical point irrespective of the system-bath coupling γ_i. This is expected as by construction the global master equation will reproduce the Gibbs state of any temperature ($T_i = 98$ in this case), which of course does not depend on the details

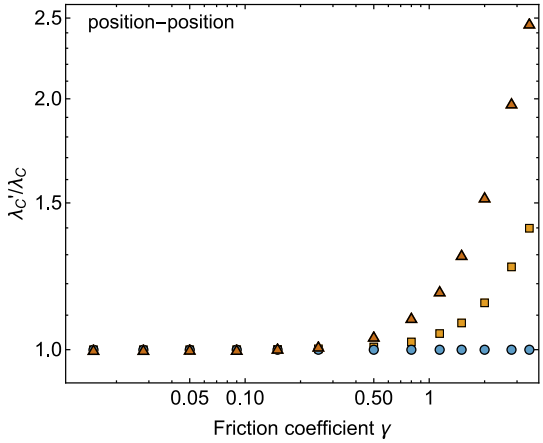

Figure 2.8: Dependence of the value of the normalized critical point λ_c'/λ_c on the coupling strength $\gamma_1 = \gamma_2 = \gamma$ in a log plot for an equilibrium $T_1 = 98 = T_2$. The global master equation (blue circles) always reproduces the system Gibbs state and thus its critical point does not change $\lambda_c' = \text{const}$. The Langevin equation (brown triangles) deviates for larger values quite strongly. Interestingly, the local approach (orange squares) seems to be able to, while not quantitatively, qualitatively reproduce the behavior of the Langevin equation. Same parameters as in Fig. 2.2.

of any system-bath coupling. However, both the Langevin (brown triangles) and the local master equation (orange squares) do depend on the coupling γ. Thus, qualitatively, the local master equation seems to be able to describe the large γ case better. Quantitatively, however, it is not able to follow the Langevin equation behavior. It is important to note, that above the critical point λ_C, the dynamics for the coupled harmonic oscillators become unstable, as the eigenvalue ω_- becomes imaginary (2.4) and thus it is questionable whether the description of the coupled harmonic oscillator model is still valid in this regime.

2.4. DISCUSSION

To conclude, we have examined the behavior of the global master equation, the Langevin equation and the local master equation for finite intra-system coupling. In particular we considered the critical behavior of these for equilibrium and nonequilibrium situations of a coupled harmonic oscillator model with position-position coupling and its rotating-wave form. This model is simple enough to allow the derivation of the (global) quantum master equation and therefore enable one to compare the local and global approach in various aspects. In particular one is able to explicitly calculate correlation measures and negativity. Yet, this model is also sufficiently complex to express quantum critical phenomena.

In the equilibrium situation, it was shown that the global master equation and the Langevin equation completely reproduce the Gibbs distribution behavior for any finite temperature and both couplings. The local master equation on the other hand is only able to reproduce this behavior 2.2 qualitatively in the position-position coupling case, but is not able to do so even qualitatively in the rotating-wave case. This is also the case

for the creation of correlations in these systems 2.4, 2.5. Interestingly, the case of vanishing temperatures $T_i = 0$ is trivial in the rotating-wave case, as all steady-states are given by the direct product of the ground state of the two oscillators. In the position-position coupling case, on the other hand, it was shown that the local approach and the Langevin equation now coincide 2.3, but only the global approach is able to reproduce the Gibbs distribution. Considering the negativity 2.5 it was further shown, that this holds true also for nonlocal properties in the form of coherences. Thus, in the equilibrium setting the global approach seems to be better in describing the correct steady-state behavior for any temperature, while the Langevin equation has difficulties at $T = 0$. The local approach only qualitatively can reproduce the critical behavior in the position-position case, but completely fails in the rotating-wave case. The difficulty at vanishing temperatures for the Langevin equation might be considered as a sign of non-Markovian effects, rendering the Langevin equation erroneous.

In the nonequilibrium situation, the same problems occur for the local approach, in particular it cannot reproduce the critical behavior there either. It is interesting, that the global master equation and the Langevin equation coincide for finite temperatures for any nonequilibrium situations in the rotating-wave case, but in the position-position case this no longer holds true. It was further shown that the sign of the temperature difference results in nontrivial differences between the global master equation and Langevin equation 2.6,2.7, which is potentially caused by the asymmetry of the oscillator frequencies ω_i of the system. It thus was shown that, depending on the chosen parameter regimes, in particular the local approach either is able to only qualitatively describe the critical behavior or not at all.

Finally, let us briefly discuss the results for the harmonic oscillators w.r.t. to the initial cause of discussing critical behavior, the phase transition behavior of the Dicke model discussed in 2.2. At vanishing temperature, only the position-position coupling is able to reproduce the critical behavior. The rotating-wave coupling case does not express any critical behavior for any of the models. Further, finite temperatures should cause the critical point to exponentially increase with β in the equilibrium situation. The fluctuations described by the harmonic oscillators, however, will always behave critically at the same critical point λ_c, irrespective of temperature. The shift of the critical point in dependence of the system-bath coupling can be reproduced using the local master equation or the Langevin equation for the position-position coupling. But the dynamics becomes unstable in the regime above the critical point λ_c as the eigenfrequency ω_- becomes imaginary. These insufficiencies might suggest, that the description of the Dicke model fluctuation using coupled harmonic oscillators might not be ideal. If one is experimentally interested only in steady-state behavior and e.g. a limited number of observables, then one can take either of the considered approaches and take γ_i as a free parameter which can be fitted to the problem. Only in cases where the behavior is also qualitatively incorrect, like the local master equation in general, or all equations at vanishing temperature $T = 0$ for the rotating-wave interaction model.

3

PERTURBATIVE TREATMENT OF QUANTUM MASTER EQUATIONS

3.1. INTRODUCTION

In the prior chapter 2 it was shown that the global approach to quantum master equations is generally able to describe quantum steady-state behavior more accurately than the local approach. However, even though the global master equation itself is a highly approximative theory [6] of open quantum dynamics that is generally too difficult to be described in its entirety, explicitly deriving the global master equation is usually rather an involved endeavor. One major difficulty in deriving such a global master equation lies in the fact that the complete eigenstates and eigenenergies have to be determined [6]. For most Hamiltonians that are quadratic in either ladder operators $a_i, a_j^\dagger$ or canonical operators p_i, q_j, there are diagonalization techniques [91, 92], which enable one to derive a global master equation analytically from first principles. However, finding the eigensystem of a general Hamiltonian is a nontrivial task. Without the eigensystem, though, the global master equation cannot be derived. Owing to this difficulty, one might be inclined to use the local approach as discussed in the prior chapter 2.

In general, if one is not able to find a proper solution of a problem, a common practice is to use perturbation theory to receive an approximative equation that can be used without the need of using the unknown complete eigensystem. The more orders are used, the more accurate one might be able to reproduce the global master equation's results. In fact, there are paper's discussing such perturbation theories for Lindblad master equations [66, 102–104]. Especially [102] showed that the correction up to first order can fix unphysical heat currents that the local approach may cause [28]. In this peturbative context, the local approach can also be considered as a zeroth order perturbation.

In this chapter, we will consider how well the steady-state results from the first order perturbative approach are able to correct errors caused by the local approach and how well it can approximate the global one. First we will consider various aspects in the equilibrium regime. Among these, the local occupation number of the first oscillator, the

quantum mutual information and further physical aspects of heat currents and the uncertainty principle. This comparison will then be reiterated for the nonequilibrium case with a stronger focus on heat currents and entropy production. Then the dependence of these approaches on the structure of the bath is considered, by comparing a flat bath spectrum with a three-dimensional one [10]. Lastly, the second order perturbation is treated and its steady-state behavior compared with the other approaches.

These equations will be compared using the example of chapter 2 considered example of the coupled harmonic oscillator (2.2). In particular the position-position coupling will be treated ($\kappa = \lambda$) in the main part. The rotating-wave case ($\kappa = 0$) can be found in the appendix 7.4. These differences will be considered for the example of the coupled harmonic oscillator model (2.2) considered in the prior chapter 2 for primarily the position-position coupling ($\kappa = \lambda$), while the for rotating-wave ($\kappa = 0$) case can be found in the appendix 7.3.

We find that while higher order perturbative terms do improve the approximation in some aspects, the obtained master equations do not necessarily reproduce the global master equation better for all properties, even for vanishing intra-system coupling $\lambda \approx 0$. It is even possible that such higher order result in unphysical density matrices.

3.2. PERTURBATION THEORY AND THERMODYNAMIC DESCRIPTION

Before comparing the first order perturbative master equation with the local and global approaches, first we will consider how perturbation theory can be applied in the derivation of the Lindblad master equation [102]. We then use these results to receive the first order perturbative master equation for the coupled harmonic oscillator model (2.2). Then we consider how various thermodynamic functions can be defined for quantum master equations.

3.2.1. PERTURBATION THEORY FOR QUANTUM MASTER EQUATIONS

The work by Shishkov et al. [102] derives an approximate master equation by expanding the interaction picture system operators about the intra-system coupling strength $\lambda = 0$, where the system Hamiltonian can be written in the general form $H_S = H_0 + \lambda H_I$. They further apply their result to spin systems. We here extend this treatment to harmonic oscillators. We will restrict ourselves to the case that the Hamiltonian H_0 only contains non-degenerate eigenvalues, i.e. $\omega_1 \neq \omega_2$ in the case of the coupled harmonic oscillators (2.2).

The interaction picture dynamics of a system operator for the system + bath dynamics coincides with the Heisenberg picture dynamics of the system-operator if the system would be considered solely. For a general operator A_n, $\vec{A} = (a_1, a_1^\dagger, a_2, a_2^\dagger)$ of the coupled harmonic oscillator model with the Hamiltonian $H_S = H_0 + \lambda H_I$, (2.2), the dynamics are given by

$$\begin{aligned} \dot{A}_n^I(t) &= i[H_S, A_n^I(t)] = iU_S^\dagger(t)[H_S, A_n]U_S(t) = iU_S^\dagger(t)[H_0 + \lambda H_I, A_n]U_S(t) \\ &= (-1)^n i\omega^{(n)} A_n^I(t) + i\lambda[H_I^I(t), A_n^I(t)], \end{aligned} \tag{3.1}$$

with $A_n^I(t) = U_S^\dagger(t) A_n U_S(t)$. It was used here, that $[H_0, A_n] = (-1)^n \omega^{(n)} A_n$, where $\omega^{(n)}$ is either ω_1 for $n = 1,2$ or ω_2 if $n = 3,4$. The goal of the perturbation theory is, that these operators can be written as a sum of time-independent operators that are multiplied by an oscillatory term $A_n^I(t) = \sum_\omega B_\omega e^{i\omega t}$ for some frequencies ω, as is needed in deriving the master equation (see chapter 2.3). The operators can be formally expanded in orders of λ, $A_n^I(t) = \sum_m \lambda^m A_n^{I(m)}(t)$ with the initial condition $A_n^I(0) = A_n$. The zeroth order differential equation for the operator thus is given by

$$\dot{A}_n^{I(0)}(t) = (-1)^n i\omega^{(n)} A_n^{I(0)}, \; A_n^{I(0)}(0) = A_n, \tag{3.2}$$

which can be solved by $A_n^{I(0)}(t) = A_n \exp\left((-1)^n i\omega^{(n)} t\right)$. Thus, the zeroth order fulfills the form necessary to be able to derive a master equation starting from Eq. (2.10). Doing so, one would reproduce the local approach (2.7). Therefore, the local approach can be regarded as the zeroth order perturbation.

For the first order, also the interaction part $H_I^I(t)$ has to be approximated. Since the Hamiltonian term $\lambda H_I^I(t)$ is already linear, $H_I^I(t)$ has to be expanded up to zeroth order $H_I^I(t) = \sum_{m,k} \varepsilon_{m,k} A_m^{I(0)}(t) A_k^{I(0)}(t)$, where the coefficient matrix is given by

$$\varepsilon_{m,k} = \frac{1}{2} \begin{pmatrix} 0 & 0 & \frac{\kappa}{\lambda} & 1 \\ 0 & 0 & 1 & \frac{\kappa}{\lambda} \\ \frac{\kappa}{\lambda} & 1 & 0 & 0 \\ 1 & \frac{\kappa}{\lambda} & 0 & 0 \end{pmatrix}. \tag{3.3}$$

The resulting differential equation for the first order correction then follows as

$$\dot{A}_n^{I(1)}(t) = (-1)^n i\omega^{(n)} A_n^{I(1)}(t) + i \sum_{m,k} \varepsilon_{m,k} [A_m A_k, A_n] e^{i\left((-1)^m \omega^{(m)} + (-1)^k \omega^{(k)} + (-1)^n \omega^{(n)}\right) t}, \tag{3.4}$$

with the initial condition $A_n^{I(1)}(0) = 0$. This operator valued differential equation may be solved by

$$A_n^{I(1)}(t) = \sum_{m,k} \frac{\varepsilon_{m,k} [A_m A_k, A_n]}{(-1)^k \omega^{(k)} + (-1)^m \omega^{(m)}} \left(e^{i\left((-1)^m \omega^{(m)} + (-1)^k \omega^{(k)}\right) t} - 1 \right) e^{(-1)^n i\omega^{(n)} t}. \tag{3.5}$$

The exact form of these first order perturbation operators $A_n^{I(1)}$ is given in 7.3. Using (3.5), one can thus write the interaction operators up to first order in the following general form

$$\begin{aligned} A_n^I(t) = &\left(A_n + \sum_m \Lambda_m A_m \right) \exp\left((-1)^n i\omega^{(n)} t\right) \\ &+ O\left(\exp\left((-1)^m i\omega^{(m)} t + (-1)^k i\omega^{(k)} t + (-1)^n i\omega^{(n)} t \right) \right), \end{aligned} \tag{3.6}$$

where the Λ_m are determined by (3.5). It is important to note that terms that oscillate at frequency $(-1)^m \omega^{(m)} + (-1)^k \omega^{(k)} + (-1)^n \omega^{(n)}$ can be simplified by the fact that $[A_m A_k, A_n]$ is only nonvanishing if A_n is equal to $A_m^\dagger$ or $A_k^\dagger$, which results in the fact that two of the three frequencies will cancel one another and therefore the additional term will result in dissipators, up to first order, that oscillate with a frequency $\omega_1 \pm \omega_2$. Because the considered perturbation theory is valid only for nonresonant situations $\omega_1 \neq \omega_2$,

these oscillatory dissipators will vanish under the secular approximation (see discussion below Eq. (2.17)). Since the first order correction in (3.6) oscillate with the same frequency as the zeroth order, the dissipators can be obtained by replacing the zeroth order operators in the local master equation (2.7) by the operators up to first order given in (3.6). Following the notation of the global approach (2.19),(2.20), the system-bath coefficients, caused by the interaction with the two baths then have the general form $\Gamma(A_n, A_m) = \Gamma_1(A_n, A_m) + \Gamma_2(A_n, A_m)$, with

$$\begin{aligned}
&\Gamma_1(a_1, a_1^\dagger) = \gamma_1(\omega_1)(N(\beta_1,\omega_1)+1),\ \Gamma_1(a_1^\dagger, a_1) = \gamma_1(\omega_1)N(\beta_1,\omega_1)\\
&\Gamma_1(a_1, a_2^\dagger) = \frac{\gamma_1(\omega_1)\lambda}{\omega_1-\omega_2}(N(\beta_1,\omega_1)+1),\ \Gamma_1(a_1, a_2) = \frac{\gamma_1(\omega_1)\kappa}{\omega_1+\omega_2}(N(\beta_1,\omega_1)+1)\\
&\Gamma_1(a_2, a_1^\dagger) = \frac{\gamma_1(\omega_1)\lambda}{\omega_1-\omega_2}(N(\beta_1,\omega_1)+1),\ \Gamma_1(a_2^\dagger, a_1^\dagger) = \frac{\gamma_1(\omega_1)\kappa}{\omega_1+\omega_2}(N(\beta_1,\omega_1)+1)\\
&\Gamma_1(a_1^\dagger, a_2) = \frac{\gamma_1(\omega_1)\lambda}{\omega_1-\omega_2}N(\beta_1,\omega_1),\ \Gamma_1(a_1^\dagger, a_2^\dagger) = \frac{\gamma_1(\omega_1)\kappa}{\omega_1+\omega_2}N(\beta_1,\omega_1)\\
&\Gamma_1(a_2^\dagger, a_1) = \frac{\gamma_1(\omega_1)\lambda}{\omega_1-\omega_2}N(\beta_1,\omega_1),\ \Gamma_1(a_2, a_1) = \frac{\gamma_1(\omega_1)\kappa}{\omega_1+\omega_2}N(\beta_1,\omega_1).
\end{aligned} \tag{3.7}$$

The second bath has the same kind of contributions and can be obtained by interchanging all subscripts ($1 \leftrightarrow 2$) in Eq. (3.7). Thus, one finds that the first order perturbative master equation does not contain local squeezing dissipators of the form $\Gamma(a_i, a_i) = 0$ et cetera, for the position-position coupling case, compared to the global master equation dissipators (2.20). This is because the used perturbation theory is done only up to first order, while the local squeezing is a higher order effect, as (2.2) itself does not contain local squeezing terms. It is further interesting to see that the first order perturbative dissipators (3.7) have thermal contributions $N(\beta_i,\omega_i)$ that are evaluated at the unperturbed frequencies ω_i, and thus are unmodified by the coupling up to first order. While there are no local squeezing dissipators $\Gamma(a_i, a_i)$ etc, in the position-position coupling case ($\lambda = \kappa$), there exist nonlocal squeezing dissipators of the form $\Gamma(a_i, a_j) \neq 0,\ i \neq j$ etc.

3.2.2. THERMODYNAMIC FUNCTIONS FOR QUANTUM MASTER EQUATIONS

Heat currents for quantum master equations, in the case of stationary Hamiltonians, can be defined [6, 28, 69] by considering the energy flux

$$\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t}\langle H_S\rangle &= \mathrm{Tr}\{H_S\dot{\rho}(t)\} = -i\mathrm{Tr}\{H_S[H_S,\rho]\} + \mathrm{Tr}\{H_S\left(\mathscr{D}_1(\rho)+\mathscr{D}_2(\rho)\right)\}\\
&= \langle\overline{\mathscr{D}}_1(H_S)+\overline{\mathscr{D}}_2(H_S)\rangle = \mathfrak{J}_1+\mathfrak{J}_2,
\end{aligned} \tag{3.8}$$

where $\overline{\mathscr{D}}_i(H_S)$ stand for the adjoint dissipators of the respective bath i. These adjoint dissipators can create a formal Heisenberg picture for the considered operators using the master equation (more details will be provided in the discussion for (4.2)). The heat currents are then defined as $\mathfrak{J}_i = \langle\overline{\mathscr{D}}_i(H_S)\rangle$. For the example of a single harmonic oscillator (2.6) with frequency ω, it can be easily shown that for a thermal initial state with temperature β_1, and bath temperature β_2, the heat current has the form $\mathfrak{J} = \gamma\omega[(N(\beta_2,\omega)+1)N(\beta_1,\omega) - (N(\beta_1,\omega)+1)N(\beta_2,\omega)]$. If the bath has a higher temperature than the system

$\beta_2 < \beta_1$, then $N(\beta_2,\omega) > N(\beta_1,\omega)$ and thus $\mathfrak{J} > 0$. Therefore, a positive heat current may be interpreted as energy (heat) flowing into the system.

For the stationary state of the dissipative dynamics $\dot{\rho} = 0$, it can be seen using (3.8), that these heat currents have to compensate each other $\mathfrak{J}_1 + \mathfrak{J}_2 = 0$. If these heat currents do not vanish individually, then the stationary state is in a nonequilibrium steady-state. Thus, Eq. (3.8) in this steady-state case can be considered as a steady-state version of the first law of thermodynamics.

A steady-state version of the second law can be given by considering the model of a system coupled to two thermal baths [28]. In the steady-state, it is physically expected that heat will flow from the hotter bath T_h to the colder one T_c. If a positive heat current corresponds to heat flowing into the system, then from (3.8) it follows that

$$\frac{\mathfrak{J}_h}{T_h} + \frac{\mathfrak{J}_c}{T_c} =: -\sigma \leq 0. \tag{3.9}$$

This defines a steady-state entropy production σ.

In the equilibrium case, thermodynamic consistency demands individually vanishing heat currents $\mathfrak{J}_i = 0$ and vanishing entropy production $\sigma = 0$.

3.3. INVESTIGATION OF THE PERTURBATIVE APPROACH

We will now consider how well the first order perturbative approach reproduces the results of the global master equation, compared to the local one. First we will treat the equilibrium case, which is then followed by the nonequilibrium one. Next, the bath structure dependence will be investigated. Lastly the second order perturbation will be briefly treated by expanding the global approach. We will focus on the position-position coupling ($\kappa = \lambda$) in the main part of this chapter. The rotating-wave Hamiltonian ($\kappa = 0$) investigation can be found in the appendix 7.4.

3.3.1. EQUILIBRIUM CASE

First, let us consider how the first order perturbative approach can describe the equilibrium situation. For the position-position coupling Hamiltonian (2.2), with $\kappa = \lambda$, the global approach was able to completely reproduce the Gibbs state behavior (see discussion in chapter 2). In Fig. 3.1, the global approach (blue circles), Gibbs state (green line), local master equation (orange squares) and the first order perturbative master equation (brown triangles) are plotted for the local occupation number $\langle a_1^\dagger a_1 \rangle$ on the left and the quantum mutual information I on the right (see definition in chapter 7.1). The main figures are evaluated at the, compared to the system frequencies, high temperature $T_i = 98$, while the insets are for the vanishing temperature case $T_i = 0$. The mean occupation number of the first order perturbative approach in Fig. 3.1 shows a similar difficulty to reproduce the global approach as the local one, with the differences that the first order perturbative approach results in a weaker mean occupation number as opposed to the local approach that is too large. This also holds true for the $T_i = 0$ case. Remarkably, even though the local occupation number of the first order perturbative approach is not able to reproduce the global behavior, the quantum mutual information (right Fig. in 3.1) is quite well reproduced for finite temperature and the considered parameter regime λ/λ_C. Even the $T_i = 0$ case is still quite well replicated for smaller λ/λ_C.

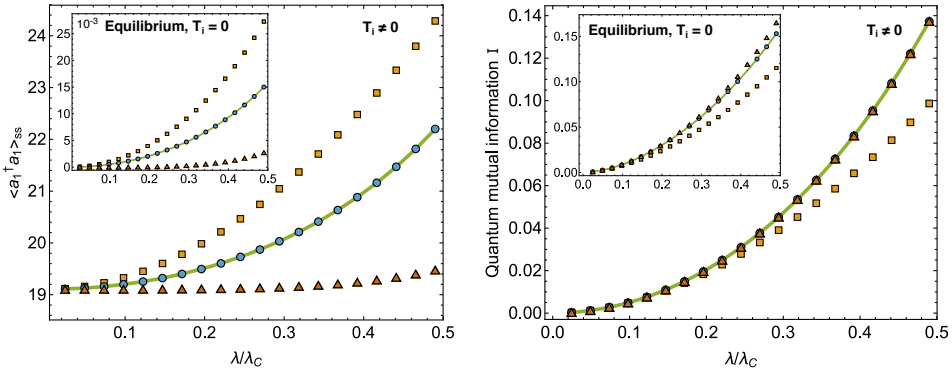

Figure 3.1: Comparison of the global, local and first order perturbative master equation for the local occupation number $\langle a_1^\dagger a_1\rangle$ (left) and the mutual information (right) of the position-position Hamiltonian (2.2) in dependence of the intra-system coupling strength λ/λ_c, for the cases of finite (equilibrium) temperature $T_i = 98$ and vanishing temperature $T_i = 0$ (insets). The global ME solution (blue circles) is shown with the reference Gibbs solution (green line). (left): The first order perturbative approach (brown triangles) has a similar difficulty reproducing the global result as the local approach (orange squares). This holds true for both finite and for vanishing temperature (inset). (right): Interestingly, the mutual information can be reproduced quite well by the first order perturbative approach in the considered λ/λ_c regime. Only for vanishing temperatures it has difficulties for higher λ/λ_c. Parameters are $\gamma_1 = \gamma_2 = 1.5\cdot 10^{-4}$, $\omega_1 = 5, \omega_2 = 2$.

A possible reason for this finding may be seen by considering the dissipators of the first order perturbation (3.7). Compared to the local dissipators (2.7), there are many additional nonlocal dissipators in the first order perturbative case. In particular also nonlocal squeezing terms $\Gamma(a_1, a_2)$, etc. Thus, the first order perturbative approach is able to better reproduce the nonlocal system-correlations. However, compared to the global approach (2.20), still lacks both local dissipators of the form $\Gamma(a_i, a_i)$ as well as any λ dependence of the dissipators $\Gamma(a_i^\dagger, a_i)$, which are of higher order in the perturbation.

Let us now dive deeper into the question whether or not the different approaches are *physical* in the equilibrium context. Figure 3.2 considers on the left how the heat current of the first bath $\mathfrak{J}_1 = \langle\overline{\mathscr{D}}_1(H_S)\rangle$ looks like using the different dissipators (How the heat currents are calculated can be found in the appendix 7.2). Since we consider the equilibrium case, it should be physically expected that the individual heat currents vanish $\mathfrak{J}_i = 0$. However, as can be seen in Fig. 3.2, only the global approach behaves as expected for finite and vanishing temperature. Both the local and first order perturbative approaches result in unphysical heat currents, regardless of temperature. The two approaches once again express a sort of inverse behavior about the correct global solution. This means, that the dissipators (2.7),(3.7) are not necessarily trustworthy in their ability to determine thermodynamic properties.

Another interesting question is, whether or not the approximative approaches can even create *physical* density operators. The uncertainty principle for covariance matrices can be stated [101, 105, 106], using the symplectic matrix $\overline{\gamma}$. A covariance matrix σ fulfills the uncertainty principle, if the matrix $\sigma - i/2\overline{\gamma}$ is positive semi-definite [101, 105, 106],

$$\sigma - i/2\overline{\gamma} \geq 0, \tag{3.10}$$

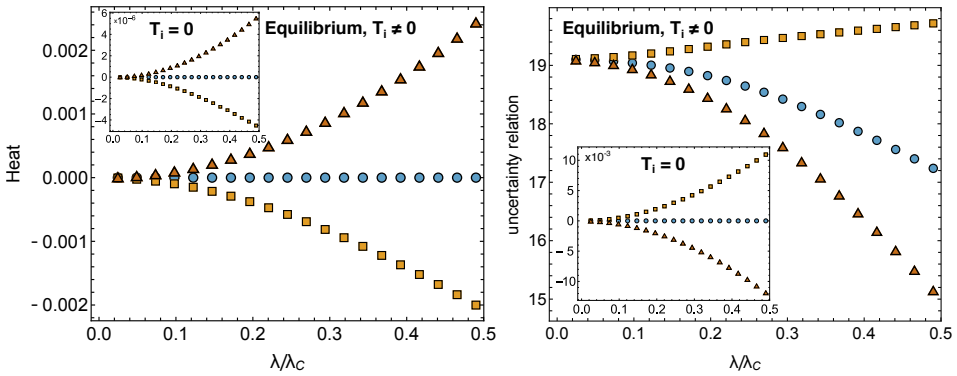

Figure 3.2: Comparison of the global, local and first order perturbative master equation heat current from the first bath $\mathfrak{J}_1$ (left) and the check whether the uncertainty principle is fulfilled (right) in dependence of the intra-system coupling λ/λ_c of the position-position Hamiltonian (2.2), for the cases of finite (equilibrium) temperature $T_i = 98$ and vanishing temperature $T_i = 0$ (insets). The global approach (blue circles) reproduces the expected behavior of no net heat current from the baths (left) as the equilibrium case is considered here. However, both the heat currents for the first order perturbative (brown triangles) and local approach (orange squares) are unphysical. The same behavior can be seen for the $T_i = 0$ case (inset). The uncertainty relation (right) is plotted for all three cases in the form of the lowest eigenvalue of the symplectic matrix $\sigma - i\widetilde{\gamma}$. As long as all values are semi-positive, the corresponding covariance matrix σ will fulfill the uncertainty relation. For the finite temperature $T_i = 98$ all three approaches therefore fulfill the uncertainty relation. For lower temperature, and in particular $T_i = 0$ (inset), the first order perturbative approach fails to fulfill the uncertainty relation, while the other two approaches always have physical covariance matrices. Parameters are the same as in Fig. 3.1.

i.e. all of its eigenvalues have to be ≥ 0. The lowest eigenvalue of this matrix (3.10) is plotted on the right in Fig. 3.2. For finite temperature $T_i = 98$, all three equations result in physical covariance matrices. In the inset, however, for T_0 and in fact also for general low temperatures, it is the case that the first order perturbative approach creates covariance matrices, and thus states, that do not fulfill the uncertainty relation.

It is interesting to note, that the local approach fulfills the uncertainty relation (3.10) for any temperature. The first order perturbation on the other hand might result in highly unphysical states, that only partly reproduce the global approach better in equilibrium situations. A possible reason for this lies in the fact that to linear order only the nonlocal dissipators of the form $\gamma(a_1, a_2)$ etc are modified by the intra-system coupling λ. While these enable the first order perturbative approach to reproduce the correlation properties of the density operator in the form of the quantum mutual information in Fig. 3.1, the incorrect local description lead to an inconsistent covariance matrix.

3.3.2. NONEQUILIBRIUM CASE

Next we consider how the nonequilibrium cases are approximated by the first order perturbative approach. Since the above consideration showed that especially at low temperatures the local/ first order perturbative approaches have the most difficulties following the global approach, we limit the nonequilibrium discussion to the high-temperature case. We set the temperature of bath 1 fixed at $T_1 = 98$.

The thermodynamic properties of a bosonic oscillator mode is described by the Bose-

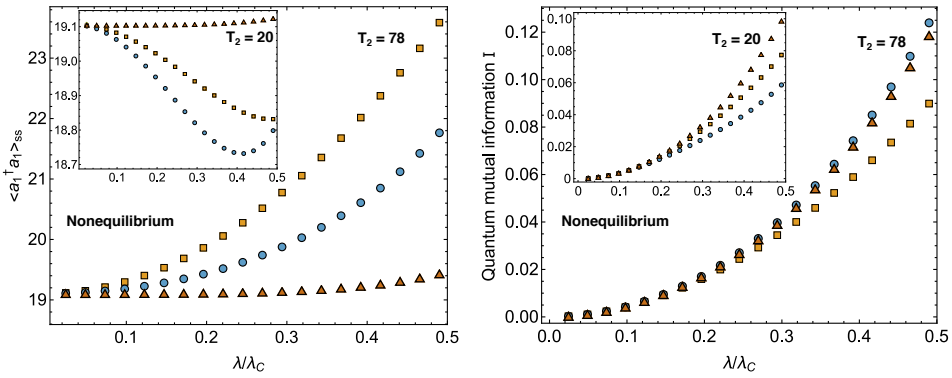

Figure 3.3: Comparison of the global, local and first order perturbative master equation for the local occupation number $\langle a_1^\dagger a_1\rangle$ (left) and the mutual information (right) in dependence of the intra-system coupling λ/λ_c of the position-position Hamiltonian (2.2), for the cases of nonequilibrium temperatures $T_1 = 98$ in the anomalous regime $T_2 = 78$ as well as in the normal regime $T_2 = 20$ (insets). The anomalous regime is close to equilibrium and thus its behavior is similar to the equilibrium case in Fig. 3.1. The normal regime shows (insets) that the mean occupation number is better described by the local approach. Even the quantum mutual information (right), while overall worse than the equilibrium case, is now better described by the local approach. Parameters are the same as in Fig. 3.1.

Einstein distribution $N(\beta_i,\omega_i)$, which is determined by the product $\beta_i\omega_i$. For non resonant oscillators $\omega_1 \neq \omega_2$, there exist two interesting nonequilibrium regimes. If $\beta_1 < \beta_2$ one normally expects $\beta_1\omega_1 < \beta_2\omega_2$, i.e. a higher temperature results in a higher occupation N; in the following this regime will be called the normal regime. However, for a steeper oscillator $\omega_1 > \omega_2$ which is coupled to the hotter bath, there exists also a regime where $\beta_1\omega_1 - \beta_2\omega_2 > 0$, i.e. even though the oscillator 1 is coupled to the hotter bath, the occupation number $N(\beta_1,\omega_1)$ is still smaller than the corresponding $N(\beta_2,\omega_2) > N(\beta_1,\omega_1)$ for the second bath; this regime will be called anomalous regime in the following. These two regimes seem relatively subtle, but they have interesting implications on the validity of the master equation approaches.

