

ABSTRACT

Title of dissertation: Nonequilibrium Quantum Systems:
 Fluctuations and Interactions

Yiğit Subaşı

Dissertation directed by: Professor Bei Lok Hu
 Department of Physics

We explore some aspects of nonequilibrium statistical mechanics of classical and quantum systems. Two chapters are devoted to fluctuation theorems which were originally derived for classical systems. The main challenge in formulating them in quantum mechanics is the fact that fundamental quantities of interest, like work, are defined via the classical concept of a phase space trajectory. We utilize the *decoherent histories* conceptual framework, in which classical trajectories emerge in quantum mechanics as a result of coarse graining, and provide a first-principles analysis of the nonequilibrium work relation of Jarzynski and Crooks's fluctuation theorem for a quantum system interacting with a general environment based on the *quantum Brownian motion* (QBM) model. We indicate a parameter range at low temperatures where the theorems might fail in their original form.

Fluctuation theorems of Jarzynski and Crooks for systems obeying classical Hamiltonian dynamics are derived under the assumption that the initial conditions are sampled from a canonical ensemble, even though the equilibrium state of an isolated system is typically associated with the microcanonical ensemble. We address

this issue through an exact analysis of the *classical Brownian motion* model. We argue that a stronger form of *ensemble equivalence* than usually discussed in equilibrium statistical mechanics is required for these theorems to hold in the infinite environment limit irrespective of the ensemble used, and proceed to prove it for this model. An exact expression for the probability distribution of work is obtained for finite environments.

Intuitively one expects a system to relax to an equilibrium state when brought into contact with a thermal environment. Yet it is important to have rigorous results that provide conditions for equilibration and characterize the equilibrium state. We consider the dynamics of open quantum systems using the Langevin and master equations and rigorously show that under fairly general conditions quantum systems interacting with a heat bath relax to the equilibrium state defined as the reduced thermal state of the system plus environment, even in the strong coupling regime. Our proof is valid to second-order in interaction strength for general systems and exact for the linear QBM model, for which we also show the equivalence of multi-time correlations.

In the final chapter we give a sampling of our investigations into *macroscopic quantum phenomena*. We work out in detail a specific example of how and under what conditions the center of mass (CoM) coordinate of a macroscopic object emerges as the relevant degree of freedom. Interaction patterns are studied in terms of the couplings they induce between the CoM and relative coordinates of two macroscopic objects. We discuss the implications of these interaction patterns on macroscopic entanglement.

Nonequilibrium Quantum Systems: Fluctuations and Interactions

by

Yiğit Subaşı

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Advisory Committee:

Professor Bei Lok Hu, Chair/Advisor

Professor Theodore L. Einstein

Professor Christopher Jarzynski

Professor Theodore R. Kirkpatrick

Professor Victor Yakovenko

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Preface

This thesis represents the majority of my research in the past seven years under the supervision of Prof. Bei Lok Hu.

Chapter 2 is based on the publication “The equilibrium states of open quantum systems in the strong coupling regime”, Y. Subaşı, C. H. Fleming, J. M. Taylor, and B. L. Hu, *Phys. Rev. E*, **86**, 061132 (2012). Chris Fleming, who was my main collaborator in this publication, did the perturbative analysis for general systems whereas I did the derivation for the linear systems. Writing of the manuscript was a joint effort.

Chapter 3 is based on the publication “Quantum and classical fluctuation theorems from a decoherent histories, open-system analysis”, Y.S. and B. L. Hu, *Phys. Rev. E*, **85**, 011112 (2012). The calculations and bulk of the analysis in this work is done by me. The manuscript is written together with Prof. Hu.

Chapter 4 is based on the publication “Microcanonical work and fluctuation relations for an open system: an exactly solvable model”, Y.S. and C. Jarzynski, *Phys. Rev. E*, **88**, 042136 (2013). The calculations and bulk of the analysis in this work is done by me. The manuscript is written together with Prof. Jarzynski.

Chapter 5 is based on a number of publications. Most of the material is from two papers: “Pathways toward understanding Macroscopic Quantum Phenomena”, B. L. Hu, and Y.S., *J. Phys.: Conf. Ser.* **442**, 012010 (2013). and “Macroscopic Quantum Phenomena from the Coupling Pattern and Entanglement Structure Perspective”, C. H. Chou, Y.S. and B. L. Hu, [arXiv:1308.4225](https://arxiv.org/abs/1308.4225). The original material

presented in this chapter is my own contribution to those papers. The discussion and analysis is a joint effort with B. L. Hu and C. H. Chou. Also the summary of two other papers is given in the introduction of this chapter: “Macroscopic Quantum Phenomena from the Correlation, Coupling and Criticality Perspectives”, C. H. Chou, B. L. Hu, and Y.S., *J. Phys.: Conf. Ser.* **330**, 012003 (2011). and “Macroscopic quantum phenomena from the large N perspective”, C. H. Chou, B. L. Hu, and Y.S., *J. Phys.: Conf. Ser.* **306**, 012002 (2011).