In Fig. 3.3 the local occupation number $\langle a_1^\dagger a_1\rangle$ (left) and mutual information (right) are plotted for the temperature case $T_2 = 78$ in the anomalous regime and $T_2 = 20$ (inset) in the normal regime. In the normal regime $T = 20$, it is visible that for both the local occupation number and the mutual information the local approach is closer to the global one than the first order perturbative approach. In the anomalous regime, however, the first order perturbative approach is better able to reproduce the mutual information, while the local occupation number is similar as in the equilibrium case Fig. 3.1. The anomalous regime is also rather close to equilibrium $T_2 \approx T_1$, thus such a behavior is somewhat expected since this is the same behavior as in the equilibrium case in Fig. 3.1. It is interesting that stronger nonequilibrium situations (in the normal regime) are better described by the local approach than the first order perturbative one.

The anomalous regime is also the one, that Levy and Kosloff [10] used to show that the local approach can violate the second law of thermodynamics for the rotating-wave Hamiltonian (2.2) ($\kappa = 0$). Further, Shishkov et al [102] showed that the first order perturbative master equation may correct the unphysical heat current. Thus, it is very inter-

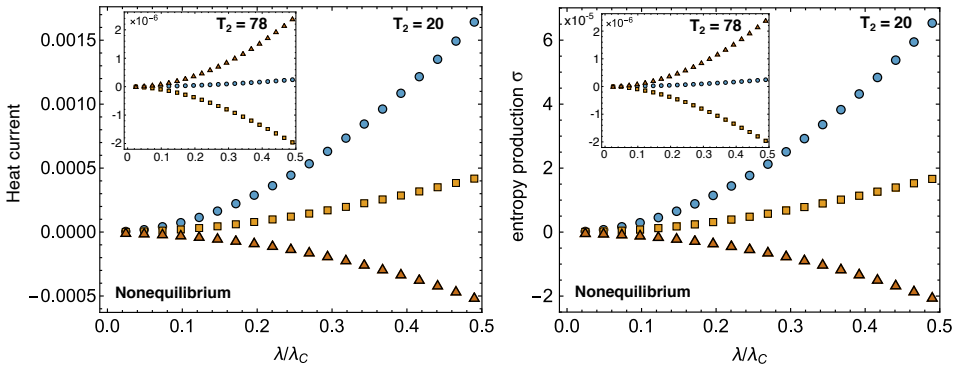

Figure 3.4: Comparison of the global, local and first order perturbative master equation heat current from the first bath $\mathfrak{J}_1$ (left) and entropy production σ in the steady-state (right) in dependence of the intra-system coupling λ/λ_C of the position-position Hamiltonian (2.2), for the cases of nonequilibrium temperatures $T_1 = 98$ in the normal regime $T_2 = 20$ as well as in the anomalous regime $T_2 = 78$ (insets). The heat current from the first bath $\mathfrak{J}_1$ (left) is positive for the local approach and negative for the first order perturbative one in the normal regime. In the anomalous regime (inset), now the heat current of the first order perturbative approach is positive, while for the local one negative. The entropy production σ on the right resembles this behavior accordingly. Parameters are the same as in Fig. 3.1.

esting to consider how the thermodynamic properties of the master equation approximations for the nonequilibrium case behave and whether the first order perturbation corrects in general such thermodynamic properties as the heat currents.

In Fig. 3.4 the heat current $\mathfrak{J}_1$ (left) and the entropy production σ (right), defined in (3.9), are shown for the normal and the anomalous regime (inset). First considering the heat currents (left), one can see that while the global one is always positive, in the normal regime only the local approach is also positive. The first order perturbative approach is actually negative and thus unphysical. However, in the anomalous regime, this is switched; the first order perturbative approach is positive, while the local one is negative. From these sign differences of the heat currents, it also follows that, depending on the regime, either the local or first order perturbative approach will have a physical (positive) entropy production σ in the steady-state. This means that the first order perturbative approach, which is a higher order perturbation as the local approach, can fix an unphysical heat current in certain situations. This is, however, not necessarily true in general even for infinitesimally small λ. Rather, it has an apparent inverse behavior compared to the local approach.

3.3.3. STEADY-STATE BEHAVIOR IN DEPENDENCE OF THE BATH STRUCTURE

We will briefly consider how the first order perturbative approach reacts to a different bath-structure, altering the system-bath couplings. For three-dimensional baths, the density of states is proportional to $J(\omega) \propto \omega^3$, where ω is the corresponding system eigenfrequency in the Markov approximation. This results in system-bath couplings of the form [10] $\gamma_i(\omega) = \gamma_i \omega^3$. For the local (2.7) and first order perturbative dissipators (3.7) these ω are just given by the respective harmonic oscillator frequencies $\gamma_i(\omega_i)$. The global approach (2.20), on the other hand has $\gamma_i \to \gamma_i(\omega_\pm)$ for the respective eigen-

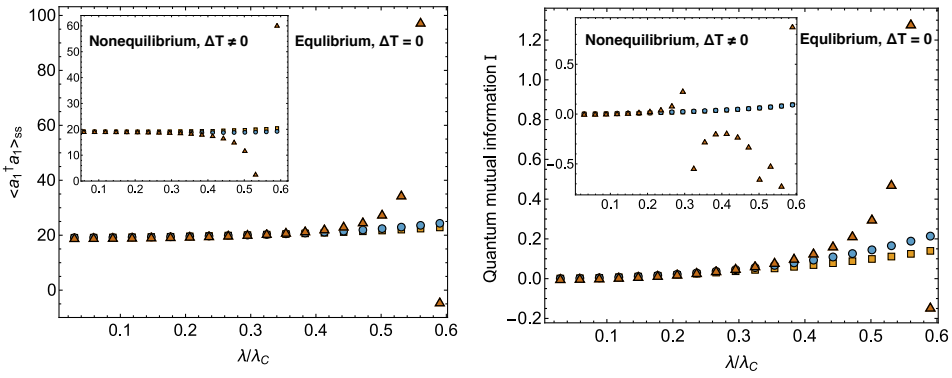

Figure 3.5: Comparison of the global, local and first order perturbative master equation local occupation number $\langle a_1^\dagger a_1 \rangle$ (left) and the quantum mutual information (right) in the steady-state of the position-position Hamiltonian (2.2) in dependence of the intra-system coupling λ/λ_c, for the equilibrium case $T_i = 98$ as well as a nonequilibrium case $T_1 = 98$, $T_2 = 20$ (inset) for three-dimensional baths $\gamma_i(\omega) = \gamma_i \omega^3$. The first order perturbative approach (brown triangles) shows signs of divergence at a consistent point in the equilibrium case for both the local occupation number (left) and the quantum mutual information (right), while the local (orange squares) and global approach (blue circles) do not. In the nonequilibrium case (insets) the first order perturbative approach behaves even more suspect, as the quantum mutual information becomes unphysical at an earlier point, compared to the mean occupation number. Parameters are the same as in Fig. 3.1, with $\gamma_i = 1.5 \cdot 10^{-4} \cdot \omega^3$, where ω is either $\omega = \omega_i$ for the i-th bath for the local or first order perturbative case and $\omega = \omega_\pm$ for the global case (2.20).

frequencies $\omega_\pm$. These modifications introduce unequally strong couplings to the two baths, $\gamma_1(\omega_1) \neq \gamma_2(\omega_2)$.

Figure 3.5 shows the local occupation number (left) and quantum mutual information (right) for this 3D-baths case, for an equilibrium and nonequilibrium constellation (insets). First considering the equilibrium case, it can be seen in Fig. 3.5 , that the first order perturbative approach shows a diverging behavior approaching a certain value $\lambda/\lambda_C \approx 0.56$ for both the local occupation number and mutual information, whereas this is not the case for the global or local approaches. Further, in the nonequilibrium case (insets) this is even worse; the first order perturbative approach shows an unphysical (negative) divergence at $\lambda/\lambda_C \approx 0.54$, while the quantum mutual information shows an earlier divergence at $\lambda/\lambda_C \approx 0.3$. One possible reason for this behavior might be the following; As mentioned above, the three dimensional baths introduce an asymmetry between the bath couplings. Because of that, one no longer can treat the bath as one bath that is coupled to the system. This higher complexity apparently cannot be correctly captured by the first order perturbative approach.

3.3.4. SECOND ORDER PERTURBATION OF THE GLOBAL MASTER EQUATION

The prior findings lead to the conclusion that the perturbation theory of the quantum master equation cannot be interpreted as just the first order approximation of a function, that describes it correctly at the point of expansion $\lambda = 0$ and in close vicinity, while becoming more inaccurate for larger λ. Rather, different aspects of the global master equations' steady-state behavior can be partly described by different orders. It is some-

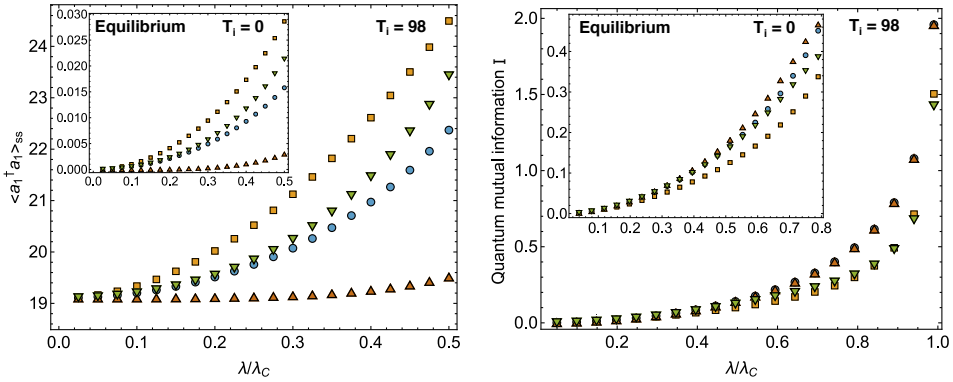

Figure 3.6: Comparison of the global, local, first order perturbative and second order perturbative master equation for the local occupation number $\langle a_1^\dagger a_1\rangle$ (left) and the mutual information (right) in dependence of the intra-system coupling λ/λ_c of the position-position Hamiltonian (2.2), for the cases of finite (equilibrium) temperature $T_i = 98$ and vanishing temperature $T_i = 0$ (insets). (left): Compared to the local (orange squares) or the first order perturbative one (brown triangles), the second order perturbative approach (green inverted triangles) is able to describe the global one (blue circles) better w.r.t. the local occupation number for both finite temperatures and vanishing ones (inset). (right): The mutual information is similar. However, for larger couplings λ/λ_c, the first order perturbative approach is still better in describing the mutual information, irrespective of temperature. Parameters are $\gamma_1 = \gamma_2 = 1.5\cdot 10^{-4}$, $\omega_1 = 5, \omega_2 = 2$.

what expected that the first order perturbative approach is not able to reproduce the global master equations' behavior, as (3.7) lacks the squeezing dissipators of the global case (2.20). In this subsection we want to consider further how well the perturbation theory can reproduce the global master equation by considering the perturbation theory up to second order. While in principle it is possible to derive this second order in the same way as for the first order (see chapter 3.2.1), we here are solely interested in how well higher order terms are able to reproduce the global dynamics. Thus, as the complete solution is known, the global master equation in our case, the second order perturbation can be directly obtained by starting with the global dissipators (2.20) and expanding each of them up to second order

$$\Gamma(A_n, A_m) = \Gamma(A_n, A_m)|_{\lambda=0} + \lambda \frac{\mathrm{d}\Gamma(A_n, A_m)}{\mathrm{d}\lambda}\bigg|_{\lambda=0} + \frac{\lambda^2}{2}\frac{\mathrm{d}^2\Gamma(A_n, A_m)}{\mathrm{d}\lambda^2}\bigg|_{\lambda=0}. \tag{3.11}$$

While the complex form of the global master equation dissipators (2.20) results in second order perturbative dissipators that are too large to be printed here, applying this expansion, one will find that the dissipators for the second order perturbation, compared to the first order (3.7), will all be modified. In particular, now there also are also local squeezing terms $\Gamma(a_i, a_i) \neq 0$ etc. Let us consider how these changes influence the accuracy of the steady-state result. In Fig. 3.6, the local mean occupation number of the first oscillator (left) and the quantum mutual information (right) are plotted, for the equilibrium cases of finite temperature $T_i = 98$ and vanishing temperature $T_i = 0$ (inset). It is interesting to see, that the second order perturbation (inverted green triangles) now can describe the global behavior (blue circles) better than the local approach (orange squares) or first order perturbative approach (brown triangles), both for finite and van-

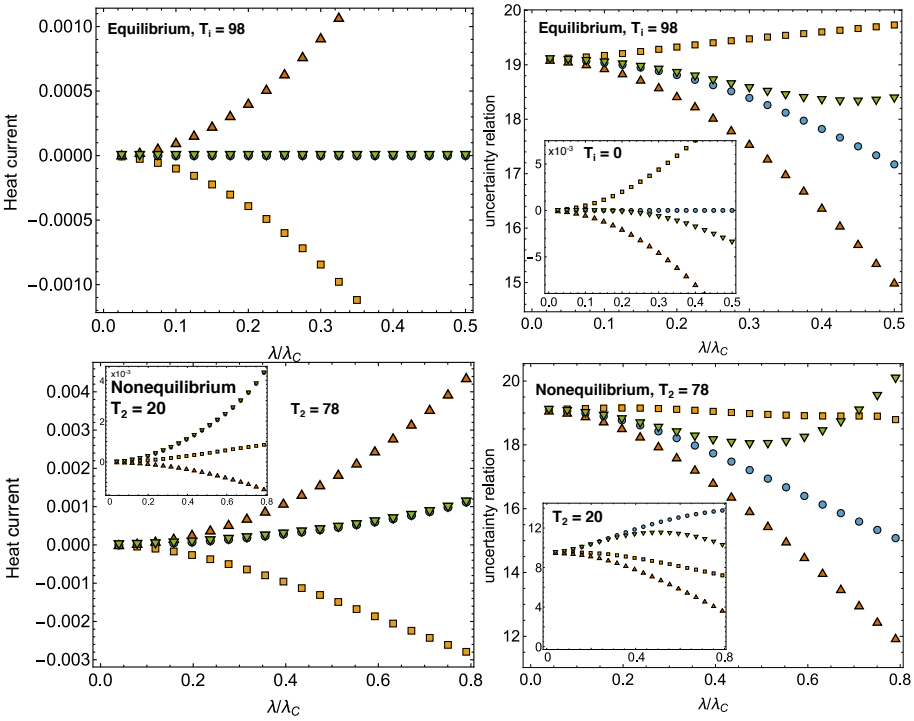

Figure 3.7: Comparison of the global, local, first order perturbative and second-order perturbative master equation heat current from the first bath $\mathfrak{J}_1$ (left) and the check whether the uncertainty principle is fulfilled (right) in dependence of λ/λ_c of the position-position Hamiltonian (2.2). The top graphs consider the equilibrium situation $T_i = 98$. The second order perturbation (inverse green triangles) is now able to correctly have a vanishing heat current $\mathfrak{J}_1$ as the global approach (blue circles). The uncertainty relation behavior of the second order perturbation is also similar to the global one for small λ/λ_c. In the $T_i = 0$ case (inset), the second order perturbation becomes for larger λ/λ_c unphysical like the first order perturbative approach (brown triangles), while the global and the local one (orange squares) stay physical. The lower graphs consider the nonequilibrium situation $T_1 = 98$ in the anomalous regime $T_2 = 78$ and the normal regime $T_2 = 20$ (insets). Interestingly, the second order perturbation manages to reproduce the global approach's heat current in both regimes. The uncertainty still is only similar for smaller λ/λ_c. Parameters are the same as in Fig. 3.6.

ishing temperature (left inset). Further, the mutual information is also improved significantly at lower coupling values, but still becomes worse than the first order perturbative approach for larger λ/λ_c.

Let us now consider the physical properties in the form of heat currents (left) and the check of the uncertainty principle (right), which are plotted in Fig. 3.7, for the equilibrium cases (upper graphs) at $T_i = 98$ and the vanishing temperature case for the uncertainty principle (inset), as well as the nonequilibrium case (lower graphs) in the normal and anomalous regime (insets). First concerning the equilibrium case, the second order perturbation is now able to suppress any unphysical heat currents, which the first order perturbative or local approaches produced. The created steady-states for the second order perturbation are further also able to follow the uncertainty of the global approach for small coupling values and finite temperatures. While it is able to now stay physi-

cal for small λ in the vanishing temperature case, it still becomes unphysical for larger λ. Interestingly, for nonequilibrium situations (lower graphs in Fig. 3.7), the second order perturbation can now reproduce the heat currents in both regimes correctly. For small coupling strengths it is also able to reproduce the same uncertainty behavior as the global approach. Only for larger coupling strengths it deviates.

Thus, the higher the order, the more properties of the global master equation can possibly be reproduced as well. While one might expect this the higher the order becomes, it is still interesting that the perturbation theory does not necessarily monotonically improve all the steady-state properties for small expansion orders about λ.

3.4. DISCUSSION

To conclude this chapter, it was mainly considered how the first order perturbation of the master equation approach reproduces the global approach compared to the local one. The result is mixed. While there are some parts, like quantum mutual information in the finite temperature equilibrium case that is being quite well reproduced by the first order perturbative approach (3.1), local observables like the mean occupation number $\langle a_1^\dagger a_1 \rangle$ behave differently than the local approach, but are not necessarily better in reproducing the global results. Further, thermodynamic properties like heat currents defined by the dissipators result in unphysical finite currents in the equilibrium case (3.2) for both the local and first order perturbative approach. Further, especially the $T_i = 0$ case is less accurately treated by the first order perturbative approach, as its steady-state fails to fulfill the uncertainty relation. For nonequilibrium situations $\Delta T \neq 0$, the first order perturbative equation can even become worse in reproducing the global behavior for large nonequilibriums 3.3. Interestingly, heat currents qualitatively seem to be behaving in an anti correlated way compared to the local approach 3.4. In regions where the local approach results in unphysical heat currents, the first order perturbative approach reproduces physical ones and vice versa. Therefore, the first order perturbative approach can be better or worse in reproducing the global behavior, but if the local approach fails, one can try the first order perturbative approach which may be able to describe the system better. This is, however, not necessarily the case.

Lastly, also the second order of the perturbation theory was considered, by expanding the global solution up to second order about $\lambda = 0$. It was shown that this order now does improve all aspects of the description, e.g. the heat current is now also physical. For low λ the behavior of the perturbation theory up to second order resembles the global approach. Thus, in the case of a perturbative approach of quantum master equations that has to produce a completely physical steady-state covariance matrix, one might have to use higher orders even for infinitesimal λ.

4

QUANTUM RESPONSE THEORY FOR NONEQUILIBRIUM STEADY-STATES

4.1. INTRODUCTION

THE prior chapters considered how well perturbative approaches to open quantum master equations can reproduce physical (steady-state) properties of systems. In the following chapters we will consider more general (dynamical) properties of quantum Markovian semigroups. In particular, we will consider in this chapter linear response theory. Response theory is a cornerstone of statistical physics. For equilibrium systems, the fluctuation-dissipation theorem connects the response to a weak external perturbation to the unperturbed correlation function between spontaneous fluctuations [108–110]. It offers a powerful tool to analyze general transport properties in numerous areas, from hydrodynamics to many-body and condensed-matter physics [25, 38, 111–113]. The fluctuation-dissipation relation has been derived for classical and closed quantum systems [25, 38, 111–113]. It is known to break down for nonequilibrium systems when detailed balance is not obeyed [114].

Over the past years, the fluctuation-dissipation theorem has been successfully generalized to classical systems in nonequilibrium steady-states, thus allowing the extension of response theory to this important class of nonthermal systems. Different theoretical formulations have been put forward [115–120], based on the Fokker-Planck equation [115], the overdamped Langevin dynamics [116–118], the Hatano-Sasa fluctuation theorem [119], or the dynamical activity [120]. Some of these modified fluctuation-response relations have been verified experimentally using colloidal particles in a toroidal optical trap [121–124].

Recently, Seifert and Speck have introduced a classification of steady-state fluctuation-dissipation theorems in the framework of stochastic thermodynamics, thus rationalizing

Parts of this chapter have been published in [107].

previous methods that lead to apparently different results [37] (see also Ref. [125]). Using a classical master equation approach, they have identified three main equivalence classes: the first variant contains a correlation function that involves no time derivatives (only functions of the steady-state distribution), the second variant is the unique form expressed in terms of time derivatives (of the stochastic entropy), whereas the last variant is the only one not requiring the explicit knowledge of the steady-state distribution. Infinitely many alternatives may be constructed via normalized linear combinations of the latter. All these variants yield the same response and are thus equivalent. However, the existence of different types of fluctuation-dissipation relations offers significant theoretical and experimental advantages. Theoretically, one kind of fluctuation-response theorem is usually easier to compute than the others, depending on the concrete application. At the same time, the choice of the form crucially affects the accuracy of the experimental determination of the nonequilibrium response function, as shown in Ref. [123]. Few attempts to extend steady-state fluctuation-dissipation theorems to open quantum systems have been presented [39–42]. However, a complete and unified picture is currently missing.

We will start by defining various different fundamental properties of quantum Markovian semigroups, how quantum detailed balance [115] may be defined for such open quantum systems and discuss briefly response theory for closed, thermal dynamics, as derived by Kubo [38, 110]. We will then develop a general framework for the steady-state linear response of open quantum systems, extending this Kubo theory, which is limited to isolated equilibrium systems with unitary dynamics [25, 38, 110–113]. We specifically derive the three equivalence classes for generalized steady-state quantum fluctuation-dissipation relations, analogously to the classical findings [37], using quantum Markovian semigroups and introduce a nonequilibrium extension of the Kubo transformation [25, 38, 110–113]. We discuss the role of the noncommutativity of quantum operators and emphasize differences to the equilibrium Kubo response theory, in particular the violation of detailed balance. We further treat how quantum detailed balance in the open context influences the general properties of these response functions. We then compare these classes with results from various authors in the quantum and classical context and show their equality.

To illustrate our unifying formalism, we consider two different examples of coupled harmonic oscillators. First we treat the rotating-wave Hamiltonian (2.2) ($\kappa = 0$) where the interaction of the two oscillators with their respective baths is described by the local master equation (2.7). This has the advantage, that the response can be calculated analytically. We use this model to stress the difference between classical and quantum responses, as well as between equilibrium and steady-state responses of Hamiltonian perturbation. Remarkably, we show that the equilibrium response function vanishes for perturbations that commute with the unperturbed Hamiltonian, whereas the steady-state response does not. This underlines the profound disparity between equilibrium and nonequilibrium quantum response theories. Lastly, we consider a second model which is given by the position-position coupling Hamiltonian (2.2) ($\kappa = \lambda$), whose open dynamic will be described by the global master equation (2.20). We show for this model how the linear response breaks down approaching a critical point and consider which differences there are for equilibrium and nonequilibrium regimes.

4.2. MATHEMATICAL CHARACTERIZATION OF QUANTUM MASTER EQUATIONS

In the following we want to consider more closely general physical properties of quantum Markovian systems. For being able to do so, it is advantageous to consider their underlying mathematical structure more closely. In particular it is interesting to consider which subtle differences in mathematical properties emerge, compared to the closed quantum dynamical case.

4.2.1. MARKOVIAN SYSTEMS AS MAPS OF POSITIVE SEMIGROUPS AND THE QUANTUM REGRESSION THEOREM

Given the full Hilbert space $\mathscr{H} = \mathscr{H}_S \otimes \mathscr{H}_B$ as a direct product of the two Hilbert spaces of the system $\mathscr{H}_S$ and the bath $\mathscr{H}_B$. The set of all bounded linear operators A acting on elements of a Hilbert space and their domains $D(A)$ create a Banach space $\mathscr{B}(\mathscr{H})$ [20, 126] with the Hilbert-Schmidt norm $||A||_\infty = \max_i \lambda_i$, where λ_i are the eigenvalues of the operator A. A special class of operators are density operators [6, 20] $\rho \in \mathscr{T}(\mathscr{H})$, which represent the possibly mixed states that a system can be found in. These constitute another Banach space $\mathscr{T}(\mathscr{H})$ with the trace norm $||\rho||_1 = \mathrm{Tr}|\{\rho\} = 1$ [20]. For the complete system + bath, the time-evolution of a density operator $\rho(t)$ and a time independent Hamilton operator H is given by the von Neumann equation $\dot{\rho}(t) = -i[H, \rho(t)] = \mathscr{L}_C(\rho(t)) = \mathscr{L}_C\rho(t)$, where the superoperator $\mathscr{L}_C$ is the generator of the closed dynamics and $\mathscr{L}_C\rho(t)$ without brackes is defined in the following as the superoperator acting on all operators to the right. This generator defines a dynamical map $V(t) : \mathscr{T}(\mathscr{H}) \to \mathscr{T}(\mathscr{H})$ that evolves the density operator in time $\rho(t) = V(t)\rho(0) = e^{\mathscr{L}_C t}\rho(0) = U(t)\rho U^\dagger(t)$, using the unitary time evolution operators $U(t) = e^{-iHt}$. The one-parameter family of maps $\{V(t)|t \in \mathbb{R}\}$ defines a positive group which is defined for positive and negative t due to the time-reversal symmetry of the unitary evolution [20].

If one traces out the degrees of freedom of the bath Hilbert space $\mathscr{H}_B$ and considers only the dynamics of the system Hilbert space $\mathscr{H}_S$, then the dynamics become in general nonunitary (see e.g. the derivation of the master equations from first principles in chapter 2.2.2). For special systems, the time evolution can still be defined via a dynamical map $\rho_S(t) = \mathrm{Tr}\{\rho(t)\}_B = V_S(t)\rho_S(t) = e^{\mathscr{L}t}\rho_S(t)$ with a generator $\mathscr{L} \neq \mathscr{L}_C$. The one-parameter family of the these maps $\{V_S(t)|t \in \mathbb{R}_+\}$ defines a positive semigroup which is defined only for positive times and fundamentally nonunitary. This is caused by the loss of information, due to not accounting for the dynamics of the bath degrees of freedom. In this work we only consider the special case of quantum Markovian semigroups (QMS) [6, 20, 22, 23, 126]. For notational simplicity and since we are interested only in the system dynamics, in the following we will write the system density operator without subscript $\rho_S = \rho$. The most general form of the generator of these semigroups, for finite dimensional Hilbert spaces of dimension $\dim(\mathscr{H}_S) = M$, are given by the Gorini Kossakowski Sudarshan Lindblad (GKSL) form [22, 23]

$$\mathscr{L}(\rho) = -i[H,\rho] + \sum_{k,\ell=0}^{M^2-1} a_{k,\ell}\left(L_k\rho L_\ell^\dagger - \frac{1}{2}\{L_\ell^\dagger L_k, \rho\}\right) = -i[H,\rho] + \mathscr{D}(\rho) \tag{4.1}$$

for a set of operators $\{L_k\}$ and a positive semidefinite matrix $\{a_{k,\ell}\}_{k,\ell} \geq 0$, defining the

open dynamics' dissipator $\mathscr{D}$. It is assumed in the following that there always exists a faithful density operator $\pi \in \mathscr{T}(\mathscr{H}_S)$, that is, the operator π fulfills $\mathscr{L}(\pi) = 0$, i.e. π is a steady-state of the dynamics. Further we assume that this operator π is uniquely defined. Using the trace operation for single time expectation values $\langle A \rangle = \mathrm{Tr}\{A\rho\}$ one can define an adjoint map with generator $\mathrm{Tr}\{A\mathscr{L}(\rho)\} = \mathrm{Tr}\{\overline{\mathscr{L}}(A)\rho\}$, which often is also a QMS [126] and its form is given by

$$\overline{\mathscr{L}}(A) = i[H, A] + \sum_{k,\ell=0}^{M^2-1} a_{k,\ell}\left(L_\ell^\dagger A L_k - \frac{1}{2}\{L_\ell^\dagger L_k, A\}\right) = i[H, A] + \overline{\mathscr{D}}(A), \tag{4.2}$$

with the adjoint dissipator $\overline{\mathscr{D}}$. The time-evolution of the operator is then given by $A(t) = e^{\overline{\mathscr{L}}t}A$. For one-time averages one thus can formally write $\langle A \rangle(t) = \mathrm{Tr}\{A(t)\rho\} = \mathrm{Tr}\{A\rho(t)\}$. If either (4.1) or (4.2) can be solved, then any one-time average can be calculated.

It should be stressed, however, that the adjoint dynamics are derived solely for one-time averages. In general the adjoint dynamics defined by the generator (4.2) does not have to hold for multi-time averages. Nevertheless, essential for response theory is the calculation of correlation functions $\langle A(t)B(t')\rangle$, which are two-time averages. In the case of unitary groups of the closed dynamics, these are always well defined as the dynamics are complete and the time evolution of the individual operators also contain the full dynamics of the complete system. However, for open dynamics this is not the case.

In general one has to rederive the time-evolution of these correlation functions from the unitary model to capture any additional effects that can be caused by the system-bath coupling [6, 15, 127].

However, a useful theorem, called the quantum regression theorem, allows one to use the adjoint dynamics also for correlation functions, for systems that are weakly coupled to their baths [6, 127]. In particular, the quantum regression theorem states that the time-evolution of the two-time correlation function is given by the dynamical properties of the one-time averages,

$$\frac{\mathrm{d}}{\mathrm{d}\tau}\langle A(t+\tau)B(t)\rangle = \left\langle \overline{\mathscr{L}}(A(t+\tau))B(t)\right\rangle. \tag{4.3}$$

This is of course not always the case, in fact, Talkner [128] showed an example where the assumption of the regression theorem for a thermal system resulted violations of the KMS condition. For the rest of this thesis we consider solely system for which the quantum regression theorem holds.

4.2.2. 0-DUAL MAPS AND QUANTUM DETAILED BALANCE

For unitary dynamics, a steady-state $\pi = e^{-\beta H}/Z$ of temperature β is given by the Gibbs state. For such states, quantum detailed balance can be defined as

$$\langle A(t)B(t')\rangle = \mu_A\mu_B\langle B(t)A(t')\rangle, \tag{4.4}$$

where the $\mu_{A,B}$ is the parity of the operator A, B under time reversal [129]. Even if the quantum Markovian semigroup's faithful state π is also given by a Gibbs state, this does not necessarily mean that detailed balance (4.4) is fulfilled in the open case[130, 131].

One way to define detailed balance in the open quantum context, is by considering the following dual generator, called the 0-dual generator $\overline{\mathscr{L}}^0$ [131], which can be defined, w.r.t. the faithful density operator π of $\mathscr{L}$, on the Banach space of operators of the system $\mathscr{B}(\mathscr{H}_s)$. The defining equation for the generators is

$$\mathrm{Tr}\left\{\rho\overline{\mathscr{L}}(A)B\right\} = \mathrm{Tr}\left\{A\mathscr{L}(B\rho)\rho^{-1}\rho\right\} =: \mathrm{Tr}\left\{\rho A\overline{\mathscr{L}}^0(B)\right\}, \tag{4.5}$$

or in short $\overline{\mathscr{L}}^0 A = \mathscr{L}(A\rho)\rho^{-1}$. Mathematically, the 0-dual is a special (physical) case of the more general s-dual which is defined for the scalar product $(A,B)_s = \mathrm{Tr}\{\rho^{1-s}A^\dagger\rho^s B\}$ [131–133]. Physically, this 0-dual comes about, because the open dynamics is characterized by a semigroup, instead of a unitary group. For closed dynamics one has

$$\langle A(t)B\rangle = \mathrm{Tr}\{U^\dagger(t)AU(t)B\rho\} = \mathrm{Tr}\{AU(t)BU^\dagger(t)U(t)\rho U^\dagger(t)\} = \langle AB(-t)\rangle \tag{4.6}$$

for steady-states $[H,\rho]=0$. For semigroups this identity with '$-t$' is not possible and thus the more general 0-dual is needed. For the semigroup's dynamical maps one can write

$$\mathrm{Tr}\left\{\rho e^{\overline{\mathscr{L}}t}(A)B\right\} = \mathrm{Tr}\left\{\rho Ae^{\overline{\mathscr{L}}^0 t}(B)\right\}. \tag{4.7}$$

The 0-dual also acts on elements of the $\mathscr{B}(\mathscr{H}_S)$. However, it is not necessarily a QMS itself [131]. Detailed balance can then be defined if the 0-dual maps generated by $\overline{\mathscr{L}}^0$ are QMS, the faithful state π commutes with the Hamiltonian and the 0-dual generator has the property $\overline{\mathscr{L}}^0 - \overline{\mathscr{L}} = 2i[H,\cdot]$.

If the QMS fulfills this detailed balance condition, then a useful property emerges. In this case, all generators $\overline{\mathscr{L}}, \mathscr{L}, \overline{\mathscr{L}}^0$ commute with the modular group [131]

$$\sigma_t(A) = \pi^{it}A\pi^{-it} \tag{4.8}$$

of the faithful state π for all $t\in\mathbb{R}$. This group can be further continued to imaginary parameters $|\mathrm{Im}(t)| \le 1/2$ [131, 133].

4.2.3. KUBO RESPONSE THEORY

Here we shortly note the results of quantum response theory for closed dynamics and equilibrium systems as derived by Kubo [38, 112]. A thermal system is perturbed by a Hamiltonian $\varepsilon(t)H_I$. The dynamics of the expectation value of an observable A can be written in linear response as $\langle A\rangle(t) = \langle A\rangle_0 + \int_0^t \varepsilon(s)\mathscr{R}_{AH_I}(t-s)\,\mathrm{d}s$, where $\langle\cdot\rangle_0$ denotes the unperturbed (equilibrium) expectation value and $\mathscr{R}_{AH_I}$ is the linear response function. Kubo derived two different forms for this response function [38]. One form is given as a commutator of the perturbation Hamiltonian and the observable of interest ($\tau = t-s$)

$$\mathscr{R}_{AH_I}(\tau) = i\langle[A(\tau),H_I]\rangle_0. \tag{4.9}$$

Upon using quantum detailed balance (4.4), Eq. (4.9) expresses a symmetry w.r.t. time-reversal, $\mathscr{R}_{AH_I}(\tau) = \mu_A\mu_{H_I}i\langle[H_I(\tau),A]\rangle_0 = \mu_A\mu_{H_I}\mathscr{R}_{H_IA}(\tau)$, where μ_{A,H_I} are the parities of the operators (more details about time-reversal will be provided in chapter 5 and in the appendix 7.8) and $\mathscr{R}_{H_IA}(\tau)$ is the response for the observable H_I and perturbation

A. For an alternative form of the response function, Kubo [38] introduced the nowadays called Kubo transform

$$\beta\overline{H_I} = \int_0^\beta d\lambda e^{-\lambda H_0} H_I e^{\lambda H_0}. \tag{4.10}$$

The response function can then be written as

$$\mathscr{R}_{AH_I}(\tau) = \beta \frac{\mathrm{d}}{\mathrm{d}t} \left\langle A(\tau)\overline{H_I} \right\rangle. \tag{4.11}$$

This transform has the advantage that (4.11) is almost identical to the classical results [25], where the dynamical variable H_I is replaced by the corresponding observable subject to the Kubo transform in the quantum case. Equation (4.11) further shows the same time-reversal symmetry as (4.9) since

$$\begin{aligned} \mathscr{R}_{AH_I}(\tau) &= \left\langle A(\tau)\overline{H_I} \right\rangle = \mu_A\mu_{H_I} \left\langle \overline{H_I}(\tau)^\dagger A \right\rangle = \mu_A\mu_{H_I} \left\langle \overline{H_I(\tau)}^\dagger A \right\rangle \\ &= \mu_A\mu_{H_I} \left\langle H_I(\tau)\overline{A} \right\rangle = \mu_A\mu_{H_I}\mathscr{R}_{H_IA}(\tau), \end{aligned} \tag{4.12}$$

where $\overline{H_I}(\tau) = \overline{H_I(\tau)}$ is the case since the Kubo transform (4.10) clearly commutes with the time-evolution unitaries.

4.3. THE EQUIVALENCE CLASSES OF OPEN QUANTUM RESPONSE THEORY

In the following we derive the general equivalence classes for steady-state quantum response functions of open quantum systems as generalizations of the classical ones in [37].

Linear response theory is defined as a small perturbation about a stationary state. The unperturbed system is described by an arbitrary QMS with unperturbed generator $\mathscr{L}_0$ of the form (4.1), assumed to possess a faithful state π_0. It is assumed that the system is subject to N individual small perturbations, whose form may be also given by (4.1) in general. The total time-dependent generator thus is given by $\mathscr{L} = \mathscr{L}_0 + \sum_n \varepsilon_n(t)\mathscr{L}_1^n$ with time-dependent parameters $\varepsilon_n(t)$. For linear response theory, such a form can always be obtained by expanding a general time-dependent generator about $\varepsilon_n = 0$ up to first order $\mathscr{L} = \mathscr{L}_0 + \sum_n \varepsilon_n(t)\mathscr{L}_1^n + O(\varepsilon_n^2)$. The complete dynamics of the system's density operator $\rho(t)$ is then given by $d_t\rho(t) = \mathscr{L}\rho(t)$ [25]. For arbitrary fixed $\varepsilon_n(t) = \varepsilon_n$, we assume that there always exists a faithful steady-state of the form $\pi_\varepsilon = \pi_0 + \sum_n \varepsilon_n\pi_1^n$, such that

$$\begin{aligned} \mathscr{L}\pi_\varepsilon &= \mathscr{L}_0\left(\pi_0 + \sum_{n=1}^N \varepsilon_n\pi_1^n\right) + \sum_{n=1}^N \varepsilon_n\mathscr{L}_1^n\pi_0 + O(\varepsilon_n^2) = 0 + O(\varepsilon_n^2) \\ &= \sum_{n=1}^N \varepsilon_n\mathscr{L}_0\pi_1^n + \varepsilon_n\mathscr{L}_1^n\pi_0 + O\left(\varepsilon_n^2\right). \end{aligned} \tag{4.13}$$

From (4.13) one can find that the steady-state density operator contributions π_1^n have the property $\mathscr{L}_0\pi_1^n = -\mathscr{L}_1^n\pi_0$ up to first order in the ε_n. If the system is initially stationary with the unperturbed steady-state π_0, the general form of the time dependent density

operator may be written as $\rho(t) = \pi_0 + \rho_1(t)$. Up to first order in $\varepsilon_n(t)$, the dynamics are then described by

$$\dot{\rho}(t) = \sum_{n=1}^{N} \varepsilon_n(t)\mathscr{L}_1^n\pi_0 + \mathscr{L}_0\rho_1(t), \tag{4.14}$$

whose formal solution is given by $\rho_1(t) = \int_0^t ds \sum_n \varepsilon_n(s) e^{\mathscr{L}_0(t-s)}\mathscr{L}_1^n\pi_0$, where $e^{\mathscr{L}_0(t-s)}$ is the map of the unperturbed system (see properties in 4.2.1). The response of the system to these small perturbations may then be calculated for any observable A up to linear order by

$$\begin{aligned}\langle A\rangle_\varepsilon(t) &= \langle A\rangle_0 + \sum_{n=1}^{N}\int_0^t ds\,\varepsilon_n(s)\mathrm{Tr}\left\{Ae^{\mathscr{L}_0(t-s)}\mathscr{L}_1^n\pi_0\right\} \\ &= \langle A\rangle_0 + \sum_{n=1}^{N}\int_0^t ds\,\varepsilon_n(s)\mathscr{R}^n(t-s).\end{aligned} \tag{4.15}$$

Here $\langle A\rangle_\varepsilon(t) = \mathrm{Tr}\{A\rho(t)\}$ denotes the perturbed expectation value of A and $\langle A\rangle_0 = \mathrm{Tr}\{A\pi_0\}$ the corresponding unperturbed expectation value. The response function for each individual perturbation is then given by $\mathscr{R}^n(\tau) = \mathrm{Tr}\left\{Ae^{\mathscr{L}_0\tau}\mathscr{L}_1^n\pi_0\right\}$ with $\tau = t - s$.

For equilibrium states π_0, one perturbation and closed dynamics, Eq. (4.15) leads to the usual Kubo response theory for closed quantum systems, as discussed in 4.2.3 for which $\mathscr{L}_0\cdot = -(i/\hbar)[H_0,\cdot]$ [25, 112]. For general (nonequilibrium) steady-states π_0 and open dynamics, it provides the basis for our quantum extension of the three equivalence classes identified in Ref. [37].

4.3.1. CLASS ONE

The first form $\mathscr{R}_1^n(\tau)$ of the quantum response function is expressed as a correlation function with a (nonHermitian) operator $B_1^n = (\mathscr{L}_1^n\pi_0)/\pi_0$. It follows from Eq. (4.15) by using the adjoint time evolution of the unperturbed dynamics $A(\tau) = e^{\overline{\mathscr{L}}_0\tau}A = Ae^{\mathscr{L}_0\tau}$, where $\mathscr{L}$ acting to the left can be interpreted as $\overline{\mathscr{L}}$ acting to the right. Assuming that the quantum regression theorem holds (see discussion in 4.2.1), the response function then reads

$$\mathscr{R}_1^n(\tau) = \langle A(\tau)B_1\rangle = \left\langle A(\tau)(\mathscr{L}_1^n\pi_0)/\pi_0\right\rangle. \tag{4.16}$$

Expression (4.16) is a quantum generalization of the linear response function derived in Ref. [115], and is often referred to as Agarwal-form for this reason [37, 125]. For the case of Hamiltonian perturbation $\mathscr{L}_1^n = -i[H_I^n,\cdot]$, this form readily shows that for a thermal stationary distribution, $\pi_0 = \exp(-\beta H_0)/Z_0$, with Z_0 the partition function for the unperturbed system, the quantum response vanishes when the perturbation commutes with the unperturbed Hamiltonian, $[H_I^n, H_0] = 0$. This is not necessarily the case for a quantum nonequilibrium steady-state, as we will discuss in detail for an example in chapter 4.6. For a general perturbation $\mathscr{L}_1^n$ it is the case that the response vanishes if π_0 is also an invariant state of $\mathscr{L}_1^n$. This variant of the fluctuation-response theorem is distinguished by the fact that it contains only state variables and no time derivatives. Its drawback is that the operator B_1 involves the stationary distribution π_0, which is not always explicitly known in concrete situations.

4.3.2. CLASS TWO

In the classical regime, the second variant is written in terms of the time derivative of the ε-derivative of the stochastic entropy of the system, $\partial_\varepsilon S_\varepsilon|_0 = -\partial_\varepsilon \ln \pi_\varepsilon|_0 = -\pi_1/\pi_0$, along single trajectories [37, 115]. We obtain the second form $\mathscr{R}_2^n(\tau)$ of the quantum response functions by using the identity derived from (4.13), $\mathscr{L}_1^n \pi_0 = -\mathscr{L}_0 \pi_1^n$, which is valid up to linear order in ε_n. Further, since $\mathscr{L}_0$ is the generator of the unperturbed open dynamics, we have $e^{\mathscr{L}_0(t-s)}\mathscr{L}_0\rho = d_t e^{\mathscr{L}_0(t-s)}\rho = d_\tau e^{\mathscr{L}_0\tau}\rho = -d_s e^{\mathscr{L}_0(t-s)}\rho$. The response function in Eq. (4.15) may therefore be rewritten as

$$\mathscr{R}_2^n(\tau) = -\mathrm{Tr}\left\{A(\tau)\mathscr{L}_0\pi_1^n\right\} = -d_\tau \left\langle A(\tau)\pi_1^n/\pi_0 \right\rangle. \tag{4.17}$$

In the limit of closed quantum systems at equilibrium and Hamiltonian perturbation, Eq. (4.17) reduces to the Kubo quantum response function (4.11), since

$$\pi_1/\pi_0 = \beta\overline{H_I} = \int_0^\beta d\lambda e^{-\lambda H_0} H_I e^{\lambda H_0} \tag{4.18}$$

is given by the Kubo transform of H_I [110] as noted in (4.10). The advantage of the Kubo transformation (4.10) is that it allows to formulate classical and quantum equilibrium response functions in the same form by simply replacing an operator by its corresponding transform. Such a procedure can be carried over to steady-state response functions for open systems.

In order to bring Eq. (4.17) in a form similar to the classical case, we first introduce a generalized Kubo transformation $\overline{\partial_\varepsilon \ln \pi_\varepsilon|_0}$ of $\partial_\varepsilon \ln \pi_\varepsilon|_0$. Using an operator identity (for the identity and its derivation see appendix 7.7), the partial derivative can be rewritten as

$$\partial_{\varepsilon_n}\pi_\varepsilon|_0/\pi_0 = \pi_1^n/\pi_0 = \int_0^1 \pi_0^\lambda(\partial_{\varepsilon_n}\ln\pi_\varepsilon)|_0\pi_0^{-\lambda}d\lambda =: \overline{\partial_{\varepsilon_n}\ln\pi_\varepsilon|_0}. \tag{4.19}$$

The latter reduces to the usual Kubo transform (4.10) even for open dynamics, if e.g. $\pi_\varepsilon = e^{-\beta(H_0+\sum_n \varepsilon_n H_I^n)}/Z_\varepsilon$ holds true. It may thus be regarded as a steady-state extension of the Kubo inner product [134] for thermal states, also called the Bogoliubov or Kubo-Mori inner product [135]. We accordingly find,

$$\mathscr{R}_2^n(\tau) = -d_\tau \left\langle A(\tau)\overline{\partial_{\varepsilon_n}\ln\pi_\varepsilon|_0}\right\rangle = d_\tau \left\langle A(\tau)\overline{\partial_{\varepsilon_n}S_\varepsilon|_0}\right\rangle, \tag{4.20}$$

where we have introduced the quantum analog of the stochastic entropy $S_\varepsilon = -\ln\pi_\varepsilon$. Noting furthermore, that two-time correlation functions for open quantum systems are defined as $\langle A(t)B(s)\rangle = \mathrm{Tr}\left\{Ae^{\mathscr{L}_0(t-s)}Be^{\mathscr{L}_0 s}\rho(0)\right\}$ [6, 24] (a more thorough discussion of open quantum correlation functions will be provided in chapter 5.3.1), we obtain, with $\rho(0) = \pi_0$ and $d_s e^{\mathscr{L}_0 s}\pi_0 = 0$,

$$\mathscr{R}_2^n(\tau) = -d_s\left\langle A(\tau)\overline{\partial_{\varepsilon_n}S_\varepsilon|_0}\right\rangle = -\left\langle A(t)d_s\overline{\partial_{\varepsilon_n}S_\varepsilon(s)|_0}\right\rangle. \tag{4.21}$$

Formula (4.21) is a quantum extension of the response function of Refs. [37, 115]. It can be formally written in Liouville space as a correlation function with the observable $B_2^n(s) = -d_s\overline{\partial_{\varepsilon_n}S_\varepsilon(s)|_0}$.

Let us consider some properties that can be deduced from the second form. Due to the noncommutativity of quantum mechanics, it is in general the case that for noncommuting operators $\partial_\varepsilon \ln(\pi_0+\varepsilon\pi_1)|_0 \neq \pi_1/\pi_0$, implying that $\overline{\partial_{\varepsilon_n} S_\varepsilon} \neq \partial_{\varepsilon_n} S_\varepsilon$. Consequently, the quantum response function (4.21) cannot be written in terms of the stochastic entropy, $-\langle A(t) d_s \partial_\varepsilon S_\varepsilon(s)|_0\rangle$, as in the classical limit, unless $[\pi_0,\pi_1]=0$. If this is the case, however, we will show that the response will vanish for Hamiltonian perturbation and thermal response, thus there is no response in these cases. We show this by considering only one perturbation $\varepsilon_n=\varepsilon$ (the same holds true for multiple perturbations if all π_1^n commute with π_0). For commuting density operators, we indeed have $\partial_\varepsilon \ln(\pi_0+\varepsilon\pi_1)|_0 = \pi_1/\pi_0$, since then π_0,π_1 possess the same eigenbasis, transforming the operator logarithm into a simple c-number equation in the common eigenbasis, for which the differentiation can be accomplished accordingly. Moreover, for a thermal state and Hamiltonian perturbation $\pi_\varepsilon \propto e^{-\beta(H_0+\varepsilon H_I)}$, the operator π_1 can be given explicitly via the Kubo transform, $\pi_1 \propto \tilde{H}_I \pi_0$ (see Eq. (7.12) in Ref. [25]). As a result, the condition $[\pi_0,\pi_1]=0$ implies that $[H_0,H_I]=0$ since the Kubo transform $\beta\tilde{H}_I = \int_0^\beta d\lambda e^{-\lambda H_0} H_I e^{\lambda H_0}$ consists only of exponentials that are proportional to H_0 and the operator H_I. Therefore, $\mathscr{L}_1\pi_0=0$ follows for Hamiltonian perturbation. The first class (4.16) then dictates that the response function has to vanish.

Interestingly, for nonequilibrium situations or general (dissipative) perturbations, i.e. $\mathscr{L}_1\rho = -i[H_I,\rho]+\mathscr{D}_I(\rho)$, there may exist cases where $[\pi_0,\pi_1]=0$, but there is still a response and thus the quantum system reacts similarly to classical systems. This is the case, if $[\pi_0,\pi_1]=0$, but $\mathscr{L}_1\pi_0\neq 0$, $\mathscr{L}_0\pi_1\neq 0$. One example for such a case is given by solely modifying the temperature of the bath of the single oscillator model (2.6).

In the unitary limit, the formal identity in Liouville space for the correlation function $\left\langle A(t) d_s \overline{\partial_{\varepsilon_n} S_\varepsilon(s)|_0}\right\rangle$ becomes one at the operator level in Hilbert space. In that case, the unitary time-evolution operators $e^{\overline{\mathscr{L}}_0 t} A = U^\dagger(t,0) A U(t,0) = A(t)$ can be used to obtain the Heisenberg representation of the generalized stochastic entropy, since for general operators $A.B$ it is then the case that $\left\{U^\dagger(t,s)AU(t,s)BU(s,0)\rho(0)U^\dagger(s,0)\right\} = \langle A(t)B(s)\rangle$. The latter equality gives the observable $-d_s\overline{\partial_{\varepsilon_n} S_\varepsilon(s)|_0}$ a meaning in Hilbert space.

To conclude the discussion of this form, let us note that the variant (4.21) is the only one where the response function is given as a correlation function with a time derivative of a state variable, namely the formal time derivative of the generalized Kubo transformed ε_n-derivative of the stochastic entropy, $\overline{\partial_{\varepsilon_n} S_\varepsilon|_0}$. Let us additionally mention that there is an alternative way of writing the quantum response function (4.21) without using any correlation function. We indeed have,

$$\mathscr{R}^n_{2,\mathrm{alt}}(\tau) = -d_\tau \partial_{\varepsilon_n} \mathrm{Tr}\{A(\tau)\pi_\varepsilon\}\big|_0 = -d_\tau \partial_{\varepsilon_n} \langle A(\tau)\rangle_\varepsilon|_0, \tag{4.22}$$

where $\langle A(\tau)\rangle_\varepsilon = \mathrm{Tr}\{A(\tau)\pi_\varepsilon\} \neq \langle A\rangle_\varepsilon(\tau)$ is the perturbed expectation value of the observable $A(\tau) = e^{\overline{\mathscr{L}}_0\tau} A$ evolved via the unperturbed dynamics. This form offers an intuitive interpretation of dynamic response theory: at any fixed time, $\partial_{\varepsilon_n}\langle A(\tau)\rangle_\varepsilon|_{\varepsilon=0}$ can be seen as the static susceptibility, that is, the static response of the system to the external perturbation [25]. The dynamic response function (4.22) then follows as the time derivative of the time-dependent susceptibility. This form often enables a simple evaluation of the response function (see section 4.6).

4.3.3. CLASS THREE

Classically, the third form is the unique one that does not explicitly involve the stationary distribution [37, 120]. This type of fluctuation-response relation is therefore of advantage when the steady-state distribution is not specifically known. Such a variant may be derived from Eq. (4.16) by realizing that the generator $\mathscr{L}_1^n$ in (4.16) can act on $A(\tau)$ by using the adjoint generator $\overline{\mathscr{L}}_1^n$. Then we obtain

$$\mathscr{R}_3^n(\tau) = \mathrm{Tr}\left\{\pi_0 \overline{\mathscr{L}}_1^n A(\tau)\right\} = \left\langle \overline{\mathscr{L}}_1^n A(\tau)\right\rangle. \tag{4.23}$$

More explicitly, for the case of Hamiltonian perturbation $\overline{\mathscr{L}}_1 \cdot = (i/\hbar)[H_I, \cdot]$ one receives

$$\mathscr{R}_3^n(\tau) = \frac{i}{\hbar}\mathrm{Tr}\left\{\pi_0 \left[H_I^n, A(\tau)\right]\right\} = \frac{i}{\hbar}\left\langle \left[H_I^n, A(\tau)\right]\right\rangle. \tag{4.24}$$

In contrast to Eqs. (4.16),(4.21),(4.22), the response function (4.23) is given as an expectation value of operators that do not explicitly depend on either π_0, π_1 or π_ε (see also Ref. [42] for an alternative approach using the eigenoperator decomposition of (4.24)). However, for generators in their general form (4.2), the response function (4.24) is not just given by a commutator. The bath effect also has to be taken into account in the form of the dissipator $\mathscr{D}_1$ in (4.23). While for Hamiltonian perturbation, the open quantum system response Eq. (4.24) resembles the Kubo quantum response form (4.9), they only coincide in the limit of unitary quantum systems at equilibrium. Interestingly, expression (4.24) indicates that for purely Hamiltonian perturbations, the quantum response function vanishes when the time evolved observable $A(\tau)$ commutes with the perturbation Hamiltonian H_I. Further, for purely Hamiltonian perturbation, the quantum response function can be identified with the expectation value of the imaginary part of the correlation function $\langle A(\tau)H_I\rangle = \langle A(\tau)H_I + H_I A(\tau)\rangle/2 + i\langle [A(\tau), H_I]/i\rangle/2$ for a nonequilibrium steady-state, as in the unitary limit [110]. General (Non-Hamiltonian) perturbations, however, destroy this strong analogy.

4.4. SYMMETRIES FOR QUANTUM DETAILED BALANCE QMS

In this section we consider what symmetries there are for master equations fulfilling quantum detailed balance as discussed in 4.2.2. Let's first consider the third form for purely Hamiltonian perturbation (4.24). For generic equilibrium correlation functions of two observables of detailed balance QMS one has $\langle AB(t)\rangle = \mu_A\mu_B\langle BA(t)\rangle$ [129]. The Hamiltonian response therefore has the symmetry

$$\mathscr{R}_m^n(\tau) = \frac{i}{\hbar}\langle [H_I^m(\tau), H_I^n]\rangle = \mu_{H_I^m}\mu_{H_I^n}\frac{i}{\hbar}\langle [H_I^n(\tau), H_I^m]\rangle = \mu_{H_I^m}\mu_{H_I^n}\mathscr{R}_n^m(\tau), \tag{4.25}$$

where $\mathscr{R}_n^m(\tau) = i/\hbar[H_I^n(\tau), H_I^m]$ corresponds to a Hamiltonian perturbation H_I^m for the observable H_I^n. Thus, up to the time-reversal parities, this response function class has a symmetric time-reversal symmetry between the operators H_I^n and H_I^m in the case of detailed balance QMS. Since this symmetry is the same for the closed dynamic's Kubo form (see (4.12)), it can be seen that the quantum detailed balance QMS resembles the closed behavior.

Next let us consider the second form (4.21) for completely general perturbations. For a set of observables $A_m = \partial_{\varepsilon_m} \ln \pi_\varepsilon|_0$ corresponding to the respective perturbations $\varepsilon_m(t)\mathscr{L}_1^m$ and quantum detailed balance QMS unperturbed dynamics described by the generator $\mathscr{L}_0$, the response function for a perturbation denoted by the letter n can be written in the second form as $\mathscr{R}_m^n(\tau) = \left\langle A_m(\tau)\overline{\partial_{\varepsilon_n} \ln \pi_\varepsilon|_0}\right\rangle$. Using the 0-dual (4.7), for which one has $\left\langle \exp\left(\overline{\mathscr{L}}_0 t\right)(A)B\right\rangle = \left\langle A \exp\left(\overline{\mathscr{L}}_0^0 t\right)B\right\rangle$, one can rewrite this response function

$$\begin{aligned}\mathscr{R}_m^n(\tau) &= \mathrm{d}_\tau \left\langle e^{\overline{\mathscr{L}}_0 \tau}(A_m)\overline{\partial_{\varepsilon_n} \ln \pi_\varepsilon|_0}\right\rangle = \mathrm{d}_\tau \left\langle A_m e^{\overline{\mathscr{L}}_0^0 \tau}\overline{\partial_{\varepsilon_n} \ln \pi_\varepsilon|_0}\right\rangle \\ &= \mathrm{d}_\tau \left\langle A_m \overline{e^{\overline{\mathscr{L}}_0^0 \tau}\partial_{\varepsilon_n} \ln \pi_\varepsilon|_0}\right\rangle = \mathrm{d}_\tau \left\langle e^{\overline{\mathscr{L}}_0^0 \tau}(\partial_{\varepsilon_n} \ln \pi_\varepsilon|_0)\overline{A_m}\right\rangle,\end{aligned} \tag{4.26}$$

where it was used that for quantum detailed balance QMS the generators $\overline{\mathscr{L}}_0^0, \overline{\mathscr{L}}_0$ commute with the modular group σ_t (defined in (4.8)) and therefore

$$\overline{\mathscr{L}}_0^0\, \overline{A} = \int_0^1 \overline{\mathscr{L}}_0^0 \rho^\lambda A \rho^{-\lambda} \mathrm{d}\lambda = \int_0^1 \overline{\mathscr{L}}_0^0 \sigma_{i\lambda}(A)\mathrm{d}\lambda = \overline{\mathscr{L}_0^0 A}. \tag{4.27}$$

It was further used that for the generalized Kubo transform (4.19) one can find $\langle A\overline{B}\rangle = \langle B\overline{A}\rangle$. Furthermore, applying time-reversal (more details about time-reversal will be provided in 5 and 7.8) on (4.26), one can use the fact, that the time-reversal of $\overline{\mathscr{L}}_0^0$, $\overline{\mathscr{L}}_0^{0R} = \overline{\mathscr{L}}_0$ is just given by the initial adjoint dynamic [131–133]. This is the case, because for quantum detailed balance QMS, the 0-dual is given by the general identity $\overline{\mathscr{L}}_0^0 - \overline{\mathscr{L}}_0 = 2i[H,\cdot]$, which has the same form [133] as the time-reversal of the $\overline{\mathscr{L}}_0$ and applying time-reversal two consecutive times onto a superoperator will result in the initial dynamics. Therefore, up to the time-reversal parities (since $A_n^{\dagger,R} = \Theta A_n^\dagger \Theta = \mu_A A_n$ for Hermitian operators), the response functions $\mathscr{R}_n^m(\tau)$, $\mathscr{R}_m^n(\tau)$ are equal,

$$\mathscr{R}_m^n(\tau) = \left\langle e^{\overline{\mathscr{L}}_0^0 \tau}(\partial_{\varepsilon_n} \ln \pi_\varepsilon|_0)\overline{A_m}\right\rangle = \mu_{A_n}\mu_{A_m}\left\langle A_m(\tau)\overline{\partial_{\varepsilon_n} \ln \pi_\varepsilon|_0}\right\rangle = \mu_{A_n}\mu_{A_m}\mathscr{R}_n^m(\tau). \tag{4.28}$$

We deduce that if the unperturbed dynamics fulfill quantum detailed balance, there is also a symmetric time-reversal symmetry for response functions connected to observables $A_n = \partial_{\varepsilon_n} \ln \pi_\varepsilon|_0$, as in the closed case (4.11) and open systems for Hamiltonian perturbation (4.25). However, the symmetry in (4.28) is valid for the more general operators $A_n = \partial_{\varepsilon_n} \ln \pi_\varepsilon|_0$ and for both Hamiltonian as well as dissipative perturbations. Interestingly, this symmetry still holds, even if the perturbation $\mathscr{L}_1$ does not fulfill quantum detailed balance, only $\mathscr{L}_0$ has to, as only the time-reversal symmetry of the unperturbed generators are relevant.

It is important to note, that in (4.27) it was used that the unperturbed QMS generators commute with $\sigma_{i\lambda}$ for $0 \leq \lambda \leq 1$ which was explicitly shown to be the case only for $0 \leq \lambda \leq 1/2$ in [131–133]. While we do not intend to generally, akin to a treatment in [131, 133], prove that this may be the case also for the larger interval, let us use an intuitive argument showing that this is in principle possible. In [131–133] it is shown that for detailed balance QMS the faithful state commutes with the Hamiltonian $[\pi_0, H_0] = 0$. This is the case for e.g. a Gibbs state $\pi_0 \propto \exp(-\beta H_0)$. Assuming purely Hamiltonian

perturbation, one has on the one hand from the third form the response function symmetry (4.25), $\mathscr{R}_{AH_I}(t) = \mu_A\mu_{H_I}\mathscr{R}_{H_IA}(t)$. On the other hand, from the second class symmetry (4.28) one can further find the following symmetry. For a Hamiltonian perturbation H_I, let us assume the corresponding perturbed steady-state is given by $\pi_{\varepsilon_1}^{H_I} = \exp(-\beta(H_0+\varepsilon_1 H_I))/Z_{\varepsilon_1}$ and for a perturbation A by $\pi_{\varepsilon_2}^{A} = \exp(-\beta(H_0+\varepsilon_2 A))/Z_{\varepsilon_2}$ and thus the general steady-state has the form $\pi_\varepsilon = \exp(-\beta(H_0+\varepsilon_1 H_I+\varepsilon_2 A)/Z_\varepsilon$. This assumption is generally used for closed linear response theory [25, 38]. One therefore has $\partial_{\varepsilon_i}\ln\pi_\varepsilon|_0 = B_i + \text{c-number}$, $B_{1,2} = H_I, A$, where the c-number term stems from the partition function Z_ε and vanishes in the response function as $\langle A(\tau)\rangle$ is constant. It then follows that $\mathscr{R}_2^1(\tau) = \mathrm{d}_\tau\langle A(\tau)\overline{H_I}\rangle$. But this is nothing else than the open dynamics' version of the Kubo response (4.11) and since this response has the same operators as the third form for pure Hamiltonian perturbation (4.25), the symmetry has to hold too. Thus one can deduce in this case

$$\mathscr{R}_2^1(\tau) = \mu_A\mu_{H_I}\mathscr{R}_1^2(\tau) = \mathrm{d}_\tau\langle A(\tau)\overline{H_I}\rangle = \mu_A\mu_{H_I}\mathrm{d}_\tau\langle H_I(\tau)\overline{A}\rangle. \tag{4.29}$$

But Eq. (4.29) is exactly the symmetry in (4.28), the generators therefore have to commute with the modular group also in the interval $0 \le \lambda \le 1$. Further, since this property is independent of the perturbation chosen, it is dependent only on the dynamical properties of the unperturbed system, it has to hold also for any kind of perturbation. While the above considerations are no definite 'proof', they are still reasonable arguments indicating that it is in general possible.

To conclude, let us stress that the above time-reversal symmetries exist only for detailed balance QMS, for general steady-states correlation functions these symmetries do not necessarily hold, even for Hamiltonian perturbation, as we show explicitly for an example below in 4.7.

4.5. COMPARISON WITH OTHER APPROACHES

In this part, the derived equivalence classes will be compared with results from various authors and their derived forms will then be put into the corresponding classes. Such quantum steady-state response functions have been obtained in e.g. Refs. [39, 40] using different methods. All of their forms are directly connected to the second class (4.17). Chetrite and Mallick have derived a steady-state fluctuation-dissipation relation from a quantum Jarzynski-Hatano-Sasa fluctuation theorem for the accompanying density operator π_t for a frozen time t [39]. This accompanying density operator satisfies for the time dependent generator $\mathscr{L} = \mathscr{L}_0 + \varepsilon(t)\mathscr{L}_1$, $\mathscr{L}_t\pi_t = 0$ for each time t, but depends explicitly on time, $d_t\pi_t \neq 0$. It is therefore different from the true density operator ρ_t. Considering a modified superoperator (using the notation of this paper) $\mathscr{L}' = \mathscr{L} + d_t\pi_t/\pi_t =: \mathscr{L} + W_t$ they have derived a quantum Jarzynski-Hatano-Sasa relation of the form,

$$\mathrm{Tr}\{\pi_t A\} = \left\langle \overrightarrow{\exp}\left(\int_0^t W_u\,du\right)A(t)\right\rangle_0, \tag{4.30}$$

where $\overrightarrow{\exp}$ is the time ordered exponential. Taking the functional derivative of Eq. (4.30) yields,

$$\mathscr{R}_{\text{Chetrite}}(t) = -\frac{d}{dt}\left\langle A(t)\pi_1\pi_0^{-1}\right\rangle, \tag{4.31}$$

which is equivalent to Eq. (4.17).

On the other hand, Mehboudi, Sanpera and Parrondo have derived a steady-state response function for open quantum systems described by Markovian completely positive and trace preserving maps $\zeta_\varepsilon(\rho)$ in terms of the symmetric logarithmic derivative (SLD)[40]. Assuming that each of these maps have an invariant state $\zeta_\varepsilon\pi_\varepsilon = \pi_\varepsilon$, they have considered these maps and their invariant states up to linear order, $\zeta_\varepsilon = \zeta_0 + \varepsilon\zeta_1$ and $\pi_\varepsilon = \pi_0 + \varepsilon\pi_1$. For discrete time steps, the time evolution is given by $\rho(t) = \zeta_{\varepsilon(t)} \circ \zeta_{\varepsilon(t-1)} \circ \cdots \circ \zeta_{\varepsilon(1)}\pi_0$. Expanding this evolution equation up to linear order in ε and taking the continuous limit, they arrived at

$$\mathscr{R}_{\text{Mehboudi}}(t) = -d_t \langle A(t)\Lambda_0 + \Lambda_0 A(t)\rangle /2, \tag{4.32}$$

where the symmetric logarithmic derivative is defined via $2\partial\varepsilon'\rho'_\varepsilon|_{\varepsilon'=\varepsilon} = (\Lambda_\varepsilon\rho_\varepsilon + \rho_\varepsilon\Lambda_\varepsilon)$. This response function can now be rewritten as,

$$\begin{aligned}\mathscr{R}_{\text{Mehboudi}}(t) &= -d_t \operatorname{Tr}\{(A(t)\Lambda_0 + \Lambda_0 A(t))\pi_0\} /2 \\ &= -d_t \operatorname{Tr}\{(\pi_0\Lambda_0 + \Lambda_0\pi_0)A(t)\} /2 \\ &= -d_t \operatorname{Tr}\{\partial\varepsilon\pi_\varepsilon|_{\varepsilon=0} A(t)\}.\end{aligned} \tag{4.33}$$

Since $\pi_\varepsilon = \pi_0 + \varepsilon\pi_1$ and $\partial\varepsilon\pi_\varepsilon|_{\varepsilon=0} = \pi_1$, we finally have,

$$\mathscr{R}_{\text{Mehboudi}}(t) = \frac{d}{dt}\operatorname{Tr}\{A(t)\pi_1\} = \frac{d}{dt}\left\langle A(t)\pi_1\pi_0^{-1}\right\rangle, \tag{4.34}$$

which is also directly connected to Eq. (4.17) of the main text.

Response functions are intimately related to the Fisher information [136]. Two quantum generalizations of the Fisher information are commonly considered: the Kubo-Mori-Bogoliubov inner product (KMB) and the symmetric logarithmic derivative (SLD) [137, 138]. Their main difference lies in the order of the noncommuting quantum observables. The SLD is most suited from the viewpoint of quantum estimation theory as it corresponds to the Cramer-Rao bound, while the KMB appears as the most natural quantum analogue of the Fisher information from the viewpoint of statistical physics; it is indeed closely related to the canonical correlation of equilibrium linear response theory [134]. Equation (4.32) can thus been regarded as a steady-state extension of the SLD approach, while Eq. (4.20) of the main text may be viewed as a steady-state and open dynamics generalization of the KMB approach.

Besides these quantum equivalences, the classes can also be compared with nonequilibrium results in the classical context. For example, the second form (4.20) can be considered as a quantum generalization of a classical NESS response derived by Prost et al. (Eq. (5) in [119]). Their form is given by

$$\mathscr{R}_{nm}(\tau) = \mathrm{d}_\tau \langle \partial_{\varepsilon_m}\Phi(\tau)\partial_{\varepsilon_n}\Phi\rangle \tag{4.35}$$

for the generalized potential $\pi_\varepsilon := e^{\Phi}$. This is clearly the classical limit of (4.20) for $A = \partial_{\varepsilon_n}\ln(\pi_\varepsilon)$.

4.6. APPLICATIONS OF LINEAR RESPONSE THEORY

Our results are applicable to general open quantum systems. As an illustration, we now consider once again different aspects of the coupled harmonic oscillator toy model from chapter 2. We will first consider the difference between steady-state and equilibrium response, for an open system subject to Hamiltonian perturbation. Afterwards we will consider the breakdown of linear response approaching a critical point and once more investigate the differences between the equilibrium and nonequilibrium case.

Despite the formal similarity between thermal $\pi_0 \propto e^{-\beta H}$ and general nonequilirium steady-states π_0, the two cases are fundamentally different: while equilibrium response for closed dynamics satisfies detailed balance [129], open (steady-state) response in general does not.

4.6.1. RESPONSE FOR THE WEAKLY COUPLED HARMONIC OSCILLATORS

The system consists of two weakly coupled harmonic oscillators, each interacting with its own reservoir at a different temperature (see Fig. 2.1 for illustration). By properly tuning the parameters of the system, this model allows one to compare different response regimes: unitary/dissipative, equilibrium/steady-state and classical/quantum. In particular, a nonequilibrium steady-state is established when the two bath temperatures are different. The (unperturbed) Hamiltonian of the system [49, 80, 139] is given by (2.2) for $\kappa = 0$,

$$H_0 = \hbar\omega_1 a_1^\dagger a_1 + \hbar(\omega_1 + \delta) a_2^\dagger a_2 + \hbar\lambda(a_1 a_2^\dagger + a_1^\dagger a_2), \tag{4.36}$$

with respective frequencies ω_1 and $\omega_2 = \omega_1 + \delta$ (with detuning δ). For being able to analyticaly treat the problem, the system-bath interaction is assumed to be such, that the dynamics are correctly described by the local master equation (see 2 for details about local master equations). For specially tuned system-bath couplings it is possible to create local dynamics, as was shown by e.g. [70].

The (unperturbed) quantum Markovian semigroup generator reads [140],

$$\mathscr{L}_0 \cdot = -(i/\hbar)[H_0, \cdot] + \sum_{j=1}^{2} \mathscr{D}_j[\cdot], \tag{4.37}$$

with the two nonunitary dissipators $\mathscr{D}_j$ given by the local approach dissipators (2.8)

$$\begin{aligned} \mathscr{D}_j[\rho] =& \gamma(N(\beta_j, \omega_j) + 1)\left[a_j \rho a_j^\dagger - \frac{1}{2}\left(a_j^\dagger a_j \rho + \rho a_j^\dagger a_j\right)\right] \\ &+ \gamma N(\beta_j, \omega_j)\left[a_j^\dagger \rho a_j + \frac{1}{2}\left(a_j a_j^\dagger \rho + \rho a_j a_j^\dagger\right)\right], \end{aligned} \tag{4.38}$$

for symmetrical system-bath couplings $\gamma_j = \gamma$. For concreteness, we apply a single type of Hamiltonian perturbation. In particular a step perturbation of the form $\varepsilon(t) H_I = \hbar\varepsilon(t)(a_1 a_2^\dagger + a_1^\dagger a_2)$, with $\varepsilon(t) = \varepsilon\Theta(t)$ is being considered, i.e. the coupling between the harmonic oscillators is being modified and calculate the response of the (dimensionless) energy of the first oscillator, $A = \beta_1 \hbar\omega_1 a_1^\dagger a_1$. We note that the unperturbed system is in a thermal state for $\lambda = 0$ and in a nonequilibrium steady-state for $\lambda \neq 0$. The two quantum oscillators are moreover closed with unitary dynamics in the absence of damping, $\gamma = 0$. Finally, the classical regime is achieved in the high-temperature limit $\beta_j \hbar\omega_j \ll 1$.

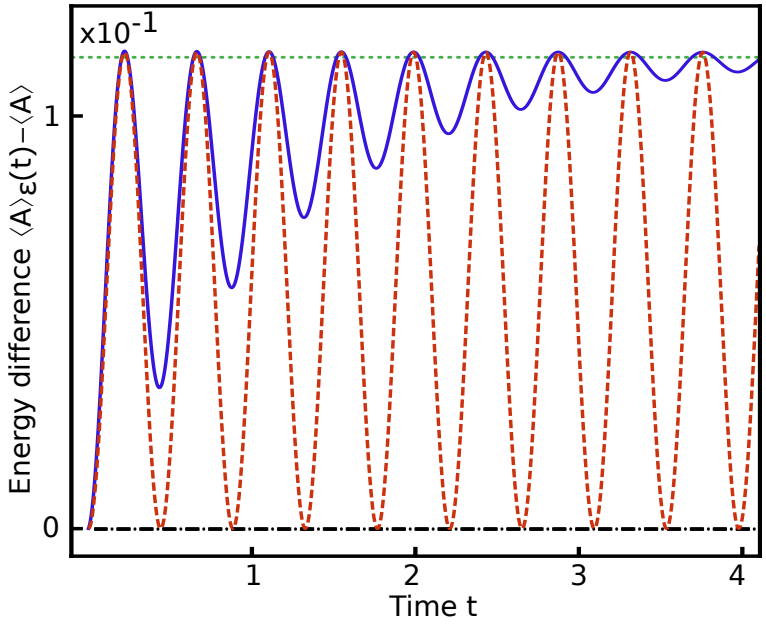

Figure 4.1: Quantum response of the (dimensionless) energy of the first oscillator, $\langle A\rangle_\varepsilon(t)-\langle A\rangle$, with $A=\beta_1\hbar\omega_1 a_1^\dagger a_1$, to a step perturbation, $\varepsilon(t)=\varepsilon\Theta(t)$, of the coupling between the two harmonic oscillators, for the rotating-wave Hamiltonian (2.2) ($\kappa=0$). The steady-state response ($\lambda\neq 0$) (blue solid), Eq. (4.39), asymptotically approaches the perturbed value (green dotted). By contrast, the equilibrium response ($\lambda=0$) (black dotted-dashed) vanishes and the unitary response ($\gamma=0$) (red dashed), Eq. (4.41), keeps oscillating and fails to reach the perturbed value of the observable A. Parameters are $\omega_1=2.4$, $\delta=10.1$, $\gamma=0.7$, $\lambda=5$, $\varepsilon=0.11$, $\beta_1=0.092$ and $\beta_2=0.0008$.

We determine the quantum response function using the forms (4.22) and (4.24). To this end, we first evaluate the steady-state density operator of the system and then calculate the time dependence of the observable A. The steady-state properties of the unperturbed system is solved using once again the symplectic space for calculating the covariance matrix σ(see appendix 7.1 for details). The time evolution of A is obtained via matrix exponentiation of a closed set of operator differential equations (see appendix 7.5 for details about solving the adjoint dynamics). We find then the response functions (see calculation details in 7.6)

$$\begin{aligned}\mathscr{R}_3(\tau)&=i\beta_1\hbar\omega_1\left\langle\left[a_1a_2^\dagger+a_1^\dagger a_2,a_1^\dagger a_1(\tau)\right]\right\rangle\\&=e^{-\gamma\tau}\frac{\gamma(\delta^2+4\lambda^2\cos z\tau)+(\gamma^2+\delta^2)z\sin z\tau}{z^2(\gamma^2+z^2)(2\lambda\Delta N\beta_1\hbar\omega_1)^{-1}}\\&=-\beta_1\hbar\omega_1 d_\tau\partial_\varepsilon\left\langle a^\dagger a(\tau)\right\rangle\Big|_{\varepsilon=0}=\mathscr{R}_{2,\mathrm{alt}}(\tau),\end{aligned}\tag{4.39}$$

where $\Delta N=N(\beta_2,\omega_2)-N(\beta_1,\omega_1)$ and $z=\sqrt{\delta^2+4\lambda^2}$. The two different forms $\mathscr{R}_{2,\mathrm{alt}}(\tau)$ and $\mathscr{R}_3(\tau)$ thus yield the same result, as expected. However, this is not obvious from their definitions (4.22) and (4.24), since $\mathscr{R}_{2,\mathrm{alt}}(\tau)$ displays an explicit ε-dependence, while

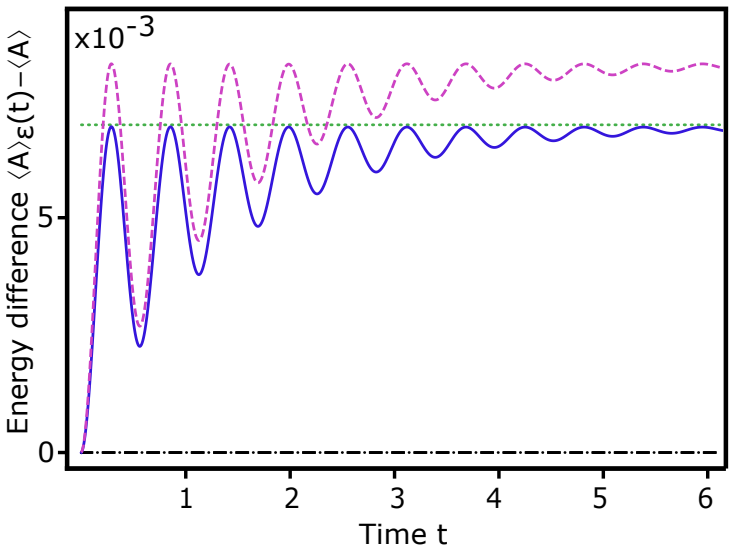

Figure 4.2: Steady-state response of the (dimensionless) energy of the first oscillator, $\langle A\rangle_\varepsilon(t)-\langle A\rangle$, with $A=\beta_1\hbar\omega_1 a_1^\dagger a_1$, to a step perturbation, $\varepsilon(t)=\varepsilon\Theta(t)$, of the coupling between the two harmonic oscillators. The quantum response ($\beta_1\hbar\omega_1\gg 1$) (blue solid), Eq. (4.39), asymptotically approaches the perturbed value (green dotted). By contrast, the classical response ($\beta_1\hbar\omega_1\ll 1$) (purple dashed), Eq. (4.42) although proportional to the quantum response fails to reach the perturbed value of the observable A. Parameters are $\omega_1=2.4$, $\delta=10.1$, $\gamma=0.7$, $\lambda=2.3$, $\varepsilon=0.11$, $\beta_1=0.164$ and $\beta_2=0.416$.

$\mathscr{R}_3(\tau)$ does not.

For this response function three different response regimes may be distinguished which can be observed in the Figs. 4.1 and 4.2, that represent the linear response difference $\langle A\rangle_\varepsilon(t)-\langle A\rangle$ for various system parameters.

(i) In the thermal limit $\lambda\to 0$, the unperturbed quantum oscillator is in an equilibrium state and the quantum response function (4.39) vanishes (black dotted-dashed line). By contrast, the steady-state response is different from zero (blue solid line) and approaches the perturbed stationary value at large times (green dotted line). This example emphasizes the fundamental difference between equilibrium and steady-state quantum response theories.

Further, it can be shown that the steady-state response, where the baths are at differing temperatures $T_1\neq T_2$, may be different from zero in cases where the equilibrium response, where $T_1=T_2$ vanishes. For zero detuning, $\delta=0$, the perturbation commutes with the unperturbed Hamiltonian, $[H_0,H_I]=\hbar\omega_1[a_1^\dagger a_1+a_2^\dagger a_2+\lambda H_I, a_1^\dagger a_2+a_1 a_2^\dagger]=0$, implying that the response vanishes for a thermal state. However, in that limit the response function (4.39) reads,

$$\mathscr{R}_{\delta=0}(\tau)=e^{-\gamma\tau}\Delta N\gamma\frac{2\lambda\cos(2\lambda\tau)+\gamma\sin(2\lambda\tau)}{(\gamma^2+4\lambda^2)(\beta_1\hbar\omega_1)^{-1}}, \tag{4.40}$$

which is in general nonzero. The $\beta_1=\beta_2$ response thus vanishes for $\lambda\neq 0$, while the steady-state response is finite. In the free oscillator limit, $\lambda\to 0$, Eq. (4.40) vanishes al-

ways.

(ii) In the unitary limit $\gamma = 0$, when the interaction with the two heat reservoirs is switched off, the quantum response function (4.39) reduces to,

$$\mathscr{R}_{\text{unitary}}(\tau) = 2\lambda\Delta N\beta_1\hbar\omega_1\frac{\delta^2}{z^3}\sin z\tau. \tag{4.41}$$

We observe (Fig. 4.1) that the perturbed observable (red dashed line) exhibits in this situation oscillations with the same oscillation period as in the nonunitary case ($\gamma \neq 0$) (blue solid line). However, it never reaches its perturbed value (green dotted line) due to the absence of external damping.

(iii) Finally, in the classical limit, $\beta_j\hbar\omega_j \ll 1$, the Bose distribution reduces to the Boltzmann distribution and the response function (4.39) simplifies to,

$$\mathscr{R}_{\text{classical}}(\tau) = 2\lambda\frac{\beta_1\omega_1 - \beta_2\omega_2}{\beta_2\omega_2}\frac{\gamma(\delta^2 + 4\lambda^2\cos z\tau) + (\gamma^2+\delta^2)z\sin z\tau}{z^2(\gamma^2+z^2)}e^{-\gamma\tau} \tag{4.42}$$

The classical response function (4.42) is proportional to the quantum response function (4.39). However, it predicts the wrong perturbed value of the observable A (purple dotted line in Fig. 4.2), stressing the difference between classical and quantum response theories.

4.6.2. DETAILED BALANCE AND NONEQUILIBRIUM BEHAVIOR

We further show that the steady-state response function of the two-oscillator model does not satisfy the detailed balance condition $\langle AB(t)\rangle = \langle BA(t)\rangle$ for two time-reversal symmetric operators (see chapter 4.2.2 for details about detailed balance). Figure 4.3 exhibits the response function and its time reversed form,

$$\begin{aligned}\mathscr{R}_3(\tau) &= i\beta_1\hbar\omega_1\left\langle[a_1^\dagger a_2 + a_1 a_2^\dagger, a_1^\dagger a_1(\tau)]\right\rangle,\\ \mathscr{R}_3^R(\tau) &= i\beta_1\hbar\omega_1\left\langle[a_1^\dagger a_1, (a_1^\dagger a_2 + a_1 a_2^\dagger)(\tau)]\right\rangle,\end{aligned} \tag{4.43}$$

where it is used that the operators $A_{1,2} = a_1^\dagger a_2 + a_1 a_2^\dagger$, $a_1^\dagger a_1$ both have positive parity under time-reversal $\mu_{A_i} = 1$. We observe that detailed balance is violated, $\mathscr{R}_3(\tau) \neq \mathscr{R}_3^R(\tau)$, especially at short times, where an offset between the two curves is visible.

It is interesting to note, that in the case of $\lambda = 0$ the response (4.39) vanishes. This is actually the case for *any* Hamiltonian perturbation when one is interested in the time dependence of the oscillator energy observables, as can be seen from the third form (4.24) and one (4.16) directly. For $\lambda = 0$ the unperturbed QMS fulfills the quantum detailed balance since this corresponds to the free evolution of the two oscillators with their corresponding baths. From the time-reversal symmetry of the response (4.25) in the case of Hamiltonian perturbation, it then follows that, for any Hamiltonian perturbation H_I, the response for the observable A is the same as the response for the observable H_I under the Hamiltonian perturbation A, modulo a sign, $\mathscr{R}^A(\tau) = i\langle[A(\tau), H_I]\rangle = -\mu_A\mu_{H_I} i\langle[H_I(\tau), A]\rangle = -\mu_A\mu_{H_I}\mathscr{R}_3^{H_I}(\tau)$. But for $\lambda = 0$ the unperturbed steady-state is a direct product of thermal states of the two oscillators with their respective bath temperature. Therefore, using the first form (4.16) the Hamiltonian perturbation with respect

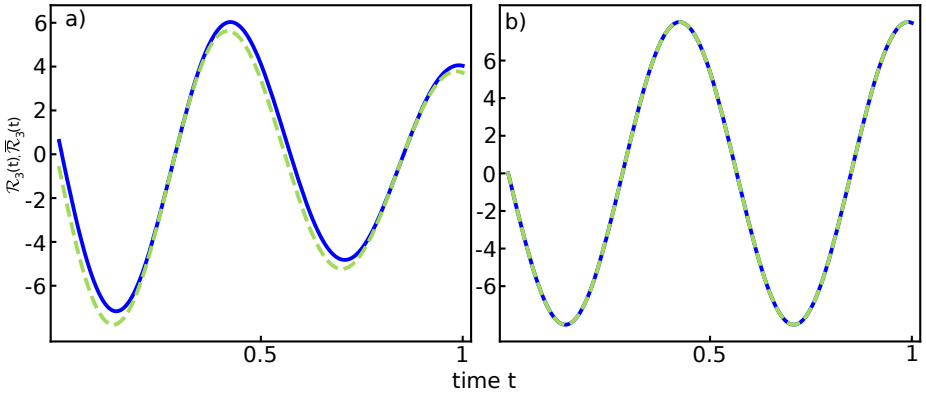

Figure 4.3: Violation of detailed balance in the two-oscillator model. a) The steady-state response $\mathscr{R}_3(\tau)$, in Eq. (4.43), (green dashed) is not equal to $\bar{\mathscr{R}}_3(\tau)$ in Eq. (4.43) (blue solid), especially at short times, as required by the detailed balance condition. b) check of detailed balance for the unitary case $\gamma = 0$. The detailed balance breaking in a) stems from the dissipative part of the dynamics.

to $H_I \propto A$ will commute with the unperturbed density operator $\mathscr{L}_1\pi_0 \propto [a_i^\dagger a_i, \pi_0] = 0$. Thus $\mathscr{R}_3^{H_I}(\tau) = 0$ has to vanish and therefore any Hamiltonian perturbation will have no effect on the unperturbed occupation number A.

4.7. BREAKDOWN OF LINEAR RESPONSE APPROACHING A CRITICAL POINT

To conclude this chapter, we will consider the limits of linear response theory. We will consider how the magnitude of a perturbation, in which the linear response is still accurate, behaves under different regimes of equilibrium and nonequilibrium situations. Further, we will consider if there is a difference between these regimes approaching a critical point. For this case, the position-position coupling Hamiltonian (2.2) for $\kappa = \lambda$ is being considered. The dynamics will be described by the global master equation (2.20) and the perturbation will be once more created by the intra-system coupling λ. Compared to the prior example using a local master equation (4.38), however, varying λ in the global case will also result in a modification of the dissipator $\mathscr{D}$. Linear response will thus contain not just a Hamiltonian part, but also a dissipative one. The linear response perturbation will be calculated by expanding the dissipator about some fixed $\lambda_{pert} = \lambda + \varepsilon$. The perturbation generator then has the form

$$\mathscr{L}_1(\rho) = -i[H_I, \rho] + \frac{\mathrm{d}}{\mathrm{d}\lambda_{\mathrm{pert}}}\mathscr{D}(\rho)\Big|_{\lambda_{pert}=\lambda}. \tag{4.44}$$

The adjoint dynamics of this problem can no longer be analytically calculated, but have to be computed numerically. In appendix 7.5, the method of calculating the adjoint dynamics numerically is shown. The perturbation itself will be chosen as an instantaneous shift $\varepsilon(t) = \Theta(t)\varepsilon$. The goal is to see the breakdown of the linear response. This will be accomplished by comparing the linear response at large (infinite) times

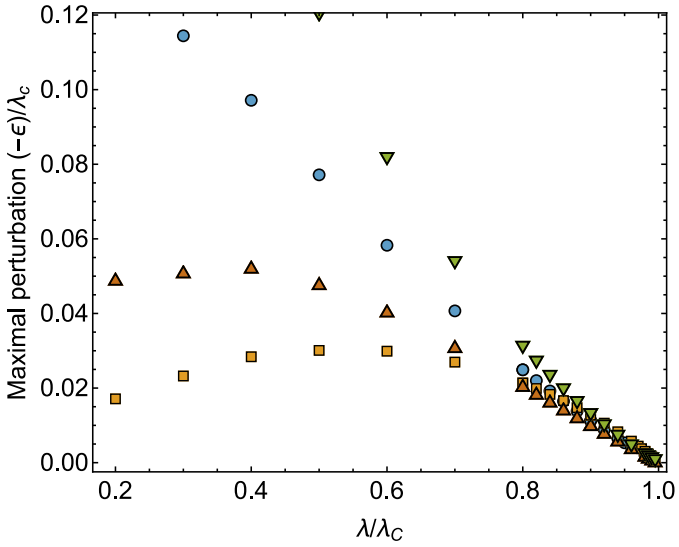

Figure 4.4: Comparison of the maximal linear perturbation strength $(-\varepsilon)/\lambda_c$ that creates an error less than δ_{err} in dependence of the system coupling λ/λ_c. The equilibrium case of finite temperature $T_i = 98$ (blue circles) possesses a larger valid linear regime compared to the equilibrium, vanishing temperature case $T_i = 0$ (orange squares) and small λ/λ_c. The nonequilibrium case of $T_1 = 20, T_2 = 98$ (brown triangles) interpolates between the two equilibrium cases as the decrease in T_1 dominates the linear regime size. The nonequilibrium case $T_1 = 98, T_2 = 20$ (inverted green triangles) on the other hand possesses a larger interval than the equilibrium case. Approaching the critical point, all regimes behave very similarly and all linear response regimes become infinitesimally small. Parameters are $\omega_1 = 5$, $\omega_2 = 2$, $\gamma_i = 1.5 \cdot 10^{-2}$.

$\langle a_1^\dagger a_1\rangle(t) = \langle a_1^\dagger a_1\rangle_0 + \int_0^t \varepsilon(s)\mathscr{R}(t-s)\mathrm{d}s$, where $\langle\cdot\rangle_0$ stands for the unperturbed system. On the other hand, solely changing the coupling strength λ_{pert}, the infinite time behavior is exactly known by calculating the steady-state behavior of the system at $\lambda + \varepsilon$, denoted $\langle a_1^\dagger a_1\rangle_\varepsilon$, using the global master equation's steady-states. Therefore, we will compare the linear response behavior with the correct result and interpret it as the failure of the linear response, if the relative error between the correct solution is larger than $\delta_{\text{err}} = 0.01$, i.e. larger than an 1% error,

$$\frac{|\langle a_1^\dagger a_1\rangle_\varepsilon - \langle a_1^\dagger a_1\rangle(\infty)|}{\langle a_1^\dagger a_1\rangle_\varepsilon} > \delta_{\text{err}}. \tag{4.45}$$

In Fig. 4.4 the breakdown of the linear response is shown for four different cases, the equilibrium finite temperature $T_i = 98$ (blue circles), the equilibrium vanishing temperature $T_i = 0$ (orange squares), the nonequilibrium $T_1 = 20$, $T_2 = 98$ (brown triangles) and the nonequilibrium $T_1 = 98$, $T_2 = 20$ case (green inverted triangles). To be precise, the maximally allowed perturbation $(-\varepsilon)/\lambda_c$ is plotted against the approach to the critical point λ/λ_c. Interestingly, the finite temperature linear response (blue circles) is generally valid in a larger perturbation interval than the vanishing temperature case. The nonequilibrium cases (triangles) possess either larger or smaller linear regimes than the equilibrium case, depending on the sign difference. The case where $T_1 = 20$ (brown triangles) possesses a smaller interval, closer to the $T_i = 0$ equilibrium case. This is ex-

pected, as the temperature of the first bath is lower and therefore interpolates between the two equilibrium regimes. Interestingly, the $T_2 = 20$ case shows actually a larger linear interval (green inverted triangles) as the corresponding equilibrium case. Thus, while nonequilibrium cases can cause a shrinking in the size of the linear interval in which the linear treatment is valid, it can also increase it for small λ/λ_c. After all, all that was checked was (4.45) and considering the nonequilibrium behavior of $\langle a_1^\dagger a_1 \rangle$ as plotted in 3.3 in chapter 3, the global approach for this strong nonequilibrium (left inset) hardly changes at smaller λ compared to close to equilibrium (left) as the energy flow to the colder mode first compensates the decrease of ω_-. Since $\langle a_1^\dagger a_1 \rangle$ hardly changes, also a larger perturbation can be applied before the linear regime becomes invalid, measured by (4.45). Approaching the critical point $\lambda/\lambda_c \approx 1$, however, one can observe that all the different approaches behave very similarly. All have a breakdown of the linear regime approaching the critical point with only small differences. This is also understandable since approaching the critical point results in a vanishing eigenfrequency $\omega_- \approx 0$ and therefore all finite temperatures will turn into a high-temperature (or quasi classical) limit.

4.8. DISCUSSION

We have generalized Kubo's quantum response theory to nonequilibrium open quantum systems described by quantum Markovian semigroups. We have, in particular, introduced a steady-state extension of the Kubo transformation, which plays a central role in the study of equilibrium isolated quantum systems. We have concretely derived quantum extensions of the equivalence classes for classical response functions introduced in Ref. [37]. We have for each of them analyzed the role of noncommuting operators and identified conditions under which the quantum response vanishes when some operators commute. We also considered the different properties these forms have depending on what kind of perturbations are applied, either Hamiltonian or dissipative. Then we considered what properties can be discerned from these classes in the case of QMS that fulfill quantum detailed balance. In particular we showed a symmetric time reversal symmetry for the second (4.22) and third (4.23) forms, in the case of quantum detailed balance. Next we have shown that the classes can be connected to various forms derived by other authors and the connection to classical response theory was further investigated. We have lastly illustrated our results with an analytically solvable model of two weakly coupled open quantum harmonic oscillators described by a local master equation and compared various response regimes including unitary/dissipative, equilibrium/steady-state, and classical/quantum limits. We have shown, in addition, that the equilibrium quantum response can vanish in instances where the steady-state quantum response does not. In the end we considered the breakdown of linear response approaching a critical point and showed that this is the case regardless whether one treats equilibrium and nonequilibrium regimes.

Our findings not only provide a unified picture of nonequilibrium quantum response theory, they also offer different, but equivalent, approaches to evaluate steady-state response functions, depending on the specific problem considered.

5

FLUCTUATION-DISSIPATION THEOREMS FOR QUANTUM MARKOVIAN SYSTEMS

5.1. INTRODUCTION

THE gist of linear response theory, as discussed in chapter 4, is that the properties of the (linear) response function is defined solely by the structure of the (unperturbed) system itself, independent of the exact time-dependence of the perturbation. This can be understood physically considering the following physical example. A Brownian particle [38] which is subject to random collisions from the surrounding gas will experience random, fluctuating, or nondirected forces. These do, however, generate also dissipative, or directed effects like friction. If the system is then subject to a small perturbation, as described by the linear response theory, it is not able to distinguish the origin of this perturbation. Therefore, its response has to be the determined by the free (unperturbed) dynamics of the system. Finding connections between these dissipative, or systematic, terms with fluctuating or random functions is what is generally termed a Fluctuation-dissipation theorem (FDT).

One such FDT [15, 25] is the connection between a function connected to the strength of the fluctuations D of a Langevin equation and its dissipative properties, caused by the interaction of a system with a thermal bath, $D = \gamma kT$, which is generally termed an Einstein relation [8]. Further, an Einstein relation for diffusion [25, 38, 110], that connects the diffusion constant with the mobility of e.g. electrons in a conductor is given by $D_0/\mu_0 = kT/e$ [141], where D_0 is the diffusion coefficient, μ_0 the mobility of the electrons and e the electric charge. One in particular interesting case of a fluctuation-dissipation theorem is the connection of response functions with specific correlation functions. In fact, the latter is a widely researched topic in both the classical [119, 142] and quantum [38–40, 110] context for both equilibrium [38, 110, 142] and nonequilibrium situations

[39, 40, 119].

Classically, the thermal fluctuation-dissipation theorem, for response functions $\mathscr{R}(t)$ and correlation functions $C^{xx}_{\mathrm{Cl}}(t)$, of a Brownian particle subject to a force can be found in Fourier space [142]. For this chapter, the notation of Fourier transforms will be given by $f(\omega) = \int_{-\infty}^{\infty} f(t)\mathrm{d}t$. The FDT for a classical, thermal, system is then given by [142]

$$C^{xx}_{\mathrm{Cl}}(\omega) = \frac{2kT}{\omega}\mathscr{R}''(\omega), \tag{5.1}$$

i.e. the Fourier transform of the position-position correlation function is connected to the imaginary part of the Fourier transformed response function, also called susceptibility $\mathscr{R}(\omega) = \mathscr{R}'(\omega) + i\mathscr{R}''(\omega)$ caused by the force. For the quantum case Kubo [25, 38, 110, 143] showed that for the Gibbs state and unitary dynamics, a similar FDT can be found. It was derived there that the imaginary part of the response function in Fourier space for general observables A, B (with positive time-reversal parity, which will be discussed in the following), here denoted as $\mathscr{R}_{AB}(\omega)$, is equal to the Fourier transform of a corresponding correlation function multiplied by a more complex term,

$$C^{AB}_{SC}(\omega) = \hbar\coth(\beta\hbar\omega/2)\mathscr{R}''_{AB}(\omega) = 2\hbar\left(N(\beta,\omega)+\frac{1}{2}\right)\mathscr{R}''_{AB}(\omega), \tag{5.2}$$

with the Bose-Einstein distribution $N(\beta,\omega) = 1/(e^{\hbar\beta\omega}-1)$. In the classical limit ($\hbar \to 0$) and $A, B = x$ this coincides with the classical form (5.1). It is interesting to note, that similar to the Callen Welton formula [109] for the radiative energy density, the quantum description contains a term corresponding to the zero point contribution $N(\beta,\omega)+\underline{1/2}$, showing that the quantum FDT (5.2) contains explicit quantum contributions.

Besides this zero point addition, the quantum setting contains further subtleties that have to be kept in mind. In quantum mechanics, operators do not commute, thus there is an ambiguity of how (two-time) correlation functions can be defined. Further, the FDT (5.2) is valid only for thermal steady-states $\pi_{eq} = \exp(-\beta H)/Z$. While many steady-states of different dynamics may be given in the form of (thermal) Gibbs states π_{eq}, this is not necessarily the case in general. Therefore, it is of interest whether it is possible to find such fluctuation-dissipation theorems for the cases of general nonequilibrium steady-states and in which way they differ from the thermal case [38]. Furthermore, (5.2) also only holds for closed (unitary) dynamics. For these, a thermal state is always their steady-state $[H,\pi_{eq}] = 0$. But since many physical problems have to be treated as fundamentally open, it is of further interest to consider how fluctuation-dissipation theorems look like for these more general cases.

The aim of this chapter is to find the general fluctuation-dissipation theorems for open quantum systems, using the general of quantum Markovian semigroups. In particular, the FDT's for the different response function classes in chapter 4 will be derived. Their structure will then be compared with classical nonequilibrium FDT'sl [119, 129], as well as open quantum attempts derived by various authors [40–42].

For being able to do so, some properties of open quantum mechanics and correlation functions have to be considered more closely. In particular we will start with rederiving the FDT (5.2) by Kubo [38]. We consider what properties the dynamics, response and correlation functions fulfill needed for deriving the FDT. This is then compared with the

properties the quantum Markovian semigroups possess and thus how the FDT's in the open context can be derived.

5.2. CLOSED QUANTUM DYNAMICS AND THE KUBO FDT

Fluctuation-dissipation theorems describe how (two-time) correlation functions are connected to response functions. Before deriving the Kubo FDT, it thus may be advantageous to first discuss how correlation functions are defined, what their properties are and how classical or quantum settings influence their structure.

5.2.1. CORRELATION FUNCTIONS IN THE QUANTUM CONTEXT

Classically, the formal definition of a correlation function [25] is given in the form

$$C^{AB}_{\text{class}}(t,t') = \langle A(t)B(t')\rangle, \tag{5.3}$$

for arbitrary dynamical variables A, B. The expectation value $\langle\cdot\rangle$ is connected to some kind of averaging. There are various definitions for how to define the averaging procedure of two variables [25]. One physically intuitive approach is defining the average by measuring the variables A, B at their respective times, and averaging their product over the number of measurements $C^{AB}_{\text{class}}(t,t') = \lim_{N\to\infty} 1/N \sum_{k=1}^{N} A_k(t)B_k(t')$. A more thermodynamic definition, for ergodic systems, is given by an ensemble average about a distribution function. For an example of a Fokker-Planck equation, this ensemble average may be defined as $C^{AB}_{\text{class}}(t,t') = \int A(X,t)B(X,t')p(X)\mathrm{d}X$, where X are the phase space variables of the system and $p(X)$ is the probability distribution function. In the case of steady-states $p(X) = p_{\text{st}}(X)$, these correlation functions fulfill the so called stationarity property $\langle A(t)B(t')\rangle = \langle A(t-t')B\rangle$ [25].

In the quantum setting, these definitions cannot be straightforwardly generalized. First, operators will in general not commute, thus (5.3) is not necessarily uniquely defined. Second, in the classical picture, the meaning of these correlation functions can be understood physically as the subsequent measurement of the corresponding variables A, B. This is not necessarily possible in the quantum case. The expectation value $\langle A(t)B(t')\rangle$ itself does not possess a direct physical meaning as in the classical case, i.e. it is not a direct measurement of B at time t', followed by a measurement of A at time t. It is only possible to connect these correlation functions mathematically to e.g. the measurements of the system response considered in chapter 4 or an intricate summation of multi-measurement setups (see e.g. p 35 in [15]). But the correlation functions themselves are not measurable, due to the simple fact, that for two Hermitian operators A, B, the correlation functions are complex $\langle A(t)B(t')\rangle^* = \langle B(t')A(t)\rangle \neq \langle A(t)B(t')\rangle$. While they can be only measured indirectly [15], these correlation functions do contain important mathematical properties of the system's dynamics.

The direct quantum generalization of the classical formula, for closed dynamics, is then given by [25, 38, 110]

$$C^{AB}_{LC}(t,t') = \langle A(t)B(t')\rangle = \mathrm{Tr}\{A(t)B(t')\rho\}, \tag{5.4}$$

where $A(t), B(t')$ are Heisenberg operators fulfilling $\mathrm{d}_t A(t) = i[H,A]$. As mentioned above, this function is in general complex. A real version can be obtained by decomposing the

complex function into a real and imaginary part $f = \mathrm{Re}(f) + i\mathrm{Im}(f)$. For two Hermitian operators A, B, the real or symmetric correlation function can be defined as

$$C_{SC}^{AB}(t,t') = \langle\{A(t), B(t')\}\rangle = C_{LC}^{AB}(t,t') + C_{LC}^{AB}(t,t')^*. \tag{5.5}$$

Let us now consider some properties of these correlation functions. Upon using time-reversal [144], there exists a physical interpretation of the symmetric form (5.5). The symmetric correlation function can be cast into a sum of a correlation function of type (5.4) and its time reversed version (see appendix 7.8 for details on time-reversal of closed quantum dynamics). Thus, the symmetric correlation function (5.5) can be written as

$$C_{SC}^{AB}(t,t') = \langle A(t)B(t')\rangle + \langle A^R(t)B^R(t')\rangle_R = (C_{LC}^{AB}(t,t') + C_{LC}^{AB}(t,t')^*), \tag{5.6}$$

where $A^R = \Theta A^\dagger \Theta$ corresponds to the time-reversal of an operator, with the antiunitary time-reversal operator $\Theta = \Theta^{-1} = \Theta^\dagger$ [144] and $\langle\cdot\rangle_R$ is the expectation value w.r.t. the time reversed density operator ρ^R. Therefore, the dynamical properties of the symmetric correlation functions are connected to the properties of time-reversal. For thermal states $\rho = \pi_{eq}$ and unitary dynamics, one can further derive additional interesting properties for these correlation functions. First, the correlation functions (5.4), (5.5) have the stationarity property $C_{iC}^{AB}(t,t') = C_{iC}^{AB}(t-t',0)$, $i = L, C$. This can be shown directly, using the fact that the equilibrium state commutes with the time evolution operator $[U(t), \pi_{eq}] = 0$ and thus

$$\begin{aligned}\langle A(t)B(t')\rangle &= \mathrm{Tr}\{U^\dagger(t)AU(t)U^\dagger(t')BU(t')\pi_{eq}\} = \mathrm{Tr}\{U(t')U^\dagger(t)AU(t)U^\dagger(t')B\pi_{eq}\} \\ &= \langle A(t-t')B\rangle.\end{aligned} \tag{5.7}$$

Further, the time-reversal operation can be interpreted as the evolution in negative time for an observable with well defined time-reversal parity μ_A, $A^R(t) = \Theta U^\dagger(t)A^\dagger U(t)\Theta = U^\dagger(-t)\Theta A\Theta U(-t) = \mu_A A(-t)$, where for the time evolution operator it is the case that $\Theta U(t)\Theta = \Theta\exp(-iHt)\Theta = \exp(iHt) = U(-t)$. Lastly one can find that $\pi_{eq}^R = \pi_{eq}$, i.e. the equilibrium state is invariant under time-reversal. Using these properties, one can derive a form of detailed balance in the quantum context [115] for closed dynamics. This states that for two Hermitian operators A, B one has

$$\langle A(t)B\rangle = \langle \Theta B A(t)\Theta^{-1}\rangle = \mu_A\mu_B\langle BA(-t)\rangle = \mu_A\mu_B\langle B(t)A\rangle. \tag{5.8}$$

From (5.8) it then follows, that the symmetric correlation function between A and B is equal to the correlation function between B and A up to the time-reversal parity, which upon using the stationarity property results in $C_{SC}^{AB}(t-t',0) = \mu_A\mu_B C_{SC}^{BA}(t-t',0)$. Further, one can use the group property and observe the properties of the negative time correlation function for the detailed balance case

$$C_{SC}^{AB}(-t,0) = C_{SC}^{BA}(t,0) \overset{(5.8)}{=} \mu_A\mu_B C_{SC}^{AB}(t,0). \tag{5.9}$$

Thus, depending on the time-reversal parities μ_A, μ_B, one can deduce from (5.9) that the symmetric correlation function is either a symmetric or antisymmetric function.

The imaginary part of a complex function can be considered in general independent of the real part. However, the imaginary part of the correlation function (5.4) is actually

not independent of (5.5). In fact, there are two kinds of connections that were established for these parts. First, there are the so called Kramers and Kronig dispersion relations which establish a general connection for a quadratic integrable function in Fourier space using complex calculus [143],

$$C(\omega) = \int_{-\infty}^{\infty} \frac{C(u)}{u-\omega} \mathrm{d}u. \tag{5.10}$$

Second there are the fluctuation-dissipation theorems, connecting the correlation function (5.4) to the response function of the closed, thermal response theory [38]. The latter we will consider more closely next.

5.2.2. THE FLUCTUATION-DISSIPATION THEOREM FOR CLOSED, THERMAL DYNAMICS

For the symmetric correlation function (5.5), the fluctuation-dissipation theorem (5.2) was derived by various authors [25, 38, 110, 143]. Response theory for closed dynamics can be obtained from the classes discussed in 4, by considering Hamiltonian perturbation of the form $\mathscr{L}_1(\cdot) = -i[B,\cdot]$ and taking the limit of vanishing system-bath coupling $\gamma_i = 0$. Utilizing the third class (4.24) in this closed dynamics limit, the response function for any observable A can then be written as a difference of two correlation function of the form (5.4),

$$\mathscr{R}_C^{AB}(\tau) = -i\left(C_{LC}^{AB}(\tau,0) - C_{LC}^{AB*}(\tau,0)\right). \tag{5.11}$$

It is interesting to note, that the response function is exactly given by the imaginary part of the correlation function (5.4). Therefore, searching for a FDT between the symmetric correlation function (5.6) and the response function (5.11) is just given by a FDT between the real and imaginary part of the correlation function (5.4). It is also noteworthy, that the response (5.11) and correlation function (5.5) are well defined for all $t \in \mathbb{R}$. In particular, these possess a symmetry about $\tau = 0$ for stationary states. The response function for $-\tau$ is given by

$$\begin{aligned}\mathscr{R}_C^{AB}(-\tau) &= -i\left(C_{LC}^{AB}(-\tau,0) - C_{LC}^{AB*}(-\tau,0)\right) = -i\left(C_{LC}^{AB}(0,\tau) - C_{LC}^{AB*}(0,\tau)\right)\\ &= i\left(C_{LC}^{BA}(\tau,0) - C_{LC}^{BA*}(\tau,0)\right) = -\mathscr{R}_C^{BA}(\tau) = -\mu_A\mu_B\mathscr{R}_C^{AB}(\tau)\end{aligned} \tag{5.12}$$

and therefore the response functions have a time-inversion symmetry that is negative (positive), when the the symmetric correlation function (5.9) has a positive (negative) symmetry. With being defined for all $t \in \mathbb{R}$, it is possible to find a FDT in Fourier space using the so called Kubo-Martin-Schwinger (KMS) boundary condition [143, 145–148]. The KMS boundary condition states that

$$\int_{-\infty}^{\infty} \langle BV(t)A\rangle f(t)\mathrm{d}t = \int_{-\infty}^{\infty} \langle (V(t)A)B\rangle f(t+i\beta)\mathrm{d}t, \tag{5.13}$$

for a continuous function $f(t)$ on $\mathbb{C}$, a general one-parameter group $\{V(t)|t \in \mathbb{R}\}$ (see also discussion in chapter 4.2.1 about group properties) and the group's steady-state being the Gibbs state $\rho = \pi_{eq}$. In the closed (unitary) case, the group is given by $V(t)A = U^\dagger(t)AU(t)$, which is continuous on $t \in \mathbb{R}$. The idea behind the KMS boundary condition

is the observation that one can define an imaginary time evolution for thermal unitary evolution in the following way

$$\langle BA(t)\rangle = \langle \rho_{eq}^{-1} A(t)\rho_{eq} B\rangle = \langle A(t-i\beta)B\rangle. \tag{5.14}$$

This KMS identity utilizes the fact that for thermal states $\pi_{eq} = e^{-\beta H}/Z$ one can identify the products of density operators A, $\pi_{eq}A(t)\pi_{eq}^{-1} = e^{-\beta H}U^\dagger(t)AU(t)e^{\beta H} = U^\dagger(t-i\beta)AU(t-i\beta)$, with an imaginary evolution since the time evolution operator $U(t) = e^{-iHt}$ in the closed dynamics case is also fully defined by the Hamiltonian H.

The KMS boundary condition now states that the Fourier transform of the correlation function can be transformed in the following way

$$\begin{aligned} C_{LC}^{AB*}(\omega) &= \int_{-\infty}^{\infty} C_{LC}^{AB*}(\tau,0)e^{i\omega\tau}\mathrm{d}\tau = \int_{-\infty}^{\infty}\langle BA(t)\rangle e^{i\omega\tau}\mathrm{d}\tau \\ &= \int_{-\infty}^{\infty}\langle A(t-i\beta)B\rangle e^{i\omega\tau}\mathrm{d}\tau \overset{\text{KMS}}{=} \int_{-\infty}^{\infty}\langle A(t)B\rangle e^{i\omega(\tau+i\beta)}\mathrm{d}\tau \\ &= e^{-\beta\omega}C_{LC}^{AB}(\omega). \end{aligned} \tag{5.15}$$

This allows one to identify the response function with the one-sided correlation function $C_{LC}^{AB}(\tau,0)$,

$$\mathscr{R}_C^{AB}(\omega) = -i(1-e^{-\beta\omega})C_{LC}^{AB}(\omega). \tag{5.16}$$

Further, the symmetric correlation function can be also identified, using (5.15), with $C_{SC}^{AB}(\omega) = (1+e^{-\beta\omega})C_{LC}^{AB}(\omega)$. Therefore, the FDT between the linear response and the symmetric correlation function can be given by

$$\mathscr{R}_C^{AB}(\omega) = -i\frac{1-e^{-\beta\omega}}{1+e^{-\beta\omega}}C_{SC}^{AB}(\omega) = -i\tanh(\beta\omega/2)C_{SC}^{AB}(\omega). \tag{5.17}$$

Next, let us consider the real and imaginary parts $\mathscr{R}_C^{AB}(\omega) = \mathscr{R}_C^{AB\prime}(\omega) - i\mathscr{R}_C^{AB\prime\prime}(\omega)$ of the susceptibility

$$\begin{aligned} \mathscr{R}_C^{AB\prime}(\omega) &= (\mathscr{R}_C^{AB}(\omega)+\mathscr{R}_C^{AB}(\omega)^*)/2 = -i\tanh(\beta\omega/2)\left(C_{SC}^{AB}(\omega)-C_{SC}^{AB}(-\omega)\right)/2 \\ \mathscr{R}_C^{AB\prime\prime}(\omega) &= i(\mathscr{R}_C^{AB}(\omega)-\mathscr{R}_C^{AB}(\omega)^*)/2 = \tanh(\beta\omega/2)\left(C_{SC}^{AB}(\omega)+C_{SC}^{AB}(-\omega)\right)/2. \end{aligned} \tag{5.18}$$

On using quantum detailed balance (5.8) and stationarity, one can simplify these terms into the full FDT for closed quantum dynamics

$$\begin{aligned} \mathscr{R}_C^{AB\prime}(\omega) &= -i\tanh(\beta\omega/2)\left(C_{SC}^{AB}(\omega)-C_{SC}^{BA}(\omega)\right)/2 \\ &= -i(1-\mu_A\mu_B)\tanh(\beta\omega/2)C_{SC}^{AB}(\omega)/2 \\ \mathscr{R}_C^{AB\prime\prime}(\omega) &= \tanh(\beta\omega/2)\left(C_{SC}^{AB}(\omega)+C_{SC}^{BA}(\omega)\right)/2 \\ &= (1+\mu_A\mu_B)\tanh(\beta\omega/2)C_{SC}^{AB}(\omega)/2. \end{aligned} \tag{5.19}$$

This reproduces the FDT derived by Kubo [38] and one can obtain (5.2), if $\hbar = 1$ is set and if both observables have the same parity $\mu_A = \mu_B$. Then it is also the case that the real part of the susceptibility vanishes $\mathscr{R}_C^{AB\prime}(\omega) = 0$. For differing parities $\mu_A \neq \mu_B$, the

imaginary part of the susceptibility vanishes $\mathscr{R}_C^{AB\prime\prime}(\omega) = 0$ and the FDT exists in this case only for the real part of the susceptibility.

It has to be noted, that for closed dynamics, the correlation functions $\langle A(t)B\rangle$ are in general nonvanishing for large times as no dissipative effects can cause a suppression of the correlations. However, a thermodynamic limit is assumed, allowing the system to have vanishing correlations for large times. Lastly, let us mention the Kramers-Kronig relation for the response function as derived in e.g. [143], which has the form

$$\begin{aligned}\mathscr{R}_C^{AB\prime}(\omega) &= \frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{\mathscr{R}_C^{AB\prime\prime}(\omega')}{\omega'-\omega}\mathrm{d}\omega' \\ \mathscr{R}_C^{AB\prime\prime}(\omega) &= -\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{\mathscr{R}_C^{AB\prime}(\omega')}{\omega'-\omega}\mathrm{d}\omega',\end{aligned} \tag{5.20}$$

with the principal value integral $\mathrm{P}\int \mathrm{d}\tau$, connecting the imaginary and real part with each other. Comparing the symmetries in (5.19) with (5.20), one can find that for operators with well defined parities μ_A, μ_B, (5.20) cannot hold, since either the real or imaginary part vanishes. Only for operators $A^R \neq \mu_A A$ (5.20) connects two finite functions.

5.3. QUANTUM MARKOVIAN SYSTEMS AND THEIR FDT'S

Considering the derivation of the closed dynamic's FDT (5.19), one observes that a multitude of properties of the closed quantum dynamics have to be used, e.g. the group property defining the correlation functions (5.4) for all times $t \in \mathbb{R}$ or the system Hamiltonian defining the complete unperturbed dynamics and the perturbation being also defined solely by a unitary operator. Also symmetry properties were of the essence, like the time-reversal symmetry.

But these used properties do not necessarily hold in the open quantum context. For one, a KMS condition of the form (5.13) cannot be found in the open context. The dynamics of an open system is not completely determined by the system's Hamiltonian and thus even a thermal steady-state density operator π_{eq} only accounts for the 0-eigenvalue eigenoperator of the generator $\mathscr{L}\pi_{eq} = 0$. But there are further dynamical properties caused by the bath structure and system-bath interaction [42]. The dynamical maps of an open system also only consistute to a semigroup [42, 132] and thus negative times have to be treated very cautiously. Due to these potential complications, it is of the essence to first consider more closely what property changes one has in the case of open quantum mechanics and how it is still possible to treat FDT's in the open quantum context of quantum Markovian semigroups in a mathematically consistent way. We start by once more considering first the essential building blocks, the correlation functions. For these it is assumed that the quantum regression theorem [25] holds (see (4.2.1) for details about quantum regression).

5.3.1. CORRELATION FUNCTIONS IN OPEN SETTINGS

Going from the classical interpretation of a correlation function (5.3) to the closed quantum dynamic's version (5.4) a number of ambiguities were introduced, as is usually the case for generalizations. Open quantum mechanics may also be treated mathematically as a generalization of closed dynamics, creating yet again new ambiguities. One such

ambiguity is caused by the fact, that the dynamical map of the open quantum system no longer constitutes to a unitary group, but a semigroup (a more detailed discussion of semigroups is given in chapter 4.2.1 or in [20]). In particular, the map $e^{\mathscr{L}t}$ cannot be written as a product of unitary operators. Therefore, one cannot reduce the time evolution of two operators $e^{\overline{\mathscr{L}}t}(AB) \neq e^{\overline{\mathscr{L}}t}(A)e^{\overline{\mathscr{L}}t}(B)$, where the generator $\overline{\mathscr{L}}$ is the adjoint super operator as defined in (4.2).

This has significant implications on the possible structures of the correlation functions, creating multiple possible definitions for them. For one, in the closed stationary case this reduction ability was used to derive the stationarity property $\mathrm{Tr}\{A(t)B(t')\pi\} = \mathrm{Tr}\{A(t-t')B\pi(t')\} = \mathrm{Tr}\{A(t-t')B\pi\}$. But these two forms are not equal in the open case. This can be seen by writing the time evolution explicitly

$$\mathrm{Tr}\left\{e^{\overline{\mathscr{L}}t}(A)e^{\overline{\mathscr{L}}t'}(B)\pi\right\} \neq \mathrm{Tr}\left\{e^{\overline{\mathscr{L}}(t-t')}(A)Be^{\mathscr{L}t}(\pi)\right\} = \mathrm{Tr}\left\{e^{\overline{\mathscr{L}}(t-t')}(A)B\pi\right\}, \tag{5.21}$$

even for thermal steady-states $\pi = \pi_{eq}$. Following e.g. [6], the plausible choice of a correlation function in the open case is the one that at least formally respects the property of stationarity $\langle A(t)B(t')\rangle = \langle A(t-t')B\rangle$. The left function in (5.21) cannot be interpreted w.r.t. stationarity. Both adjoint evolutions of A, B start at $t, t' = 0$ and even if $t = t'$ one can observe a dynamic behavior for different t. A consistent definition that takes stationarity into account is then given by

$$C^{AB}_{LO}(t,t') = \langle A(t)B(t')\rangle := \mathrm{Tr}\left\{Ae^{\mathscr{L}(t-t')}\left(Be^{\mathscr{L}t'}(\rho)\right)\right\} = \mathrm{Tr}\left\{e^{\overline{\mathscr{L}}(t-t')}(A)Be^{\mathscr{L}t'}(\rho)\right\}, \tag{5.22}$$

where for the last equation the quantum regression theorem is used (see 4.2.1). This can be interpreted [6] as first the density matrix ρ is being evolved from 0 to time t'. Then the operator B is multiplied to the left and this new operator $Be^{\mathscr{L}t'}(\rho)$ is evolved from t' to t and finally the operator A is being multiplied. For a stationary state the first time evolution acts trivially on the stationary state $e^{\mathscr{L}t}\pi = \pi$ and the last term in (5.22) can be simplified where the dynamical behavior is solely defined by the adjoint dynamics of the operator A, $C^{AB}_{LO}(t,t') = \langle A(t-t')B\rangle = \langle A(t)B(t')\rangle$. Defining the correlation function in that way (5.22), one can reproduce formally the stationarity property. Further, this is exactly the form of correlation functions that occur in the response functions of the open dynamics linear response theory (see chapter 4), justifying this choice.

Let us consider now what properties the open dynamic's correlation function (5.22) possesses. As in the case of closed dynamics, the correlation function (5.22) is a complex function. A real version can be once again achieved by symmetrization

$$C^{AB}_{SO}(t,t') = C^{AB}_{LO}(t,t') + C^{AB}_{LO}(t,t')^*. \tag{5.23}$$

Writing the complex conjugation of the correlation function explicitly $C^{AB}_{LO}(t,t')^* = \langle A(t)B(t')\rangle^* = \mathrm{Tr}\left\{Be^{\overline{\mathscr{L}}(t-t')}(A)e^{\mathscr{L}t'}(\rho)\right\}$ one can observe that the above definition of the symmetric correlation function (5.23) is not the sum of two correlation function of the form (5.22). Thus, for being able to reproduce the stationarity property, this symmetry property of the correlation function is sacrificed in the open context. While the second function in (5.23) may not be a direct correlation function, using time-reversal enables one to interpret both terms in (5.23) as correlation functions. Using the time reversed

generator of the adjoint dynamics $\overline{\mathscr{L}}^R$ (see appendix 7.8), the complex conjugate of the correlation function (5.22) can be rewritten as

$$C_{LO}^{AB}(t,t')^* = \mathrm{Tr}\left\{B e^{\overline{\mathscr{L}}(t-t')}(A)\pi\right\} = \mathrm{Tr}\left\{e^{\overline{\mathscr{L}}^R(t-t')}(A^R)B^R\rho^R\right\} = \left\langle A^R(t-t')B^R\right\rangle_R, \quad (5.24)$$

where $A^R(t) := e^{\overline{\mathscr{L}}^R t}A^R$ is given by the time reversed adjoint dynamics. Thus, the symmetric correlation function can be interpreted, like in the closed case (5.6), as a sum of the correlation function (5.22) and its time-reversed version

$$C_{SO}^{AB}(t,t') = \langle A(t-t')B\rangle + \left\langle A^R(t-t')B^R\right\rangle_R. \quad (5.25)$$

The time-reversal operation (7.24) can further be used, in conjunction with the 0-dual semigroup $\overline{\mathscr{L}}^0$, to generalize the quantum detailed balance condition as was done in 4.2.2. It should be stressed, however, that in the closed dynamics case this 0-dual semigroup is just given by $\langle A(-t)B\rangle = \langle AB(t)\rangle$, i.e. the 0-dual is necessary because the open dynamics is only a semigroup and not a unitary group. Thus, the defining negative times for the correlation functions (5.23) and (5.22) is a nontrivial task. Interestingly, Weidlich [42] put forward a heuristic argument extending the correlation function (5.25) to negative times by defining just the negative time behavior as it is in the closed dynamic's case, $C_{SO}^{AB}(-\tau,0) := C_{SO}^{BA}(\tau,0)$, $\tau > 0$. This transforms the correlation functions into C_0 functions for all $t \in \mathbb{R}$, which coincides with the closed dynamic's definition in the limit of vanishing system-bath coupling. This definition is advantageous in the sense, that for these correlation functions it is possible to use the normal Fourier transform as in the closed case. However, this definition also does not respect the time-irreversibility of the open dynamics. It is therefore rather a mathematical trick than a physical property. Due to this ambiguity, we will not continue the correlation functions to negative times, but rather use one-sided Fourier transforms as in [41].

5.3.2. FLUCTUATION-DISSIPATION THEOREMS OF QUANTUM MARKOVIAN SYSTEMS

In this subsection we will reconsider the various response formulas derived in chapter 4 w.r.t. to the possibility of finding a fluctuation-dissipation theorem.

FDT'S FROM THE THIRD CLASS

Starting with the third form (4.24), the response function is given by $\mathscr{R}_3(\tau) = \left\langle \overline{\mathscr{L}}_1 A(\tau)\right\rangle$. For the case of purely Hamiltonian perturbation, the response has the form $\mathscr{R}_3^{AB}(\tau) = -i\langle[A(t),B]\rangle$. Formally, this completely resembles the response function of the closed case (5.11). However, due to the different properties of the correlation function of the open dynamics case considered above 5.3.1, it is not possible to apply the KMS boundary condition (5.15). This is also something that is expected, in the open case the dynamics are governed by the generator $\mathscr{L}$, whose structure is not completely defined by the steady-state π even in the thermal case. Thus, it should be expected that for any fluctuation-dissipation theorem the eigensystem of the generator $\mathscr{L}$ has to be accounted for in the open case. Interestingly, Weidlich [42] used an eigenoperator decompositions

of the generators $\mathscr{L}$, $\overline{\mathscr{L}}$ to derive a FDT for this Hamiltonian perturbation. The derived FDT is of the form [42]

$$C_{SO}^{AB}(\omega) = i(R_{A\mathscr{B}}(\omega) - R_{\mathscr{A}B}(\omega)) + \mathrm{P}\int_{-\infty}^{\infty} \frac{1}{\omega}(R_{A\mathscr{B}}(\omega) + R_{\mathscr{A}B}(\omega))\mathrm{d}\omega, \tag{5.26}$$

where the operators $\mathscr{A}, \mathscr{B}$ use special eigenoperator projections of the generators (see [42] for more details) and P denotes the principal value integral. This shows, that for general open quantum systems, the FDT for the same type of response as for the closed dynamics' case (5.18), is significantly more complex in form.

While one could in principle also apply these eigenoperator decompositions to all the other classes, we will follow a different approach. In particular, we relax the condition that the correlation function has to be a product of two observables $\langle A(t)B\rangle$, but allow that the operator B can be non Hermitian. This is consistent insofar, as the first (4.16) and second form 4.21 are exactly such correlation functions. The third form (4.24) for commutative perturbation then can be rewritten as such correlation functions $\mathscr{R}_3(\tau) = \langle A(\tau)B_1'\rangle + \langle B_1'^{\dagger}A(\tau)\rangle$, with $B_1' = iH_I$, where without loss of generality only one perturbation is assumed. However, this is exactly a symmetric correlation function $\mathscr{R}_3^n(\tau) = C_{SO}^{AB_1'}(\tau)$. The FDT for this case is then given in Fourier space by

$$R(\omega) = C_{SO}^{AB_1'}(\omega) \tag{5.27}$$

Next, the case of a purely dissipative perturbation in the general form [20] $\overline{\mathscr{L}}_1(A) = \overline{\mathscr{D}}_I(A) = \sum_n \Gamma_n(B_nAB_n^{\dagger} - \{B_nB_n^{\dagger}, A\}/2)$ for a set of operators $\{B_n\}$ is being considered. In this case, the response function can even no longer be written as the combination of two correlation functions of type (5.22) for observables. However, one is able to find the non Hermitian operators C_n such that

$$\mathscr{R}_3(\tau) = \sum_n \Gamma_n(\langle A(\tau)C_n\rangle + \langle C_n^{\dagger}A(\tau)\rangle) = \sum_n \Gamma_n C_{SO}^{AC_n}(\tau). \tag{5.28}$$

These operators have the form $C_n = (B_n^{\dagger}\pi B_n\pi^{-1} - B_nB_n^{\dagger})/2$. (5.28) is then also a sum of symmetric correlation functions, and thus the FDT has the form

$$R(\omega) = \sum_n \Gamma_n C_{SO}^{AC_n}(\omega). \tag{5.29}$$

This form is similar in notion to a classical NESS FDT by Agarwal [115] for Fokker-Planck dynamics. There it was shown that for detailed balance dynamics it is the case that the response can be written in the form $R(\omega) = \sum_j (D^{-1})_{ij}R_{ij}(\omega)$, where D is the diffusion matrix. For the general case of dissipative + Hamiltonian perturbation one can also find a non Hermitian operator, using the decomposition of the dissipative form, $D_n = iH_I + C_n$.

FDT'S USING THE FIRST AND SECOND FORMS

Compared to the third form, the first (4.16) and second forms (4.17),(4.20) are asymmetric correlation functions of type (5.4). Furthermore, these are directly a product of the Hermitian observable A and a non Hermitian operator B.

For the first form $\mathscr{R}_1(\tau) = \langle A(\tau)B_1\rangle$, the reality of the response function means that $\langle A(\tau)B_1\rangle = \langle A(\tau)B_1\rangle^* = \langle B_1^\dagger A(\tau)\rangle$. Thus, the response function can be connected to the symmetric correlation function in the form

$$\mathscr{R}_1(\tau) = \left(\langle A(\tau)B_1\rangle + \langle B_1^\dagger A(\tau)\rangle\right)/2 = C_{SO}^{AB_1}(t)/2. \tag{5.30}$$

The FDT then is once more straight forward

$$\mathscr{R}_1(\omega) = C_{SO}^{AB_1}(\omega)/2. \tag{5.31}$$

This form is very similar to the one derived for purely dissipative perturbation (5.29). The difference is, that $B_1 = (\mathscr{L}_1\pi_0)/\pi_0$ hides all the system-bath interactions within B_1. The second form (4.21) is given by $\mathscr{R}_2(\tau) = -\mathrm{d}_\tau\langle A(\tau)\overline{\partial_\varepsilon S_\varepsilon|_0}\rangle = \mathrm{d}_\tau\langle AB_2\rangle$. Using the same argument of reality of the response function, the response can be written in dependence of a correlation function,

$$\mathscr{R}_2(\tau) = \mathrm{d}_\tau C_{SO}^{AB_2}(\tau)/2. \tag{5.32}$$

For the FDT, it is important to emphasize that the response and correlation functions are only defined for $t \geq 0$. This results in the fact, that in the partial integration in the (one-sided) Fourier transform of (5.32) a boundary term remains

$$\begin{aligned}\mathscr{R}_2(\omega) = \int_0^\infty \mathscr{R}_2(\tau)e^{i\omega\tau}\mathrm{d}\tau = \int_0^\infty e^{i\omega\tau}\frac{\mathrm{d}}{\mathrm{d}\tau}C_{SO}^{AB_2}(\tau,0)\mathrm{d}\tau = \left[e^{i\omega\tau}\frac{\mathrm{d}}{\mathrm{d}\tau}C_{SO}^{AB_2}(\tau,0)\right]_0^\infty \\ - i\omega\int_0^\infty C_{SO}^{AB_2}(\tau,0)e^{i\omega\tau}\mathrm{d}\tau = C_{SO}^{AB_2}(0,0) - i\omega C_{SO}^{AB_2}(\omega).\end{aligned} \tag{5.33}$$

From this follows the FDT, by noting that because of the reality of the response function one can deduce $\mathscr{R}_2(\omega) - \mathscr{R}_2(-\omega) = \mathscr{R}_2(\omega) - \mathscr{R}_2(\omega)^* = -i2\mathscr{R}_2''(\omega)$,

$$\mathscr{R}_2''(\omega) = \omega C_{SO}^{AB_2}(\omega). \tag{5.34}$$

This form of the frequency FDT is similar in form to a classical NESS FDT by Prost et al. [119]. In the classical case, they showed that $R_{\alpha\gamma}(\omega) - R_{\gamma\alpha}(-\omega) = i\omega C_{\alpha\gamma}(\omega)$ and thus the difference of susceptibilities is given by a correlation function.

5.4. DISCUSSION

The differences between classical, closed quantum and open quantum systems were considered w.r.t. correlation functions as the building blocks of fluctuation-dissipation theorems. Rederiving the closed quantum FDT by Kubo [38], it was emphasized that the form (5.2) is a consequence of the properties of the closed, thermal dynamics. In the open case of quantum Markovian semigroups it was first shown that for these many properties are not reproduced. It is therefore expected, that FDT's for such systems will not be necessarily be as elusive as (5.2). It is the case, that for open quantum systems, the dissipative part of the dynamics is determined by the bath properties and system-bath interaction. Therefore, these have to either be contained within the connection between the susceptibility $R(\omega)$ and the symmetric correlation function $C_S(\omega)$ or hidden in the operators within these functions. We showed then that for the classes found in chapter

4, there can be found a FDT for all of them, which are simple in form, by storing the system-bath interaction in non Hermitian operators. For purely dissipative perturbation the FDT for the third form was shown to be a sum of the friction coefficients (5.29). While the second form's FDT has a trivial connection between the susceptibility and a correlation function (5.31), the second form's FDT connects the imaginary part of the susceptibility to a correlation function times ω (5.34).

6

CONCLUSION AND OUTLOOK

We investigated different aspects of quantum Markovian master equations. First different perturbative methods were analyzed in their ability to correctly describe the steady-state properties of a model which is simple enough to be described by global master equations, yet sufficiently complex model that it still expressed nontrivial behavior. Second the dynamical properties were investigated by deriving response function classes and fluctuation-dissipation theorems.

In chapter 2 global and local master equation were compared in their ability to describe critical phenomena. In particular, the steady state properties of two coupled harmonic oscillators each coupled to their respective baths was considered. It was shown that in the equilibrium case the global approach is able to correctly reproduce the Gibbs distribution for both considered intra-system coupling models. The local approach, on the other, hand had difficulties to quantitatively reproduce the expected equilibrium behavior. Especially for the rotating-wave intra-system coupling, it was shown that the local approach completely fails to reproduce even qualitatively the critical behavior. It was further seen, that nonlocal properties like the quantum mutual information is generally smaller for the local approach as it cannot create the same amount of correlations. For vanishing temperatures, the local approach is further not able to create sufficiently strong quantum coherences in the position-position coupling case, approaching the critical point. In the nonequilibrium situation, the approaches were compared with a quantum Langevin equation solution. The local approach showed a qualitative difference in the error behavior depending on the sign of the temperature difference. Interestingly, the global approach also differs from the quantum Langevin equation solution in the position-position case, while they completely agree in the rotating-wave case.

In the context of perturbation theory, the local approach is nothing else than the zeroth order perturbation. Chapter 2 can thus be interpreted as an investigation in how well the zeroth order perturbation is able to describe steady-state behavior of quantum master equations. In chapter 3 we went further and consider what effects higher order perturbations create and if these become monotonously better in describing the global approach. The major focus was put into the first order perturbative correction, which

was compared with the global and local approaches using the same coupled harmonic oscillator model, with the focus on the position-position coupling. From the expansion of ordinary functions, perturbation theory is expected to behave better the higher the order of perturbation becomes, in particular for small perturbations. It was shown there, that in the equilibrium situation, this improvement could be observed partially, especially for finite temperatures and nonlocal observables such as the quantum mutual information. Local observables, as the considered mean occupation number of one of the oscillators, however, behaves differently than the local approach, but does not necessarily reproduce the global result better. Calculating thermodynamic properties, however, showed that both the local and first order perturbative approach lead to unphysical heat currents from the baths in the equilibrium case. And even more pressing, in the vanishing temperature case, the first order perturbative approach led to a steady-state covariance matrix that violated the uncertainty principle. The nonequilibrium regime further showed, that there exist, for the nonresonant oscillators ($\omega_1 \neq \omega_2$), two regimes in the case that the hotter bath is coupled to the oscillator with the higher frequency. One can be termed the 'normal' regime where the hotter bath results in a higher local occupation number for its corresponding oscillator compared to the colder bath and an 'anomalous' regime where the hotter bath coupled to the higher frequency still creates a lower occupation number than the colder bath, i.e. $N(\beta_1,\omega_1) < N(\beta_2,\omega_2)$, but $\beta_1 < \beta_2$. In the normal regime, the local approach can reproduce the global approach's local and nonlocal behavior better than the first order perturbative one. In particular heat currents and entropy production are physical and qualitiatively behaving like in the global case, whereas the first order perturbative approach is unphysical. In the anomalous regime, however, this is reversed, in particular the heat currents and entropy production are now physical for the first order perturbative approach, while the local one becomes unphysical. Therefore, the result of approximative approaches to circumvent the full derivation of a quantum master equation is rather mixed. While both the local and the first order perturbative approach are able to reproduce the global behavior qualitatively, they do so with differing levels of success, depending on the exact choice of temperature, coupling strength, small differences in the Hamiltonian and equilibrium or nonequilibrium situations. Especially the first order perturbative approach, while being of first order compared to the local approach's zeroth order, is interestingly not necessarily better than the local approach, even for infinitesimally small intra-system coupling. If the order of perturbation is further increased, as in the considered case of second order, the perturbative description improves significantly and considered properties were following the global approach's behavior more closely and all unphysical properties vanished for small perturbations.

The last two chapters then treated some general dynamical properties of quantum Markovian master equations. In chapter 4 quantum response theory of open quantum systems was treated. In particular, three different classes of response functions were derived akin to a classical treatment [37]. The first class is a response function that does not contain a time-derivative and is directly dependent on the unperturbed steady-state density operator. One can further conclude from this form, that for Hamiltonian perturbation and a thermal system, where the perturbation Hamiltonian commutes with the system Hamiltonian, the response vanishes. The second form derived was shown to be

identified as a correlation function containing a transformed stochastic entropy, which is also an open system, nonequilibrium generalization of the canonical correlation response form derived by Kubo [38] in the closed thermal case. For this form the quantum detailed balance condition was used to show symmetry properties of the quantum response. The third form is the one not depending explicitly on any density operator, neither the unperturbed, nor the perturbed ones. For Hamiltonian perturbations, the third form is given as a commutator between the perturbation Hamiltonian and the observable of interest. For general (dissipative) perturbations, the adjoint generator of the perturbation acts on the observable of interest, creating the correlation function for the third form. These classes were then compared with classical nonequilibrium forms as well as other quantum response functions derived by various authors. It was also investigated that there exists a symmetry for the response function in the case of quantum detailed balance quantum master equations. Finally two examples were considered. First the rotating-wave Hamiltonian of the two harmonic oscillators subject to a local master equation was used to consider the different regimes of equilibrium and nonequilibrium systems, for a Hamiltonian perturbation. The nonequilibrium regime showed detailed balance breaking in its response function and the equivalence of the different classes was stressed. The second example treated the breakdown of linear response approaching a critical point by using the position-position Hamiltonian and the global master equation approach. It was shown that regardless which regime the dynamics are considered in, all equilibrium and nonequilibrium cases show a similar breakdown close to the critical point.

Lastly, In chapter 5, the response function classes were used to derive fluctuation-dissipation theorems for quantum Markovian master equations. This was done by first recapping the classical form for fluctuation-dissipation theorems and considering the properties of classical and quantum correlation functions. In the quantum case, the closed, thermal, form of the fluctuation-dissipation theorem depends fundamentally on the KMS property and boundary condition. In the open, in general nonequilibrium steady state case, this does not hold. Further, the dynamics in the open case are not completely defined by the system Hamiltonian, but are generated by the bath properties and the system-bath interaction. Therefore, it is expected that the FDT's for the open quantum systems do not necessarily reproduce the Kubo FDT. Thus the fluctuation-dissipation theorems cannot resemble the closed form. The FDT's from the third form are for purely dissipative perturbations given by a sum of Fourier transformed correlation functions that are multiplied by the corresponding friction coefficients of the master equation. The first class is already a correlation function and thus the FDT is a trivial identity. Since the second form contains a time derivative, the Fourier transform between these contains a linear frequency dependence.

To summarize, we showed in the first two chapters that perturbative approaches to quantum master equations have difficulties reproducing especially thermodynamic properties, even for the simple example of two coupled harmonic oscillators. Depending on the chosen parameter, the first order perturbative approaches have to be used with care and in general lower perturbation orders can be unreliable even at infinitesimal coupling strengths. This suggests that these approximative approaches are rather unreliable and better suited as being used as an approximative model whose parameter

are rather to be fitted, as long as the local or first order perturbative equation reproduces the already expected behavior qualitatively, instead of using their properties as necessarily the correct behavior of a system. The response classes derived may be helpful different situations. The second form showed interesting capabilities for deriving mathematical properties of such open systems as the shown symmetry properties for quantum detailed balance quantum Markovian master equations, while the third form is exceptionally helpful in direct calculation of weakly driven systems. These classes were also useful in deriving the different variants of FDT's and we expect these classes to be useful for future studies of the quantum response of nonequilibrium steady-state systems. Further, the consideration of the different properties of correlation functions and the investigation of the differing properties between open and closed dynamics in the light of FDT's may further be hopeful to be used as a stepping stone for deriving even more forms of FDT's that expose more intricate properties of such systems, akin to the closed quantum form of the FDT (5.2), that expresses clearly its quantum structure.

7

APPENDIX

7.1. METHODS FOR SOLVING THE COUPLED OSCILLATORS

A Gaussian system is completely defined by its first and second moments. We explicitly solve the linear local and global quantum master equations by computing the first and second moments in symplectic space [6]. The symmetric characteristic function is defined by $\chi(\alpha_1,\alpha_2)=\langle D_1(\alpha_1)\otimes D_2(\alpha_2)\rangle$, where $D_i(\alpha_i)=\exp(\alpha_i a_i^\dagger-\alpha_i^* a_i)$ is the displacement operator. The (symmetric) moments are then obtained by differentiation [140],

$$\langle a_i^{\dagger k} a_j^l\rangle_s = \frac{d^k}{d\alpha_i^k}\frac{d^l}{(-\alpha_j^*)^l}\chi(\alpha_1,\alpha_2)\bigg|_{\alpha_1=\alpha_2=0}, \tag{7.1}$$

where $\langle\cdot\rangle_s$ is the expectation value of the symmetrized version of the operators $a_i^{\dagger k}a_j^l$. The evolution of the characteristic function is derived from the master equation

$$\frac{d}{dt}\chi(\alpha_1,\alpha_2)=\mathrm{Tr}\{D_1(\alpha_1)\otimes D_2(\alpha_2)\dot{\rho}\}, \tag{7.2}$$

together with the identities,

$$\begin{aligned} D_i a_i^\dagger &= \left(-\frac{\alpha_i^*}{2}+\frac{d}{d\alpha_i}\right)D_i,\ D_i a_i = \left(-\frac{\alpha_i}{2}-\frac{d}{d\alpha_i^*}\right)D_i,\\ a_i^\dagger D_i &= \left(\frac{\alpha_i^*}{2}+\frac{d}{d\alpha_i}\right)D_i,\ a_i D_i = \left(\frac{\alpha_i}{2}-\frac{d}{d\alpha_i^*}\right)D_i. \end{aligned} \tag{7.3}$$

with $\alpha_i=x_i+ip_i$ and $d/d\alpha_i=(d/dx_i-id/dp_i)/2$ and using the Gaussian ansatz for the characteristic function $\chi(x_1,p_1,x_2,p_2)=\exp(i\vec{P}\vec{\overline{y}}-\vec{P}^T\overline{\sigma}\vec{P}/2)$ with $\vec{P}=(x_1,p_1,x_2,p_2)$ and $\vec{\overline{y}}=(\overline{y}_1,\overline{z}_1,\overline{y}_2,\overline{z}_2)$. The over lined terms $\overline{y},\overline{\sigma}$ correspond to the symplectic transformed first and second moments y,σ. Since the Hamiltonian (2.2) is quadratic, the steady-state values for the first moments always vanish $\overline{y}_i=0=\overline{z}_i$ and the system is completely described by the second moments, simplifying the description. Writing these second

moments in symplectic space in vector form $\vec{\sigma} = (\overline{\sigma}_{x1x1}, \overline{\sigma}_{x1p1}, \overline{\sigma}_{x1x2}, \overline{\sigma}_{x1p2}, \overline{\sigma}_{p1p1}, \overline{\sigma}_{p1x2}, \overline{\sigma}_{p1p2}, \overline{\sigma}_{x2x2}, \overline{\sigma}_{x2p2}, \overline{\sigma}_{p2p2})$, one may denote the steady-state set of equations as $\vec{G} = \underline{\Lambda}\vec{\sigma}$. The 10×10 matrix $\underline{\Lambda}$ can be written row-wise, using the collectives terms for the Γ's, $2\Gamma(i,j,k,l,m,n,o,p) = (-1)^i\Gamma(a_1,a_2) + (-1)^j\Gamma(a_1,a_2^\dagger) + (-1)^k\Gamma(a_1^\dagger,a_2) + (-1)^l\Gamma(a_1^\dagger,a_2^\dagger) + (-1)^m\Gamma(a_2,a_1) + (-1)^n\Gamma(a_2,a_1^\dagger) + (-1)^o\Gamma(a_2^\dagger,a_1) + (-1)^p\Gamma(a_2^\dagger,a_1^\dagger)$. We have

$$
\begin{aligned}
\underline{\Lambda}_1 =& (\Gamma(a_1,a_1^\dagger) - \Gamma(a_1^\dagger,a_1), -\omega_1, \Gamma(1,0,1,0,0,0,1,1), -i\Gamma(1,1,1,1,0,0,0,0) - (\kappa+\lambda),\\
& 0,0,0,0,0,0)\\
\underline{\Lambda}_2 =& (\omega_1, 2\Gamma(a_1,a_1^\dagger) - 2\Gamma(a_1^\dagger,a_1), i\Gamma(1,0,0,1,0,1,1,0) - (\kappa-\lambda), \Gamma(0,0,1,1,1,0,1,0),\\
& -\omega_1, \Gamma(1,0,1,0,0,0,1,1), i\Gamma(1,1,1,1,0,0,0,0) - (\kappa+\lambda), 0,0,0)\\
\underline{\Lambda}_3 =& (\Gamma(0,0,1,1,1,0,1,0), i\Gamma(0,0,0,0,1,1,1,1) - (\kappa+\lambda), \Gamma(a_1,a_1^\dagger) - \Gamma(a_1^\dagger,a_1) + \Gamma(a_2,a_2^\dagger)\\
& - \Gamma(a_2^\dagger,a_2), -\omega_2, 0, -\omega_1, 0, \Gamma(1,0,1,0,0,0,1,1), i\Gamma(1,1,1,1,0,0,0,0) - (\kappa+\lambda), 0)\\
\underline{\Lambda}_4 =& (-(\kappa-\lambda) + i\Gamma(0,1,1,0,1,0,0,1), \Gamma(1,0,1,0,0,0,1,1), \omega_2, \Gamma(a_1,a_1^\dagger) - \Gamma(a_1^\dagger,a_1)\\
& + \Gamma(a_2,a_2^\dagger) - \Gamma(a_2^\dagger,a_2), 0,0, -\omega_1, 0, \Gamma(1,0,1,0,0,0,1,1), -(\kappa+\lambda) + i\Gamma(1,1,1,1,0,0,0))\\
\underline{\Lambda}_5 =& (0, \omega_1, 0, 0, \Gamma(a_1,a_1^\dagger) - \Gamma(a_1^\dagger,a_1), -(\kappa-\lambda) + i\Gamma(1,0,0,1,0,1,1,0),\\
& \Gamma(0,0,1,1,1,0,1,0), 0,0,0)\\
\underline{\Lambda}_6 =& (0, \Gamma(0,0,1,1,1,0,1,0), \omega_1, 0, i\Gamma(0,0,0,0,1,1,1,1) - (\kappa+\lambda), \Gamma(a_1,a_1^\dagger) - \Gamma(a_1^\dagger,a_1)\\
& + \Gamma(a_2,a_2^\dagger) - \Gamma(a_2^\dagger,a_2), -\omega_2, i\Gamma(1,0,0,1,0,1,1,0) + (\lambda-\kappa), \Gamma(0,0,1,1,1,0,1,0), 0)\\
\underline{\Lambda}_7 =& (0, i\Gamma(0,1,1,0,1,0,0,1) + (\lambda-\kappa), 0, \omega_1, \Gamma(1,0,1,0,0,0,1,1), \omega_2, \Gamma(a_1,a_1^\dagger) - \Gamma(a_1^\dagger,a_1)\\
& + \Gamma(a_2,a_2^\dagger) - \Gamma(a_2^\dagger,a_2), 0, i\Gamma(1,0,0,1,0,1,1,0) + (\lambda-\kappa), \Gamma(0,0,1,1,1,0,1,0))\\
\underline{\Lambda}_8 =& (0,0,\Gamma(0,0,1,1,1,0,1,0), 0,0, i\Gamma(0,0,0,0,1,1,1,1) - (\kappa+\lambda), 0,\\
& \Gamma(a_2,a_2^\dagger) - \Gamma(a_2^\dagger,a_2), -\omega_2, 0)\\
\underline{\Lambda}_9 =& (0,0, i\Gamma(0,1,1,0,1,0,0,1) + (\lambda-\kappa), \Gamma(0,0,1,1,1,0,1,0), 0, \Gamma(1,0,1,0,0,0,1,1),\\
& i\Gamma(0,0,0,0,1,1,1,1) - (\kappa+\lambda), \omega_2, 2\Gamma(a_2,a_2^\dagger) - 2\Gamma(a_2^\dagger,a_2), -\omega_2)\\
\underline{\Lambda}_{10} =& (0,0,0, i\Gamma(0,1,1,0,1,0,0,1) + (\lambda-\kappa), 0,0, \Gamma(1,0,1,0,0,0,1,1), 0, \omega_2,\\
& \Gamma(a_2,a_2^\dagger) - \Gamma(a_2^\dagger,a_2)).
\end{aligned}
\tag{7.4}
$$

The steady-state vector $\vec{G}$ depends on the dissipators and explicitly reads $\vec{G} =$

$$
\begin{aligned}
&(\Gamma(a_1,a_1) + \Gamma(a_1,a_1^\dagger) + \Gamma(a_1^\dagger,a_1) + \Gamma(a_1^\dagger,a_1^\dagger), 2i\Gamma(a_1,a_1) - 2i\Gamma(a_1^\dagger,a_1^\dagger), \Gamma(a_1,a_2) + \Gamma(a_1,a_2^\dagger)\\
&+\Gamma(a_1^\dagger,a_2) + \Gamma(a_1^\dagger,a_2^\dagger) + \Gamma(a_2,a_1) + \Gamma(a_2,a_1^\dagger) + \Gamma(a_2^\dagger,a_1) + \Gamma(a_2^\dagger,a_1^\dagger), i(\Gamma(a_1,a_2) - \Gamma(a_1,a_2^\dagger)\\
&+\Gamma(a_1^\dagger,a_2) - \Gamma(a_1^\dagger,a_2^\dagger) + \Gamma(a_2,a_1) + \Gamma(a_2,a_1^\dagger) - \Gamma(a_2^\dagger,a_1) - \Gamma(a_2^\dagger),a_1^\dagger), -\Gamma(a_1,a_1) + \Gamma(a_1,a_1^\dagger)\\
&+\Gamma(a_1^\dagger,a_1) - \Gamma(a_1^\dagger,a_1^\dagger), i(\Gamma(a_1,a_2) + \Gamma(a_1,a_2^\dagger) - \Gamma(a_1^\dagger,a_2) - \Gamma(a_1^\dagger,a_2^\dagger) + \Gamma(a_2,a_1) - \Gamma(a_2,a_1^\dagger)\\
&+\Gamma(a_2^\dagger,a_1) - \Gamma(a_2^\dagger,a_1^\dagger)), -\Gamma(a_1,a_2) + \Gamma(a_1,a_2^\dagger) + \Gamma(a_1^\dagger,a_2) - \Gamma(a_1^\dagger,a_2^\dagger) - \Gamma(a_2,a_1) + \Gamma(a_2,a_1^\dagger)\\
&+\Gamma(a_2^\dagger,a_1) - \Gamma(a_2^\dagger,a_1^\dagger), \Gamma(a_2,a_2) + \Gamma(a_2,a_2^\dagger) + \Gamma(a_2^\dagger,a_2) + \Gamma(a_2^\dagger,a_2^\dagger), 2i(\Gamma(a_2,a_2) - \Gamma(a_2^\dagger,a_2^\dagger)),\\
&-\Gamma(a_2,a_2) + \Gamma(a_2,a_2^\dagger) + \Gamma(a_2^\dagger,a_2) - \Gamma(a_2^\dagger,a_2^\dagger)).
\end{aligned}
\tag{7.5}
$$

Solving this system of equations (numerically) leads to the symplectic covariance matrix. The actual covariance matrix is obtained after symplectic transformation: $\sigma_{xixj} = \overline{\sigma}_{pipj}/2$, $\sigma_{pipj} = \overline{\sigma}_{xixj}/2$, $\sigma_{xjpi} = -\overline{\sigma}_{xipj}/2$. The steady-state occupation numbers are finally calculated via $\langle a_1^\dagger a_1\rangle_{\mathrm{ss}} = (\sigma_{x1x1} + \sigma_{p1p1} - 1)/2$.

7.2. MUTUAL INFORMATION, NEGATIVITY AND HEAT CURRENTS FOR GAUSSIAN SYSTEMS

The quantum mutual information for a Gaussian system can be calculated from the covariance matrix as $I(\sigma) = f(a) + f(b) - f(n_-(\sigma)) - f(n_+(\sigma))$ [105], with $a = \sqrt{\det(\alpha)}$, $b = \sqrt{\det\beta}$, $f(x) = (x+1/2)\ln(x+1/2) - (x-1/2)\ln(x-1/2)$, $\Delta(\sigma) = \det\alpha + \det\beta + 2\det\gamma$, $n_\mp(\sigma) = \sqrt{\left(\Delta(\sigma) \mp \sqrt{\Delta(\sigma)^2 - 4\det\sigma}\right)/2}$, for the covariance matrix defined as $\sigma_{ij} = \langle x_i x_j + x_j x_i\rangle/2$, $x_i = (x_1, p_1, x_2, p_2)$. In this form, the covariance matrix contains the sub-matrices of interest via $\sigma = ((\alpha,\gamma),(\gamma^T,\beta))$.

The negativity of a Gaussian system can also be calculated from its second moment. [101]. The solutions $\zeta_{1,2,3,4}$ of the characteristic function

$$\zeta^4 + (\det\alpha + \det\beta - 2\det\gamma)\zeta^2 + \det\sigma) = 0 \tag{7.6}$$

are explicitly given by

$$\begin{aligned}\zeta_{1,2} &= \pm\frac{1}{\sqrt{2}}\sqrt{2\det\gamma - \det\alpha - \det\beta + \sqrt{(\det\alpha + \det\beta - 2\det\gamma)^2 - 4\det\sigma}}\\ \zeta_{3,4} &= \pm\frac{1}{\sqrt{2}}\sqrt{2\det\gamma - \det\alpha - \det\beta + \sqrt{(\det\alpha + \det\beta - 2\det\gamma)^2 - 4\det\sigma}}.\end{aligned} \tag{7.7}$$

The absolute values of these solutions correspond to the symplectic eigenvalues of the partially transposed matrix ρ^{T_A}. The negativity thus is given by

$$\mathcal{N} = \frac{1}{2}\left(\sum_n |\zeta_i| - 1\right). \tag{7.8}$$

The heat currents are defined as $\mathfrak{J}_i = \langle\overline{\mathscr{D}}_i(H_S)\rangle$ with the adjoint dissipator $\overline{\mathscr{D}}_i$ of bath i. The general form of any adjoint dissipator is given by (4.2). Since the system Hamiltonian (2.2) is quadratic, applying the dissipators will result in expectation values of the form

$$\left\langle\overline{\mathscr{D}}^{A_iA_j}(H_S)\right\rangle = \Gamma(A_i,A_j)(\langle[A_j,H_S]A_i - A_j[A_i,H_S]\rangle), \tag{7.9}$$

where $\overline{\mathscr{D}}^{A_iA_j}(B) := \Gamma(A_i,A_j)(A_jBA_i - \{A_jA_i,B\}/2)$ and the commutators of the Hamiltonian and the jump operators A_i result in quadratic expectation values that can be directly calculated from the covariance matrix. Summing all contributions for the different dissipators of the respective baths leads to the heat currents $\mathfrak{J}_i$.

7.3. EXPANSION OF THE INTERACTION OPERATORS

The interaction operators of the coupled harmonic oscillators, $a_1^I(t), a_1^{\dagger I}(t), a_2^I(t), a_2^{\dagger I}(t)$, in the first order of the perturbation about $\lambda = 0$ have the form

$$\begin{aligned}
a_1^{(1)}(t) &= -\frac{a_2}{\omega_1-\omega_2}\left(e^{-i\omega_2 t}-e^{-i\omega_1 t}\right)-\frac{\kappa/\lambda a_2^\dagger}{\omega_1+\omega_2}\left(e^{i\omega_2 t}-e^{-i\omega_1 t}\right)\\
a_1^{\dagger(1)}(t) &= -\frac{a_2^\dagger}{\omega_1-\omega_2}\left(e^{i\omega_2 t}-e^{i\omega_1 t}\right)-\frac{\kappa/\lambda a_2}{\omega_1+\omega_2}\left(e^{-i\omega_2 t}-e^{i\omega_1 t}\right)\\
a_2^{(1)}(t) &= \frac{a_1}{\omega_1-\omega_2}\left(e^{-i\omega_1 t}-e^{-i\omega_2 t}\right)-\frac{\kappa/\lambda a_1^\dagger}{\omega_1+\omega_2}\left(e^{i\omega_1 t}-e^{-i\omega_2 t}\right)\\
a_2^{\dagger(1)}(t) &= \frac{a_1^\dagger}{\omega_1-\omega_2}\left(e^{i\omega_1 t}-e^{i\omega_2 t}\right)+\frac{\kappa/\lambda a_1}{\omega_1+\omega_2}\left(e^{-i\omega_1 t}-e^{i\omega_2 t}\right).
\end{aligned} \tag{7.10}$$

Here it can also be seen that the used perturbation theory is restricted to off-resonant oscillators, as only for $\omega_1 \neq \omega_2$ will the dissipators be well defined.

7.4. INVESTIGATING THE ROTATING-WAVE HAMILTONIAN FOR THE FIRST ORDER PERTURBATIVE MASTER EQUATION

Here we consider the perturbative approach applied to the rotating-wave ($\kappa = 0$) Hamiltonian (2.2). Comparing the global dissipators (2.19), with the first order perturbative approach ones (3.7), one can observe that the number of nonvanishing dissipators is the same. Thus, compared to the position-position coupling, the occurring differences will be solely due to incorrect dissipator values. In Fig. 7.1 one can observe the local observable of mean occupation number of the first oscillator (left) and mutual information (right) for the equilibrium case $T_i = 98$. In the insets, one can see the nonequilibrium situation, where $T_1 = 98$, $T_2 = 20$. First considering the mean occupation number, one can observe that the first order perturbative approach (brown triangles) is also not able to reproduce the global results (green circles), similarly to the local approach. In particular, the critical behavior discussed in chapter 2 cannot be reproduced by this approach either for the local occupation number.

As in the position-position coupling case, the quantum mutual information can be quite well approximated by the first order perturbative approach in the equilibrium case. Qualitatively, also in the nonequilibrium case it is behaving similarly. also the nonequilibrium case can be approximated. Remarkably, in the equilibrium case, the mutual information can reproduce the critical behavior. However, since the mean occupation number does not increase accordingly, the physicality of the first order perturbative steady-state may be rather questionable approaching the critical point. This can be seen by considering the physical properties in the form of the heat current of the first oscillator $\mathfrak{J}_1$ (left) and uncertainty relation for the steady-state (right) in Fig. 7.2 for the equilibrium situation $T_1 = T_2 = 98$ and nonequilibrium case $T_1 = 98$, $T_2 = 20$ (inset). Once more, in the equilibrium case, the heat current of the first order perturbative approach is also unphysical as the local one. The nonequilibrium case in the normal regime (inset in Fig. 7.2) shows also that the heat current of the first order perturbative approach

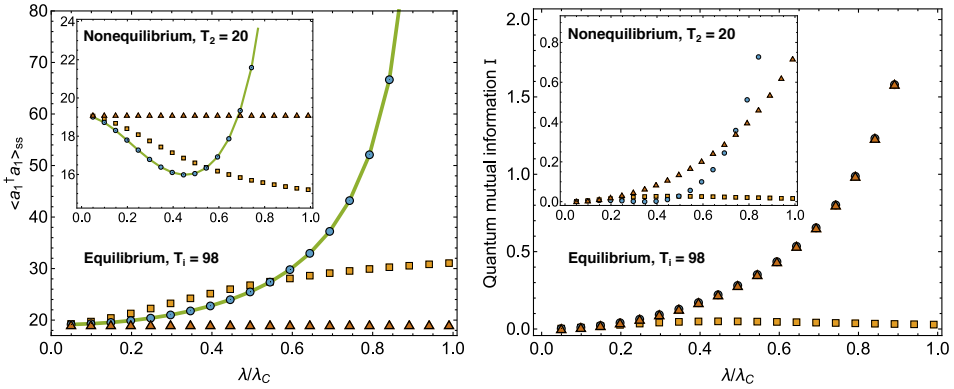

Figure 7.1: Comparison of the global, local and first order perturbative master equation for the local mean occupation number $\langle a_1^\dagger a_1\rangle$ (left) and the quantum mutual information (right) in dependence of the intra-system coupling λ/λ_c of the rotating-wave Hamiltonian (2.2), for the cases of (equilibrium) temperature $T_i = 98$ and nonequilibrium $T_1 = 98$, $T_2 = 20$ (inset). The global ME solution (blue circles) is shown with the reference quantum Langevin solution (green line). (left): The first order perturbative approach (brown triangles) has a similar difficulty reproducing the global result as the local approach (orange squares). This holds true for both the equilibrium and nonequilibrium cases (inset). Interestingly, neither equation can reproduce the critical behavior of the global approach. (right): The mutual information on the other hand can be quite well reproduced by the first order perturbative approach in the considered λ/λ_c regime. In particular, the critical behavior seems to be reproduced here using the first order perturbative approach. Parameters are $\gamma_1 = \gamma_2 = 1.5\cdot 10^{-4}$, $\omega_1 = 5, \omega_2 = 2$.

can be unphysical there. Considering the uncertainty relation, one can observe that regardless whether equilibrium or nonequilibrium, the first order perturbative approach always becomes unphysical approaching the critical point. This is clear, since the local observables like the mean occupation numbers stay approximately constant, but the mutual information, i.e. the intra-system correlations increase strongly. Therefore, the perturbation of the rotating-wave Hamiltonian shows similar difficulties of the orders in describing the full thermodynamic properties of the system.

7.5. ADJOINT DYNAMICS CALCULATION FOR THE HARMONIC OSCILLATOR MODEL

The goal is to calculate the adjoint dynamics given by the generator (4.2) in the Banach space of the operators $\mathscr{B}(\mathscr{H}_S)$. Even after simplifying the resulting equation using commutation relations, one in general receives an operator differential equation which is typically nontrivial to solve. One can note that the Hamiltonian of the two coupled harmonic oscillators (2.2) is a quadratic Hamiltonian. Due to that, one is able to find a closed set of (operator) differential equations that can be used to find a solution for the adjoint dynamics using matrix exponentiation [6]. For the considered quadratic system, this set of operators is given by $\vec{v}(t)^T = \Big(a_1^\dagger a_1(t), a_1^2(t), a_1^{\dagger 2}(t), a_2^\dagger a_2(t), a_2^2(t), a_2^{\dagger 2}(t), a_1 a_2(t), a_1 a_2^\dagger(t), a_1^\dagger a_2(t), a_1^\dagger a_2^\dagger(t), \mathbb{1}\Big)$, where $\mathbb{1}$ is the identity operator. Using the generator

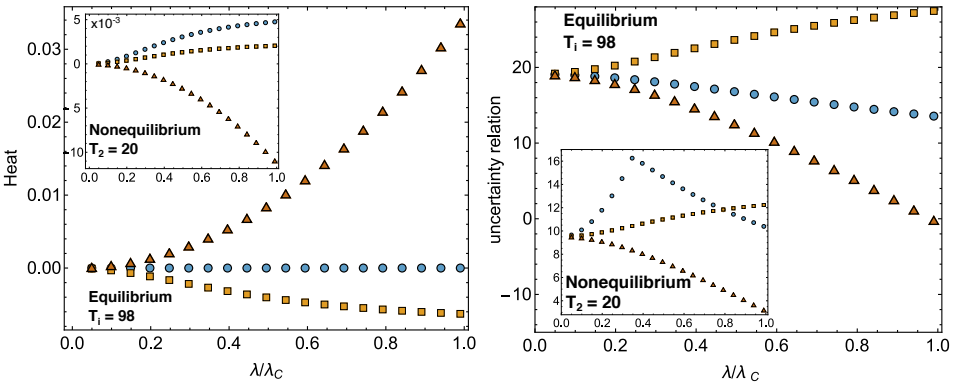

Figure 7.2: Comparison of the global, local and first order perturbative master equation heat current from the first bath $\mathfrak{J}_1$ (left) and the check whether the uncertainty principle is fulfilled (right) in dependence of the intra-system coupling λ/λ_c of the rotating-wave Hamiltonian (2.2), for the cases of the (equilibrium) temperature $T_i = 98$ and nonequilibrium $T_1 = 98$, $T_2 = 20$ (inset). (left): The global approach (blue circles) reproduces the expected behavior of no net heat current from the baths as the equilibrium case is considered here. However, both the heat currents for the first order perturbative (brown triangles) and local approach (orange squares) are unphysical. The nonequilibrium case (inset) shows that, in the considered normal regime, the first order perturbative approach has unphysical heat currents . Considering the fulfillment of the uncertainty relation (right), the first order perturbative approach becomes unphysical for any finite temperature approaching the critical point $\lambda/\lambda_c = 1$, regardless whether it is a equilibrium or nonequilibrium situation. Parameters are the same as in Fig. 7.1.

(4.2), one receives the following general form of the closed set of differential equations

$$\frac{d}{dt}\vec{v}(t) = M\vec{v}(t). \tag{7.11}$$

If the matrix M has a sufficiently simple eigensystem, this may be analytically solved by matrix exponentiation $\vec{v}(t) = e^{Mt}\vec{v}(0)$.

For the position-position coupling Hamiltonian (2.2) ($\kappa = \lambda$) and the global master equation treatment (2.20) cannot be solved analytically, even for the stationary properties (see 7.1 for details of calculating steady-state covariance matrices). The breakdown of the linear response considered in chapter 4.7 further necessitates the knowledge of the dynamical properties of the system by virtue of the adjoint dynamics (4.2). However, matrix exponentiation in this case does not work, as the general form of the dissipators and the Hamiltonian result a dynamical matrix M that has a complex dependence on the system parameters. However, it is still possible to find such a matrix and therefore one can at least solve the adjoint dynamics numerically. In fact, since one can find this closed set of differential equations for the operators $\vec{v}(t)$, one can solve the differential equation (7.11) using the ansatz

$$\begin{aligned} v_i = &f_i(t)a_1^\dagger a_1 + g_i(t)a_1^2 + h_i(t)a_1^{\dagger 2} + j_i(t)a_2^\dagger a_2 + l_i(t)a_2^2 + m_i(t)a_2^{\dagger 2} \\ &+ n_i(t)a_1 a_2 + p_i(t)a_1 a_2^\dagger + q_i(t)a_1^\dagger a_2 + r_i(t)a_1^\dagger a_2^\dagger + s_i(t). \end{aligned} \tag{7.12}$$

This will then transform the set of operator differential equations (7.11) into a set of c-number differential equations for the 121 functions f_i, g_i etc, which can be numerically

calculated with the starting condition $f_i(0) = \delta_{i1}$, $g_i(0) = \delta_{i2}$, etc. Using these solutions for the observable of interest $a_1^\dagger a_1(t)$, then the response (4.23) can be directly calculated.

7.6. LINEAR RESPONSE FOR THE LOCAL COUPLED-OSCILLATOR MODEL

Using the methods considered in 7.1 for covariance matrices of the quadratic system with the help of the symplectic space, one has for the steady-state solution vanishing first moments, $\langle x_{1,2}\rangle = \langle p_{1,2}\rangle = 0$, and the covariance matrix,

$$\sigma = \zeta \begin{pmatrix} D+n_1+\frac{1}{2} & 0 & -\delta C & -\gamma C \\ 0 & D+n_1+\frac{1}{2} & \gamma C & -\delta C \\ -\delta C & \gamma C & D+n_2+\frac{1}{2} & 0 \\ -\gamma C & -\delta C & 0 & D+n_2+\frac{1}{2} \end{pmatrix}, \tag{7.13}$$

where $n_i = N(\beta_i, \omega_i)$ and with the three parameters,

$$\zeta = \frac{\gamma^2+\delta^2}{4\lambda^2+\gamma^2+\delta^2},\ D = \frac{2\lambda^2(n_1+n_2+1)}{\gamma^2+\delta^2},\ C = \frac{\lambda(n_1-n_2)}{\gamma^2+\delta^2}. \tag{7.14}$$

The adjoint dynamics in this case can be analytically calculated by matrix exponentiation as treated in (7.5). The matrix M, where diag(γ) is a diagonal matrix with elements γ, explicitly reads $M + \text{diag}(\gamma) =$

$$\begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & i\lambda & -i\lambda & 0 & n_1\gamma \\ 0 & -2i\omega_1 & 0 & 0 & 0 & 0 & -2i\lambda & 0 & 0 & 0 & 0 \\ 0 & 0 & 2i\omega_1 & 0 & 0 & 0 & 0 & 0 & 0 & 2i\lambda & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & -i\lambda & i\lambda & 0 & n_2\gamma \\ 0 & 0 & 0 & 0 & -2i\omega_2 & 0 & -2i\lambda & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 2i\omega_2 & 0 & 0 & 0 & 2i\lambda & 0 \\ 0 & -i\lambda & 0 & 0 & -i\lambda & 0 & -i\omega_{12} & 0 & 0 & 0 & 0 \\ i\lambda & 0 & 0 & -i\lambda & 0 & 0 & 0 & -i\Delta\omega & 0 & 0 & 0 \\ -i\lambda & 0 & 0 & i\lambda & 0 & 0 & 0 & 0 & i\Delta\omega & 0 & 0 \\ 0 & 0 & i\lambda & 0 & 0 & i\lambda & 0 & 0 & 0 & i\omega_{12} & 0 \end{pmatrix}, \tag{7.15}$$

with $\omega_{12} = \omega_1 + \omega_2$ and $\Delta\omega = \omega_1 - \omega_2$. The time dependence of the number operator, $a_1^\dagger a_1$, of the first oscillator then has the following general form

$$\begin{aligned} a_1^\dagger a_1(t) = & f(t)a_1^\dagger a_1 + g(t)a_1^2 + h(t)a_1^{\dagger 2} + j(t)a_2^\dagger a_2 + l(t)a_2^2 + m(t)a_2^{\dagger 2} \\ & + n(t)a_1 a_2 + p(t)a_1 a_2^\dagger + q(t)a_1^\dagger a_2 + r(t)a_1^\dagger a_2^\dagger + s(t), \end{aligned} \tag{7.16}$$

for the closed set of operators $\vec{v}(t)$. The various functions appearing in Eq. (7.16) are

given explicitly by the exponentiation and have the following form,

$$\begin{aligned} f(t) &= e^{-\gamma t}\left(\delta^2+2\lambda^2+2\lambda^2\cos zt\right)/z^2, \\ j(t) &= -2\lambda^2 e^{-\gamma t}\left(\cos zt-1\right)/z^2, \\ p(t) &= \lambda e^{-\gamma t}\left(-\delta+iz\sin zt+\delta\cos zt\right)/z^2, \\ q(t) &= \lambda e^{-\gamma t}\left(-\delta-iz\sin zt+\delta\cos zt\right)/z^2, \\ r(t) &= 0,\ g(t)=0,\ h(t)=0, \\ l(t) &= 0,\ m(t)=0,\ n(t)=0, \end{aligned} \tag{7.17}$$

together with the term proportional to the identity $\mathbb{1}$ which defines the large-time behavior of the adjoint dynamics,

$$\begin{aligned} s(t) = &\frac{1}{z^3\left(\gamma^2+\delta^2+4\lambda^2\right)}e^{-\gamma t} \\ &\times\left\{z\left[\left(\gamma^2+z^2\right)\left(\delta^2 n_1+2\lambda^2(n_1+n_2)\right)\right.\right. \\ &-z^2e^{\gamma t}\left(n_1\left(\gamma^2+\delta^2\right)+2\lambda^2(n_1+n_2)\right)\left.\right] \\ &+2\gamma\lambda^2(n_1-n_2)\left(\gamma z\cos zt-z^2\sin zt\right)\left.\right\}. \end{aligned} \tag{7.18}$$

For $\lambda=0$, Eq. (7.16) simplifies to $a_1^\dagger a_1(t)=e^{-\gamma t}a_1^\dagger a_1+(1-e^{-\gamma t})n_1$, as expected for a thermal oscillator [6].

7.7. DERIVATION OF THE GENERALIZED KUBO TRANSFORM

To proof the generalized Kubo transform (4.19), let us first consider a general operator identity. The following operator identity holds true for arbitrary positive operators $A.B$ [135]

$$A-B=\int_0^1 B^\lambda(\ln A-\ln B)A^{1-\lambda}\mathrm{d}\lambda. \tag{7.19}$$

This can be proven by differentiating the following formula w.r.t. t

$$\begin{aligned} &\frac{\mathrm{d}}{\mathrm{d}t}(1-B^tA^{-t})=\frac{\mathrm{d}}{\mathrm{d}t}\int_0^t B^\lambda(\ln A-\ln B)A^{1-\lambda}\mathrm{d}\lambda \\ \Leftrightarrow\ &-B^t\ln(B)A^{-t}+B^t\ln(A)A^{-t}=B^t(\ln A-\ln B)A^{-t}, \end{aligned} \tag{7.20}$$

since $\mathrm{d}_tA^t=\mathrm{d}_te^{t\ln A}=A^t\ln A$. Thus both sides have to fulfill the same first order differential equation. For $t=0$ both sides also vanish and therefore are equal for all t. Multiplying (7.20) with A^t on the right one receives identity (7.19).

Now let's consider a general density operator π_ε that depends continuously on a parameter ε. The partial derivative w.r.t. the parameter ε is defined as a limit process, analogously as for c-number functions. Using (7.19) thus results in

$$\begin{aligned} \partial_\varepsilon\pi_\varepsilon &:= \lim_{\delta\to0}\frac{\pi_{\varepsilon+\delta}-\pi_\varepsilon}{\delta}\overset{(7.19)}{=}\lim_{\delta\to0}\frac{1}{\delta}\int_0^1\pi_\varepsilon^\lambda(\ln\pi_{\varepsilon+\delta}-\ln\pi_\varepsilon)\pi_{\varepsilon+\delta}^{1-\lambda}\,\mathrm{d}\lambda \\ &=\int_0^1\pi_\varepsilon^\lambda(\partial_\varepsilon\ln\pi_\varepsilon)\pi_\varepsilon^{1-\lambda}\,\mathrm{d}\lambda. \end{aligned} \tag{7.21}$$

Further, the faithful state for fixed ε_n has the form $\pi_\varepsilon = \pi_0 + \sum_n \varepsilon_n \pi_1^n$ and thus $\partial_{\varepsilon_n}\pi_\varepsilon = \pi_1^n$. Using the identity (7.21), we therefore obtain the following identity, for $\varepsilon_n = 0$, and the density operator π_ε, $\partial_{\varepsilon_n}\pi_\varepsilon|_0/\pi_0 = \int_0^1 \pi_0^\lambda (\partial_{\varepsilon_n} \ln \pi_\varepsilon)|_0 \pi_0^{-\lambda} d\lambda =: \overline{\partial_{\varepsilon_n} \ln \pi_\varepsilon|_0}$, which proves the identity used for the generalized Kubo transform (4.19).

7.8. TIME-REVERSAL FOR CLOSED AND OPEN DYNAMICS

CLOSED CASE

Time-reversal in the closed quantum setting can be defined using the antiunitary time-reversal operator $\Theta = \Theta^{-1} = \Theta^\dagger$ [144]. This time-reversal operation attempts to take the classical notion that certain variables change under time-reversal their sign while others do not. For example, the time-reversal of the position operator should not change $\Theta x \Theta^{-1} = x$ while the momentum operator should have a sign change $\Theta p \Theta^{-1} = -p$; in general $\Theta A \Theta^{-1} = \mu_A A$, $\mu = \pm 1$ for Hermitian operators. Let us consider the time-reversal of a (general) Heisenberg operator $A(t)$ with well defined time-reversal parity μ_A. The dynamics of the time-reversed operator $A^R(t) = \Theta A^\dagger(t)\theta^{-1}$ can be derived from the invariance of the trace operation upon time-reversal

$$\begin{aligned} \mathrm{Tr}\{A(t)\rho\} &= \sum_n (\langle n|\Theta)(A(t)\rho\Theta|n\rangle) = \sum_n \langle n|(\Theta A(t)\rho\Theta|n\rangle)^* \\ &= \sum_n \langle n|(\Theta A(t)\rho\Theta)^\dagger|n\rangle = \mathrm{Tr}\{\Theta A^\dagger(t)\Theta\rho^R\}. \end{aligned} \tag{7.22}$$

OPEN CASE

Open quantum dynamics are, compared to closed dynamics, fundamentally irreversible, i.e. not time-reversal invariant. Therefore, in this context, any 'time-reversal' operation cannot be taken literally by replacing t with $-t$ as in the closed case. Nevertheless, if the quantum Markovian dynamics are being derived from first principles of a system + bath, then time-reversal symmetry has to hold. in fact, this implies that regardless which time direction one chooses, the system will always experience open dynamics. How these exactly look like, however is in general not clear. Using the generic form of a quantum Markovian generator

$$\mathscr{L}(\rho) = -i[H,\rho] + \mathscr{D}(\rho), \tag{7.23}$$

with a selfadjoint dissipator $\mathscr{D}$, a straightforward definition of time-reversal is given by [39, 115, 133, 149, 150] only changing the unitary part of the dynamics akin to a closed treatment

$$\mathscr{L}^R(\rho) = i[H,\rho] + \mathscr{D}(\rho). \tag{7.24}$$

This form can be derived from the generator (7.23) by once again using the time-reversal invariance of the trace operation, denoting the time-reversal operation here as $\mathscr{L}^R = \widetilde{\mathscr{L}}$,

$$\mathrm{Tr}\{A\mathscr{L}(\rho)\} = \mathrm{Tr}\{\widetilde{A\mathscr{L}(\rho)}\} = \mathrm{Tr}\{\Theta\mathscr{L}(\rho)^\dagger\Theta\widetilde{A}\} = \mathrm{Tr}\{\widetilde{\mathscr{L}}(\widetilde{\rho})\widetilde{A}\}, \tag{7.25}$$

or in operator notation $\widetilde{\mathscr{L}(\rho)} =: \widetilde{\mathscr{L}}(\widetilde{\rho})$. The same holds true for the adjoint superoperator $\overline{\mathscr{L}} \to \overline{\mathscr{L}}^R$. It is interesting to note, that using the generators $\mathscr{L}, \mathscr{L}^R$ one can observe fundamental differences between the possible stationary states of open quantum Markovian maps compared to unitary maps. Not all stationary states have to be time-reversal

symmetric. In fact, only steady-states that commute with the Hamiltonian can be time-reversal symmetric, as it is then the case that $\mathscr{L}(\pi_{ss}) = -i[H,\pi_{ss}] + \mathscr{D}(\pi_{ss}) = \mathscr{D}(\pi_{ss}) = 0$, i.e. both the Hamiltonian and dissipative part vanish individually. Only then this π_{ss} is also invariant under the time-reversed generator (7.24). However, in general it is given that $\pi_{ss}^R \neq \pi_{ss}$.

BIBLIOGRAPHY

[1] M. Jammer, *The conceptual development of quantum mechanics* (Tomash Publishers, 1989).

[2] A. Kramida, *A critical compilation of experimental data on spectral lines and energy levels of hydrogen, deuterium, and tritium,* Atomic Data and Nuclear Data Tables **96**, 586 (2010).

[3] A. Pais, *Einstein and the quantum theory,* Rev. Mod. Phys. **51**, 863 (1979).

[4] A. Messiah, *Quantum mechanics, Vol 1* (North-Holland, Amsterdam, 1961).

[5] I. Prigogine, *Vom Sein zum Werden* (Piper Verlag, München, 1979).

[6] H.-P. Breuer and F. Petruccione, *The Theory of Open Quantum Systems* (Springer, Berlin, 2002).

[7] R. A. van Santen and J. W. Niemantsverdriet, *The rate equation,* in *Chemical Kinetics and Catalysis* (Springer US, Boston, MA, 1995) pp. 21–72.

[8] N. G. Van Kampen, *Stochastic processes in physics and chemistry*, Vol. 1 (Elsevier, 1992).

[9] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom photon interactions* (Wiley, Weinheim, 2004).

[10] A. Levy and R. Kosloff, *The local approach to quantum transport may violate the second law of thermodynamics,* EPL (Europhysics Letters) **107**, 20004 (2014).

[11] H. Risken, *The Fokker-Planck Equation: Methods of Solution and Applications* (Springer Berlin Heidelberg, Berlin, Heidelberg, 1996) pp. 63–95.

[12] G.-L. Ingold, *Coherent Evolution in Noisy Environments* (Springer Berlin Heidelberg, Berlin, Heidelberg, 2002) pp. 1–53.

[13] S. Albeverio, L. Cattaneo, S. Mazzucchi, and L. Di Persio, *A rigorous approach to the feynman-vernon influence functional and its applications. i,* Journal of Mathematical Physics **48**, 102109 (2007).

[14] U. Weiss, *Quantum Dissipative Systems* (World Scientific, Singapore, 2008).

[15] C. Gardiner and P. Zoller, *Quantum Noise* (Springer, Berlin, 2004).

[16] W. Vogel and D.-G. Welsch, *Quantum optics* (Wiley Verlag, Weinheim, 2007).

[17] Y. Tanimura, *Numerically "exact" approach to open quantum dynamics: The hierarchical equations of motion (heom),* The Journal of Chemical Physics **153**, 020901 (2020).

[18] K. Mølmer, Y. Castin, and J. Dalibard, *Monte carlo wave-function method in quantum optics,* J. Opt. Soc. Am. B **10**, 524 (1993).

[19] W. Pauli, *Über das H-Theorem vom Anwachsen der Entropie vom Standpunkt der neuen Quantenmechanik* (Hirzel, Leipzig, 1928).

[20] R. Alicki and K. Lendi, *Quantum Dynamical Semigroups and Applications* (Springer, Berlin, 2007).

[21] A. Rivas and S. F. Huelga, *Open Quantum Systems* (Springer, Berlin, 2012).

[22] V. Gorini, A. Kossakowski, and E. C. G. Sudarshan, *Completely positive dynamical semigroups of n-level systems,* Journal of Mathematical Physics **17**, 821 (1976).

[23] G. Lindblad, *On the generators of quantum dynamical semigroups,* Communications in Mathematical Physics **48**, 119 (1976).

[24] H. Carmichael, *An Open Systems Approach to Quantum Optics* (Berlin, Springer, 1993).

[25] R. Zwanzig, *Nonequilibrium Statistical Mechanics* (Oxford University Press, Oxford, 2001).

[26] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, Cambridge, 2000).

[27] Á. Rivas, A. D. K. Plato, S. F. Huelga, and M. B. Plenio, *Markovian master equations: a critical study,* New Journal of Physics **12**, 113032 (2010).

[28] R. Kosloff, *Quantum thermodynamics: A dynamical viewpoint,* Entropy **15**, 2100 (2013).

[29] A. Dhahri, *A lindblad model for a spin chain coupled to heat baths,* Journal of Physics A: Mathematical and Theoretical **41**, 275305 (2008).

[30] F. Benatti, R. Floreanini, and L. Memarzadeh, *Bath-assisted transport in a three-site spin chain: Global versus local approach,* Phys. Rev. A **102**, 042219 (2020).

[31] P. H. Guimarães, G. T. Landi, and M. J. de Oliveira, *Nonequilibrium quantum chains under multisite lindblad baths,* Phys. Rev. E **94**, 032139 (2016).

[32] L. Banchi, P. Giorda, and P. Zanardi, *Quantum information-geometry of dissipative quantum phase transitions,* Phys. Rev. E **89**, 022102 (2014).

[33] F. Dimer, B. Estienne, A. S. Parkins, and H. J. Carmichael, *Proposed realization of the dicke-model quantum phase transition in an optical cavity qed system,* Phys. Rev. A **75**, 013804 (2007).

[34] W. Kopylov, C. Emary, and T. Brandes, *Counting statistics of the dicke superradiance phase transition,* Phys. Rev. A **87**, 043840 (2013).

[35] T. Prosen and I. Pižorn, *Quantum phase transition in a far-from-equilibrium steady state of an xy spin chain,* Phys. Rev. Lett. **101**, 105701 (2008).

[36] V. V. Albert and L. Jiang, *Symmetries and conserved quantities in lindblad master equations,* Phys. Rev. A **89**, 022118 (2014).

[37] U. Seifert and T. Speck, *Fluctuation-dissipation theorem in nonequilibrium steady states,* EPL (Europhysics Letters) **89**, 10007 (2010).

[38] R. Kubo, *The fluctuation-dissipation theorem,* Reports on Progress in Physics **29**, 255 (1966).

[39] R. Chetrite and K. Mallick, *Quantum fluctuation relations for the lindblad master equation,* Journal of Statistical Physics **148**, 480 (2012).

[40] M. Mehboudi, A. Sanpera, and J. M. R. Parrondo, *Fluctuation-dissipation theorem for non-equilibrium quantum systems,* Quantum **2**, 66 (2018).

[41] L. Campos Venuti and P. Zanardi, *Dynamical response theory for driven-dissipative quantum systems,* Phys. Rev. A **93**, 032101 (2016).

[42] W. Weidlich, *Fluctuation-dissipation theorem for a class of stationary open systems,* Zeitschrift für Physik **248**, 234 (1971).

[43] J. O. González, L. A. Correa, G. Nocerino, J. P. Palao, D. Alonso, and G. Adesso, *Testing the validity of the 'local' and 'global' gkls master equations on an exactly solvable model,* Open Systems & Information Dynamics **24**, 1740010 (2017).

[44] M. Konopik and E. Lutz, *Local master equations may fail to describe dissipative critical behavior,* arXiv preprint arXiv:2012.09907 (2020).

[45] S. Diehl, A. Micheli, A. Kantian, B. Kraus, H. P. Büchler, and P. Zoller, *Quantum states and phases in driven open quantum systems with cold atoms,* Nature Physics **4**, 878 (2008).

[46] S. Diehl, A. Tomadin, A. Micheli, R. Fazio, and P. Zoller, *Dynamical phase transitions and instabilities in open atomic many-body systems,* Phys. Rev. Lett. **105**, 015702 (2010).

[47] F. Brennecke, R. Mottl, K. Baumann, R. Landig, T. Donner, and T. Esslinger, *Real-time observation of fluctuations at the driven-dissipative dicke phase transition,* Proceedings of the National Academy of Sciences **110**, 11763 (2013).

[48] C. Carr, R. Ritter, C. G. Wade, C. S. Adams, and K. J. Weatherill, *Nonequilibrium phase transition in a dilute rydberg ensemble,* Phys. Rev. Lett. **111**, 113901 (2013).

[49] M. Aspelmeyer, T. J. Kippenberg, and F. Marquardt, *Cavity optomechanics,* Rev. Mod. Phys. **86**, 1391 (2014).

[50] R. Labouvie, B. Santra, S. Heun, and H. Ott, *Bistability in a driven-dissipative superfluid,* Phys. Rev. Lett. **116**, 235302 (2016).

[51] M. Soriente, T. Donner, R. Chitra, and O. Zilberberg, *Dissipation-induced anomalous multicritical phenomena,* Phys. Rev. Lett. **120**, 183603 (2018).

[52] D. Karevski and T. Platini, *Quantum nonequilibrium steady states induced by repeated interactions,* Phys. Rev. Lett. **102**, 207207 (2009).

[53] I. Carusotto, D. Gerace, H. E. Tureci, S. De Liberato, C. Ciuti, and A. Imamoğlu, *Fermionized photons in an array of driven dissipative nonlinear cavities,* Phys. Rev. Lett. **103**, 033601 (2009).

[54] T. Prosen and M. Žnidarič, *Long-range order in nonequilibrium interacting quantum spin chains,* Phys. Rev. Lett. **105**, 060603 (2010).

[55] T. Prosen, *Open xxz spin chain: Nonequilibrium steady state and a strict bound on ballistic transport,* Phys. Rev. Lett. **106**, 217206 (2011).

[56] T. Prosen, *Exact nonequilibrium steady state of a strongly driven open xxz chain,* Phys. Rev. Lett. **107**, 137201 (2011).

[57] T. Prosen and E. Ilievski, *Nonequilibrium phase transition in a periodically driven xy spin chain,* Phys. Rev. Lett. **107**, 060403 (2011).

[58] M. Vogl, G. Schaller, and T. Brandes, *Criticality in transport through the quantum ising chain,* Phys. Rev. Lett. **109**, 240402 (2012).

[59] J. Cui, J. I. Cirac, and M. C. Bañuls, *Variational matrix product operators for the steady state of dissipative quantum systems,* Phys. Rev. Lett. **114**, 220601 (2015).

[60] M. Foss-Feig, J. T. Young, V. V. Albert, A. V. Gorshkov, and M. F. Maghrebi, *Solvable family of driven-dissipative many-body systems,* Phys. Rev. Lett. **119**, 190402 (2017).

[61] F. Carollo, E. Gillman, H. Weimer, and I. Lesanovsky, *Critical behavior of the quantum contact process in one dimension,* Phys. Rev. Lett. **123**, 100604 (2019).

[62] V. Popkov, T. Prosen, and L. Zadnik, *Exact nonequilibrium steady state of open xxz/xyz spin-1/2 chain with dirichlet boundary conditions,* Phys. Rev. Lett. **124**, 160403 (2020).

[63] D. F. Walls, *Higher order effects in the master equation for coupled systems,* Zeitschrift für Physik A Hadrons and nuclei **234**, 231 (1970).

[64] H. J. Carmichael and D. F. Walls, *Master equation for strongly interacting systems,* Journal of Physics A: Mathematical, Nuclear and General **6**, 1552 (1973).

[65] P. D. Manrique, F. Rodríguez, L. Quiroga, and N. F. Johnson, *Nonequilibrium quantum systems: Divergence between global and local descriptions,* Advances in Condensed Matter Physics **2015**, 615727 (2015).

[66] A. S. Trushechkin and I. V. Volovich, *Perturbative treatment of inter-site couplings in the local description of open quantum networks,* EPL (Europhysics Letters) **113**, 30005 (2016).

[67] P. P. Hofer, M. Perarnau-Llobet, L. D. M. Miranda, G. Haack, R. Silva, J. B. Brask, and N. Brunner, *Markovian master equations for quantum thermal machines: local versus global approach,* New Journal of Physics **19**, 123037 (2017).

[68] J. T. Stockburger and T. Motz, *Thermodynamic deficiencies of some simple lindblad operators,* Fortschritte der Physik **65**, 1600067 (2017).

[69] M. T. Naseem, A. Xuereb, and O. E. Müstecaplıoğlu, *Thermodynamic consistency of the optomechanical master equation,* Phys. Rev. A **98**, 052123 (2018).

[70] G. D. Chiara, G. Landi, A. Hewgill, B. Reid, A. Ferraro, A. J. Roncaglia, and M. Antezza, *Reconciliation of quantum local master equations with thermodynamics,* New Journal of Physics **20**, 113024 (2018).

[71] M. T. Mitchison and M. B. Plenio, *Non-additive dissipation in open quantum networks out of equilibrium,* New Journal of Physics **20**, 033005 (2018).

[72] M. Cattaneo, G. L. Giorgi, S. Maniscalco, and R. Zambrini, *Local versus global master equation with common and separate baths: superiority of the global approach in partial secular approximation,* New Journal of Physics **21**, 113045 (2019).

[73] H. Wichterich, M. J. Henrich, H.-P. Breuer, J. Gemmer, and M. Michel, *Modeling heat transport through completely positive maps,* Phys. Rev. E **76**, 031115 (2007).

[74] A. Purkayastha, A. Dhar, and M. Kulkarni, *Out-of-equilibrium open quantum systems: A comparison of approximate quantum master equation approaches with exact results,* Phys. Rev. A **93**, 062114 (2016).

[75] D. Boyanovsky and D. Jasnow, *Heisenberg-langevin versus quantum master equation,* Phys. Rev. A **96**, 062108 (2017).

[76] M. Ludwig, K. Hammerer, and F. Marquardt, *Entanglement of mechanical oscillators coupled to a nonequilibrium environment,* Phys. Rev. A **82**, 012333 (2010).

[77] M. Aspelmeyer, T. J. Kippenberg, and F. Marquardt, *Cavity optomechanics,* Rev. Mod. Phys. **86**, 1391 (2014).

[78] R. H. Dicke, *Coherence in spontaneous radiation processes,* Phys. Rev. **93**, 99 (1954).

[79] M. Tavis and F. W. Cummings, *Exact solution for an n-molecule—radiation-field hamiltonian,* Phys. Rev. **170**, 379 (1968).

[80] T. Brandes, *Coherent and collective quantum optical effects in mesoscopic systems,* Physics Reports **408**, 315 (2005).

[81] P. Kirton, M. M. Roses, J. Keeling, and E. G. Dalla Torre, *Introduction to the dicke model: From equilibrium to nonequilibrium, and vice versa,* Advanced Quantum Technologies **2**, 1800043 (2019).

[82] C. Emary and T. Brandes, *Chaos and the quantum phase transition in the dicke model,* Phys. Rev. E **67**, 066203 (2003).

[83] N. Lambert, C. Emary, and T. Brandes, *Entanglement and the phase transition in single-mode superradiance,* Phys. Rev. Lett. **92**, 073602 (2004).

[84] V. Sudhir, M. G. Genoni, J. Lee, and M. S. Kim, *Critical behavior in ultrastrong-coupled oscillators,* Phys. Rev. A **86**, 012316 (2012).

[85] Y. K. Wang and F. T. Hioe, *Phase transition in the dicke model of superradiance,* Phys. Rev. A **7**, 831 (1973).

[86] K. Hepp and E. H. Lieb, *On the superradiant phase transition for molecules in a quantized radiation field: the dicke maser model,* Annals of Physics **76**, 360 (1973).

[87] H. Carmichael, C. Gardiner, and D. Walls, *Higher order corrections to the dicke superradiant phase transition,* Physics Letters A **46**, 47 (1973).

[88] J.-F. Huang, J.-Q. Liao, and L.-M. Kuang, *Ultrastrong jaynes-cummings model,* Phys. Rev. A **101**, 043835 (2020).

[89] P. Forn-Díaz, L. Lamata, E. Rico, J. Kono, and E. Solano, *Ultrastrong coupling regimes of light-matter interaction,* Rev. Mod. Phys. **91**, 025005 (2019).

[90] R. Landig, F. Brennecke, R. Mottl, T. Donner, and T. Esslinger, *Measuring the dynamic structure factor of a quantum gas undergoing a structural phase transition,* Nature Communications **6**, 7046 (2015).

[91] J. Colpa, *Diagonalization of the quadratic boson hamiltonian,* Physica A: Statistical Mechanics and its Applications **93**, 327 (1978).

[92] P. T. Nam, M. Napiórkowski, and J. P. Solovej, *Diagonalization of bosonic quadratic hamiltonians by bogoliubov transformations,* Journal of Functional Analysis **270**, 4340 (2016).

[93] H. Grabert, U. Weiss, and P. Talkner, *Quantum theory of the damped harmonic oscillator,* Zeitschrift für Physik B Condensed Matter **55**, 87 (1984).

[94] R. Karrlein and H. Grabert, *Exact time evolution and master equations for the damped harmonic oscillator,* Phys. Rev. E **55**, 153 (1997).

[95] H. Grabert, U. Weiss, and P. Talkner, *Quantum theory of the damped harmonic oscillator,* Zeitschrift für Physik B Condensed Matter **55**, 87 (1984).

[96] A. Anfossi, P. Giorda, A. Montorsi, and F. Traversa, *Two-point versus multipartite entanglement in quantum phase transitions,* Phys. Rev. Lett. **95**, 056402 (2005).

[97] R. R. P. Singh, M. B. Hastings, A. B. Kallin, and R. G. Melko, *Finite-temperature critical behavior of mutual information,* Phys. Rev. Lett. **106**, 135701 (2011).

[98] F. C. Alcaraz and M. A. Rajabpour, *Universal behavior of the shannon mutual information of critical quantum chains,* Phys. Rev. Lett. **111**, 017201 (2013).

[99] G. De Tomasi, S. Bera, J. H. Bardarson, and F. Pollmann, *Quantum mutual information as a probe for many-body localization,* Phys. Rev. Lett. **118**, 016804 (2017).

[100] C. Walsh, P. Sémon, D. Poulin, G. Sordi, and A.-M. S. Tremblay, *Local entanglement entropy and mutual information across the mott transition in the two-dimensional hubbard model,* Phys. Rev. Lett. **122**, 067203 (2019).

[101] G. Vidal and R. F. Werner, *Computable measure of entanglement,* Phys. Rev. A **65**, 032314 (2002).

[102] V. Y. Shishkov, E. S. Andrianov, A. A. Pukhov, A. P. Vinogradov, and A. A. Lisyansky, *Perturbation theory for lindblad superoperators for interacting open quantum systems,* Phys. Rev. A **102**, 032207 (2020).

[103] C. Müller and T. M. Stace, *Deriving lindblad master equations with keldysh diagrams: Correlated gain and loss in higher order perturbation theory,* Phys. Rev. A **95**, 013847 (2017).

[104] B. M. Villegas-Martínez, F. Soto-Eguibar, and H. M. Moya-Cessa, *Application of perturbation theory to a master equation,* Advances in Mathematical Physics **2016**, 9265039 (2016).

[105] A. Serafini, F. Illuminati, and S. D. Siena, *Symplectic invariants, entropic measures and correlations of gaussian states,* Journal of Physics B: Atomic, Molecular and Optical Physics **37**, L21 (2003).

[106] J. Eisert, S. Scheel, and M. B. Plenio, *Distilling gaussian states with gaussian operations is impossible,* Phys. Rev. Lett. **89**, 137903 (2002).

[107] M. Konopik and E. Lutz, *Quantum response theory for nonequilibrium steady states,* Phys. Rev. Research **1**, 033156 (2019).

[108] H. Nyquist, *Thermal agitation of electric charge in conductors,* Phys. Rev. **32**, 110 (1928).

[109] H. B. Callen and T. A. Welton, *Irreversibility and generalized noise,* Phys. Rev. **83**, 34 (1951).

[110] R. Kubo, *Statistical-mechanical theory of irreversible processes. i. general theory and simple applications to magnetic and conduction problems,* Journal of the Physical Society of Japan **12**, 570 (1957).

[111] P. Hänggi and H. Thomas, *Stochastic processes: Time evolution, symmetries and linear response,* Physics Reports **88**, 207 (1982).

[112] R. Kubo, M. Toda, and N. Hatshitsume, *Statistical Physics II* (Springer, Berlin, 1978).

[113] U. M. B. Marconi, A. Puglisi, L. Rondoni, and A. Vulpiani, *Fluctuation–dissipation: Response theory in statistical physics,* Physics Reports **461**, 111 (2008).

[114] L. F. Cugliandolo, D. S. Dean, and J. Kurchan, *Fluctuation-dissipation theorems and entropy production in relaxational systems,* Phys. Rev. Lett. **79**, 2168 (1997).

[115] G. S. Agarwal, *Fluctuation-dissipation theorems for systems in non-thermal equilibrium and applications,* Zeitschrift für Physik A Hadrons and nuclei **252**, 25 (1972).

[116] L. F. Cugliandolo, J. Kurchan, and G. Parisi, *Off equilibrium dynamics and aging in unfrustrated systems,* J. Phys. I France **4**, 1641 (1994).

[117] T. Speck and U. Seifert, *Restoring a fluctuation-dissipation theorem in a nonequilibrium steady state,* Europhysics Letters (EPL) **74**, 391 (2006).

[118] R. Chetrite, G. Falkovich, and K. Gawedzki, *Fluctuation relations in simple examples of non-equilibrium steady states,* Journal of Statistical Mechanics: Theory and Experiment **2008**, P08005 (2008).

[119] J. Prost, J.-F. Joanny, and J. M. R. Parrondo, *Generalized fluctuation-dissipation theorem for steady-state systems,* Phys. Rev. Lett. **103**, 090601 (2009).

[120] M. Baiesi, C. Maes, and B. Wynants, *Fluctuations and response of nonequilibrium states,* Phys. Rev. Lett. **103**, 010602 (2009).

[121] V. Blickle, T. Speck, C. Lutz, U. Seifert, and C. Bechinger, *Einstein relation generalized to nonequilibrium,* Phys. Rev. Lett. **98**, 210601 (2007).

[122] J. R. Gomez-Solano, A. Petrosyan, S. Ciliberto, R. Chetrite, and K. Gawędzki, *Experimental verification of a modified fluctuation-dissipation relation for a micron-sized particle in a nonequilibrium steady state,* Phys. Rev. Lett. **103**, 040601 (2009).

[123] J. Mehl, V. Blickle, U. Seifert, and C. Bechinger, *Experimental accessibility of generalized fluctuation-dissipation relations for nonequilibrium steady states,* Phys. Rev. E **82**, 032401 (2010).

[124] S. Ciliberto, R. Gomez-Solano, and A. Petrosyan, *Fluctuations, linear response, and currents in out-of-equilibrium systems,* Annual Review of Condensed Matter Physics **4**, 235 (2013).

[125] M. Baiesi and C. Maes, *An update on the nonequilibrium linear response,* New Journal of Physics **15**, 013004 (2013).

[126] J. van Neerven, *The Adjoint of a Semigroup of Linear Operators* (Springer-Verlag, Berlin, 1992).

[127] M. Lax, *Formal theory of quantum fluctuations from a driven state,* Phys. Rev. **129**, 2342 (1963).

[128] P. Talkner, *The failure of the quantum regression hypothesis,* Annals of Physics **167**, 390 (1986).

[129] G. S. Agarwal, *Open quantum markovian systems and the microreversibility,* Zeitschrift für Physik A Hadrons and nuclei **258**, 409 (1973).

[130] A. M. Alhambra and M. P. Woods, *Dynamical maps, quantum detailed balance, and the petz recovery map,* Phys. Rev. A **96**, 022118 (2017).

[131] F. Fagnola and V. Umanità, *Generators of detailed balance quantum markov semigroups,* Infinite Dimensional Analysis, Quantum Probability and Related Topics **10**, 335 (2007).

[132] F. Fagnola and V. Umanita, *Generators of kms symmetric markov semigroups on $\mathscr{B}(h)$ symmetry and quantum detailed balance,* Commun. Math. Phys. **298** (2010).

[133] F. Fagnola and V. Umanità, *Detailed balance, time reversal, and generators of quantum markov semigroups,* Mathematical Notes **84**, 108 (2008).

[134] D. Petz and G. Toth, *The bogoliubov inner product in quantum statistics,* Letters in Mathematical Physics **27**, 205 (1993).

[135] S. Amari and H. Nagaoka, *Methods of information geometry* (Oxford University Press, Oxford, 2000).

[136] T. Shitara and M. Ueda, *Determining the continuous family of quantum fisher information from linear-response theory,* Phys. Rev. A **94**, 062316 (2016).

[137] M. Hayashi, *Two quantum analogues of fisher information from a large deviation viewpoint of quantum estimation,* Journal of Physics A: Mathematical and General **35**, 7689 (2002).

[138] M. Hayashi, *Quantum information: an introduction* (Springer-Verlag, Berlin, 1992).

[139] J. Javanainen, *Oscillatory exchange of atoms between traps containing bose condensates,* Phys. Rev. Lett. **57**, 3164 (1986).

[140] S. Campbell, G. De Chiara, and M. Paternostro, *Equilibration and nonclassicality of a double-well potential,* Scientific Reports **6**, 19730 (2016).

[141] V. K. Arora, *Drift diffusion and einstein relation for electrons in silicon subjected to a high electric field,* Applied Physics Letters **80**, 3763 (2002).

[142] S. Blundell and K. Blundell, *Concepts in Thermal Physics*, oup ed. (Oxford University Press, USA, 2006).

[143] J. des Cloizeaux et al., *Theory of condensed matter* (IAEA, Wien, 1967).

[144] A. Messiah, *Quantum mechanics volume II* (North-Holland Publishing company, Amsterdam, 1965).

[145] J. S. Cohen, H. A. M. Daniëls, and M. Winnink, *On generalizations of the kms-boundary condition,* Commun. Math. Phys. **84** (1982).

[146] R. Haag, N. M. Hugenholtz, and M. Winnink, *On the equilibrium states in quantum statistical mechanics,* Commun. Math. Phys. **5** (1982).

[147] O. Bratteli and D. Kastler, *Relaxing the clustering condition in the derivation of the kms property,* Commun. Math. Phys. **46** (1976).

[148] R. Haag, D. Kastler, and E. Trych-Pohlmeyer, *Stability and equilibrium states,* Commun. Math. Phys. **38** (1974).

[149] W. A. Majewski, *On the relationship between the reversibility of detailed balance conditions,* Ann. Inst.H. Poincare Sec. A(N.S.) **39**, 45 (1983).

[150] W. A. Majewski, *The detailed balance condition in quantum statistical mechanics,* Journal of Mathematical Physics **25**, 614 (1984).

Ausführliche deutsche Zusammenfassung

Diese Arbeit behandelte verschiedene Aspekte der quantenmechanischen Markoff'schen Mastergleichung. Begonnen wurde mit verschiedenen approximativen Ansätzen, deren physikalische Eigenschaften analysiert wurden, besonders deren Fähigkeit das stationäre Verhalten eines beispielhaften Modells zu reproduzieren, das zwar hinreichend simpel ist, damit eine globale Mastergleichung hergeleitet werden kann, jedoch auch hinreichend komplex, damit es kritisches Verhalten aufzeigt. Danach wurden dynamische Eigenschaften allgemeiner Markoff'scher Mastergleichungen betrachtet. Es wurden verschiedene Klassen von Antwortfunktionen hergeleitet, sowie für diese Klassen jeweils ein Fluktuations-Dissipations Theorem.

Kapitel 2 betrachtete die globale und lokale Mastergleichung und verglich deren Fähigkeit kritische Phänomene aufzulösen. Insbesondere wurden stationäre Eigenschaften des Modells zweier harmonischer Oszillatoren, die jeweils an ein thermisches Bad gekoppelt sind, betrachtet. Es wurde gezeigt, dass im Gleichgewichtsfall gleicher Temperaturen der Bäder die globale Mastergleichung in der Lage ist, das analytische Verhalten der Gibbsverteilung für die betrachteten Oszillatorwechselwirkungen fehlerfrei zu reproduzieren. Die lokale Mastergleichung war generell jedoch nicht dazu in der Lage dies fehlerfrei zu tun. Im Falle der 'rotating-wave'-Oszillatorenwechselwirkung versagte die lokale Mastergleichung sogar komplett und konnte kein kritisches Verhalten zeigen. Dieses Versagen ist konsistent mit der Beobachtung dass die lokale Beschreibung generell nichtlokale Eigenschaften wie Quanten-'mutual information', oder Negativität im Falle verschwindender Temperatur, nicht in gleicher Stärke produzieren kann wie die globale Mastergleichung. Im Nichtgleichgewichtsfall verschiedener Temperaturen zwischen den Bädern wurden die beiden Gleichungen mit der Quanten-Langevingleichung verglichen. Während die lokale Gleichung stets stärkere Unterschiede zeigte deren qualitatives Verhalten auch vom Vorzeichen des Temperaturunterschiedes abhängt, so zeigte die globale Mastergleichung nur für die 'rotating-wave'-Wechselwirkung eine völlige Übereinstimmung, die 'position-position'-Wechselwirkung offenbarte Unterschiede.

Während man die lokale Mastergleichung üblicherweise heuristisch angibt, so kann diese im Kontext der Störungsrechnung als nullte Ordnung betrachtet werden. Kapitel 2 ist also nichts anderes als eine Betrachtung der nullten Störungsordnung. In Kapitel 3 wurden nun weitere Ordnungen bestimmt und analysiert, inwiefern sich die stationären Eigenschaften dieser Mastergleichungen verbessern. Primär behandelte dieses Kapitel über die erste Ordnung, die mit der globalen und lokalen Mastergleichung verglichen wurde für das $x - x$-Wechselwirkungsmodell. Werden normale Funktionen um einen Punkt entwickelt, so erwartet man, dass die Beschreibung besser wird mit der Höhe der Ordnung, insbesondere für sehr kleine Variationen. In manchen stationären

Aspekten war dies auch der Fall, insbesondere endliche Temperaturen im thermischen Fall und nichtlokale Observable wie die Quanten-'mutual-information'. Lokale Observablen jedoch, wie der Erwartungswert der Besetzungszahl eines der Oszillatoren, unterschieden sich zwar von dem Ergebnis der lokalen Mastergleichung, die erste Ordnung ist dort jedoch nicht zwangsläufig besser. Interessanterweise zeigte die Betrachtung themodynamischer Eigenschaften, dass sowohl die lokale Mastergleichung als auch die erste Ordnung unphysikalische endliche Wärmeströme aufzeigten im thermischen Fall. Insbesondere der Fall verschwindender Temperaturen zeigte auf, dass die erste Ordnung eine stationäre Dichtematrix erzeugt, die das Unschärfeprinzip verletzt. Im nichtthermischen Fall verschiedener Temperaturen der Bäder wurde gezeigt, dass für Oszillatoren mit verschiedenen Frequenzen ($\omega_1 > \omega_2$) zwei Bereiche existieren, wenn das Bad, das an die höhere Frequenz gekoppelt ist, eine höhere Temperatur besitzt als das andere $\beta_1 < \beta_2$. Zwei Bereiche wurden untersucht. Ein 'normaler' Bereich in dem die höhere Temperatur auch zu einer höheren Besetzungszahl des Oszillators führen würde und ein 'anormaler' Bereich in dem die thermische Besetzung des ersten Oszillators niedriger ist, trotz höherer Temperatur, da für die Bose-Einstein-Verteilung gelten kann, dass $N(\beta_1,\omega_1) < N(\beta_2,\omega_2)$, trotz $\beta_1 < \beta_2$. Interessanterweise wurde gezeigt, dass im normalen Bereich die lokale Mastergleichung besser war, sowohl lokales als auch nichtlokales Verhalten wiederzugeben, im Vergleich zu der ersten Ordnung. Interessanterweise erzeugte die lokale Mastergleichung Wärmeströme und eine Entropieproduktion, die qualitativ das korrekte Verhalten aufzeigten, während die erste Ordnung Unphysikalische besaß. Im anormalen Bereich zeigte sich jedoch, dass die erste Ordnung besser in der Lage war, qualitativ das richtige Verhalten zu reproduzieren, sowohl die Wärmeströme als auch die Entropieproduktion hatten dort das korrekte Vorzeichen, während der lokale Fall Negative aufzeigte. Als Gesamtresult dieser Betrachtungen lässt sich sagen, dass die Versuche die Herleitung einer Mastergleichung mittels Störungsrechnung oder heuristischer Methoden zu umgehen, einen eher mittelwertigen Erfolg erzielen. Auch wenn in manchen Fällen diese Gleichungen in der Lage sind das Verhalten der Globalen qualitativ wiederzugeben, je nach Wahl der Temperaturen, der Intrasystemwechselwirkungsstärke, kleine Veränderungen in der Struktur des Hamiltonoperators und je nach Gleichgewichts- oder Nichtgleichgewichtssituationen, kann deren qualitative oder quantitative Korrektheit unvorhersehbar variieren. Insbesondere zeigte sich dies im Vergleich zwischen der ersten Ordnung und der lokalen Mastergleichung, die die nullte Ordnung wiederspiegelt. Selbst für nur infinitesimale Variationen der Parameter um den Entwicklungspunkt zeigte sich, dass die erste Ordnung nicht zwangsläufig besser ist als die nullte Ordnung. Bei der ebenfalls betrachteten höheren zweiten Ordnung zeigte sich dann, dass diese Störungsordnung nun zu einer starken Verbesserung aller Eigenschaften führte und in einem moderaten Parameterbereich auch besser war, das Verhalten der Globalen zu reproduzieren.

Die letzten beiden Kapitel fokussierten sich dann auf einige dynamische Eigenschaften von allgemeinen quantenmechanischen Mastergleichungen. Kapitel 4 behandelte die quantenmechanische lineare Antworttheorie. Es wurden drei verschiedene Klassen für die Antwortfunktion hergeleitet, auf Grundlage der Ergebnisse einer klassischen Nichtgleichgewichtsbetrachtung [37]. Die erste Klasse beschreibt die Antwortfunktion in einer Form die keine Zeitableitung enthält und direkt abhängig von der ungestörten sta-

tionären Dichtematrix des Systems ist. Aus dieser Klasse geht ebenfalls hervor, im Falle einer Störung der Form eines reinen kommutativen Termes, deren Störung mit dem Hamiltonoperator des Systems kommutiert, dass die Antwort eines thermischen Systems verschwinden wird. Die zweite Klasse hat die Form einer Korrelationsfunktion, die die betrachtete Observable enthält, sowie eine transformierte stochastiche Entropie. Diese Form ist ebenfalls eine Verallgemeinerung der Kubo kanonischen Form [38], für offene Systeme und Nichtgleichgewichtsfälle. Für diese Klasse wurde im Falle einer Mastergleichung, die die 'detailed balance' erfüllt, gezeigt, dass die Antwortfunktion eine bestimmte Symmetrie zwischen verschiedenen Observablen und Störungen enthält. Die dritte Klasse ist die einzige, die nicht explizit von irgendeiner Form von Dichteoperator des Systems abhängt, weder die Ungestörte noch die Gestörte. Für Störungen mit ausschließlich kommutativem Teill, wurde gezeigt, dass die dritte Form als ein Kommutator zwischen der betrachteten Observable und dem Störungshamiltonoperator ausgedrückt werden kann. Für allgemeine Störungen ist der adjungierte Generator der Störung nötig, der auf die Observable wirkt. Diese Klassen wurden dann verglichen mit verschiedenen klassischen und quantenmechanischen Arbeiten. Es wurde des Weiteren betrachtet, welche Symmetrien die Antwortfunktion besitzt, im Falle dass die Mastergleichung Quanten-'detailed balance' erfüllt. Zuletzt wurden verschiedene Beispiele analysiert. Für das Modell der gekoppelten harmonischen Oszillatoren wurden die verschiedenen Bereiche von Gleichgewichts- und Nichtgleichgewichtssituationen betrachtet, für eine Störung in der Form eines reinen Störhamiltonoperators. Im Nichtgleichgewichtsfall wurde das Versagen der 'detailed balance' mittels der Antwortfunktionen aufgezeigt. Ein anderes Beispiel zeigte das Versagen der linearen Antworttheorie in der Nähe eines kritischen Punktes auf. Unabhängig des betrachteten Bereiches, ob Gleichgewicht oder Nichtgleichgewicht, versagte die lineare Antwort konsistent am kritischen Punkt.

Im letzten Kapitel 5 wurden für diese Klassen der Antwortfunktion die zugehörigen Fluktuations-Dissipations Theoreme (FDT) für Quanten-Markoff'sche Mastergleichungen hergeleitet. Dies wird vorbereitet durch eine Wiederholung der klassischen FDT und eine Betrachtung der Definition der klassischen und quantenmechanischen Korrelationfunktionen. Im thermischen, geschlossenen Fall wurde das Fluktuations-Dissipations Theorem wiederholt hergeleitet und die Anzahl der nötigen Eigenschaften für diese Herleitung betont. Insbesondere die KMS-Eigenschaft und die KMS-Randbedingung ist nötig für dessen Herleitung. Für offene, im Allgemeinen nichtthermische, stationäre Systeme gelten diese Eigenschaften nicht. Ausschließlich im Falle geschlossener Systeme, charakterisiert der Hamiltonoperator des Systems die gesamte Dynamik.

Für allgemeine offene Systeme wird die Dynamik auch durch die Badeigenschaften und System-Bad-Wechselwirkung definiert. Deswegen ist zu erwarten, dass die FDT's für diese Systeme nicht zwangsläufig die Form einer Kubo-FDT haben werden. Die FDT für die dritte Klasse kann geschrieben werden als Verknüpfung der Antwortfunktion mit Korrelationsfunktionen im Fourierraum, die multipliziert werden mit Reibungskoeffizienten der Mastergleichung und dadurch den Einfluss der System-Bad Wechselwirkung aufzeigen. Die erste Klasse hat formal bereits die Form einer Korrelationsfunktion und daher ist deren FDT trivial. Die Zeitableitung der zweiten Klasse führt zu einer FDT, die eine lineare Funktion der Fouriervariablen ist, multipliziert mit einer entsprechenden Korrelationsfunktion.

Zusammenfassend wurden in den ersten zwei Kapiteln verschiedene approximative Methoden für Quanten-Mastergleichungen betrachtet und gezeigt, dass diese besonders Probleme haben thermodynamische Eigenschaften von Systemen korrekt zu beschreiben, selbst für das relativ einfache Beispiel zweier gekoppelter harmonischer Oszillatoren. Je nach genauer Parameterwahl, zeigte die erste Störungsordnung der Mastergleichung nur bedingt besseres Verhalten, als die nullte Ordnung. Die Nutzung der Störungsrechnung in der Herleitung der Mastergleichung, muss daher mit Vorsicht benutzt werden, da leicht unphysikalische Gleichungen entstehen können, selbst im Falle infinitesimaler Störungen. Da diese approximativen Ansätze nicht sehr vertrauenswürdig zu sein scheinen, ist es fragwürdig ob Ergebnisse dieser benutzt werden können, um allgemeine Aussagen über ein System treffen zu können. Derartige Gleichungen scheinen eher dazu geeignet, als mathematisches Modell mit freier Parameterwahl zu fungieren, die entsprechend an z.B. ein experimentelles System gefittet werden, solange sie qualitativ bereits erwartbares Verhalten reproduzieren. Die Klassen für die Antwortfunktion der linearen Antworttheorie sind potentiell nutzvoll, um weitere mathematische Eigenschaften der Mastergleichung erforschen zu können, wie es im Falle der Quanten-'detailed-balance' für die Symmetrie der zweiten Klasse der gezeigtu wurde. Die hergeleiteten FDT's mögen potentiell ebenfalls, in Zukunft, als Startpunkt nützlich sein, um eine FDT für allgemeine offene Nichtgleichgewichtssysteme herzuleiten, die noch stärker die Eigenschaften des Systems und die System-Bad Wechselwirkung offenbart.

LIST OF PUBLICATIONS

1. M. Konopik. A. Friedenberger and E. Lutz, *Nonequilibrium information erasure below kTln2*, EPL **131** 60004 (2020)

2. M. Konopik and E. Lutz, *Quantum response theory for nonequilibrium steady states*, Phys. Rev. Research **1**,3 (2019)

3. M. Konopik and E. Lutz, *Local master equations may fail to describe dissipative critical behavior*, arXiv preprint arXiv:2012.09907 (2020)

4. M. Konopik, T. Korten, E. Lutz and H. Linke, *Fundamental energy cost of finite-time computing*, arXiv preprint arXiv:2101.07075 (2021)

5. M. Rademacher, M. Konopik, M. Debiossac, D. Grass, E. Lutz and N. Kiesel, *Nonequilibrium control of thermal and mechanical changes in a levitated system*, arXiv preprint arXiv: 2103.10898 (2021)

Curriculum Vitæ

Michael Konopik

Persönliche Daten

Geburtsdatum	19.11.1991
Geburtsort	Nürnberg

Ausbildung

1998-2002	**Bertleinschule, Lauf a. d. Pegnitz**
2002-2008	**Staatliche Realschule Röthenbach, Röthenbach** Mittlere Reife (mit Auszeichnung)
2008-2010	**Lothar-von-Faber-Schule, Nürnberg** Fachhochschulreife (mit Auszeichnung)
2010-2011	**Staatliche Fachoberschule, Erlangen** Fachgebundene Hochschulreife
2011-2015	**B.SC. in Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg** Bachelorarbeit: Nonequilibrium Landauer principle Abschlussnote 1,19 (mit Auszeichnung)
2015-2017	**M. SC. in Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg** Masterarbeit: Energetic cost of finite time information erasure and parallel computation Abschlussnote 1,04 (mit Auszeichnung)
2017-2018	**Promotionsstudium, Friedrich-Alexander-Universität Erlangen-Nürnberg** Beginn des Promotionsstudium unter Prof. Eric Lutz am Institut für Theoretische Physik 2, mit Wechsel der Forschungsgruppe and die Universität Stuttgart

2018-2021 **Promotion, Universität Stuttgart**
Abschluss der Promotion am 1. Institut für Theoretische Physik
Doktorarbeit: Nonequilibrium steady-state physics with quantum master equations

BERUFLICHE TÄTIGKEITEN

2014-2017 **Tutor, Friedrich-Alexander-Universität Erlangen-Nürnberg**
verschiedne Tutorenstellen im Fachbereich Mathematik und Physik

03-05/2017 **Wissenschaftlicher Mitarbeiter, Universität Wien**
Experimentelles Projekt in der START Gruppe um Prof. Nikolai Kiesel

2017-2018 **Wissenschaftlicher Mitarbeiter, Friedrich-Alexander-Universität Erlangen-Nürnberg**
Arbeit am Institut für Theoretische Physik 2
Forschung, Administration, Erstellung von Übungenblättern und Übungsgruppenleitung

2018-2021 **Wissenschaftlicher Mitarbeiter, Universität Stuttgart**
Arbeit am 1. Institut für Theoretischen
Forschung, Administration, Erstellung von Übungenblättern, Übungsgruppenleitung, Betreuung von Bachelorarbeiten

WISSENSCHAFTLICHE VORTRÄGE UND KONFERENZEN

- II Workshop on quantum information and thermodynamics 2019, Natal, Brasilien, Vortrag
- Quantum ThermoDynamics conference 2019, Espoo, Finnland, Vortrag
- FOR2724 Kick-off meeting 2019, Berlin, Deutschland, Postervortrag
- Quantm Thermodynamics for Young Scientists 2020, Bad Honnef, Deutschland, Postervortrag

Eidesstattliche Erklärung

Ich versichere, die vorliegende Arbeit selbstständig und lediglich unter Benutzung der angegebenen Quellen und Hilfsmittel verfasst zu haben. Ich erkläre weiterhin, dass die vorliegende Arbeit noch nicht im Rahmen eines anderen Prüfungsverfahrens eingereicht wurde.

Gez. Michael Konopik Stuttgart 25.06.2021

ACKNOWLEDGEMENTS

First I want to thank my supervisor Prof. Dr. Eric Lutz for the opportunity to pursue my ideas and enabled me to understand the groundworks of physical practice. I also want to thank Prof. Dr. Nikolai Kiesel for the opportunity to work for a small project in a lab, giving me the opportunity to understand the experimental side of physical research. From Erlangen I want to thank my former colleagues Alexander, Paul, Andreas, Benedikt for cool evening events. From Stuttgart I want to thank Johannes, Jörg and all people of the Main group for the lunch times and coffee break discussions. I further want to thank Arthur, Franklin and Sinan for a nice time in Stuttgart and Kaonan Micadei for many delightful conversations about many different philosophical topics.

I especially want to thank Tobias Denzler for a great time and I am grateful for doing the PHD concurrently. Being able to attend various conferences together was delightful and a lot of fun.

And lastly I want to thank my family to help through these sometimes stressful times and allowing me to pursue my goals.