*To my beautiful wife Shannon,
For her unwavering support and love.*

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List of Abbreviations

QBM	Quantum Brownian Motion
NHO	N Harmonic Oscillators
QFT	Quantum Fluctuation Theorem
FT	Fluctuation Theorem
MQP	Macroscopic Quantum Phenomena
OQS	Open Quantum System
IF	Influence Functional
BEC	Bose-Einstein Condensate
QSM	Quantum Statistical Mechanics
CoM	Center of Mass
FDR	Fluctuation Dissipation Relation
KMS	Kubo, Martin and Schwinger
<i>dechis</i>	decoherent histories
<i>envdec</i>	environment-induced decoherence
\mathcal{H}	Hilbert space
$\beta = 1/k_B T$	inverse temperature
\mathcal{W}	Wigner function
\mathbf{f}	classical phase space density
$Z(\beta)$	partition function
$Z[j(\cdot)]$	generating functional
F	free energy
f	external driving force
ξ	noise
ν	noise kernel
γ	damping kernel
μ	dissipation kernel
ν_r	Matsubara frequencies
W	work
$P(W)$	probability distribution of work
$P_\xi[\xi]$	probability distribution of noise
Σ	density of states
σ	precision of trajectory
σ_W	standard deviation of work
B_n	n'th Bernoulli number

Chapter 1: Introduction

1.1 Motivation

Statistical Mechanics. It is generally accepted that all the physics at the microscopic level is governed by quantum mechanics. At the macroscopic level we encounter a completely different world with a rich variety of phenomena. For instance elements occur in different phases with qualitatively different properties and transitions between phases occur although the underlying microscopic theory is unchanged. The nature of the phenomena is also very different. In microscopic theory the dynamics is reversible whereas at the macroscopic level irreversibility is far more common. Even the objects of study of micro and macro physics are different; quantities of interest to macrophysics like temperature, entropy, heat etc. and events like phase transitions do not have *direct analogues* in microphysics. These macroscopic phenomena emerge from the complexity of the underlying microphysics due to the large number of constituents. Although the large number of constituents makes a straightforward application of the underlying Schrödinger equation impractical, it makes a statistical description of macroscopic objects feasible. The aim of statistical physics is to study complex systems composed of many degrees of freedom using the tools of probability theory and statistics and provide a bridge between micro and

macro scales.

The most developed branch of statistical physics is equilibrium statistical mechanics. It provides a unifying framework in which various results can be interpreted and is extremely successful in describing a vast range of phenomena [1]. In equilibrium statistical mechanics systems are described by various ensembles, like canonical, microcanonical and grand canonical depending on the macroscopic constraints. These ensembles assign a probability to microstates of the system consistent with the observed macrostate. The central object of study of equilibrium statistical mechanics is the partition function, from which various quantities of physical interest can be derived by differentiation. This universal recipe can be applied to any equilibrium system, barring technical difficulties. The success of this approach led to its application to various fields outside physics like biology and economics [2].

Nonequilibrium Statistical Mechanics. Even though equilibrium statistical physics is familiar and powerful, nonequilibrium phenomena are ubiquitous in nature. Any transition between two equilibrium states that happens in finite time necessarily involves nonequilibrium states. Moreover, physical phenomena of great theoretical as well as practical importance like heat transfer (Fourier Law [3], diffusion, dissipation etc.) are inherently nonequilibrium. Nonequilibrium also plays a very important role in biological systems since most processes in cells occur under nonequilibrium circumstances which can mostly be approximated by steady states [4].

Unlike equilibrium statistical mechanics, where the ensemble approach and partition function methods provide a recipe to study a wide variety of systems,

there is no prescription for nonequilibrium phenomena of comparable generality. Moreover, some of the trusted methods of equilibrium theory do fail under nonequilibrium conditions, among which one can mention divergences in the virial expansion for the calculation of transport coefficients of dense gases [5–8].

A large number of results in this field are obtained for individual models. Although solvable models play an important role in our understanding, the ultimate goal is to place them all in a unifying framework. Recently there have been serious attempts to achieve this goal for a restricted class of states, i.e. nonequilibrium steady states. These are nonequilibrium states in which the parameters that describe the state do not change in time. This raises the possibility to describe such systems via nonequilibrium ensembles, which are meant to be generalizations of equilibrium ensembles. [9, 10] It is desirable to have general physical principles to motivate these ensembles similar to the use of *the principle of equal a priori probabilities* together with conservation laws in the construction of microcanonical ensemble. Nonequilibrium steady states are also interesting because they exhibit long-range correlations [6, 7].

Fluctuation Theorems. Fluctuations at or near equilibrium have been relatively well understood. The famous fluctuation-dissipation relation, most often proven within the linear response regime, relates the response of the system away from equilibrium to the properties of fluctuations at equilibrium. An important set of results in linear response theory are the Green-Kubo relations which relate transport coefficients to integrals of two-time correlation functions. Thus equilibrium fluctuations contain some information about behavior under nonequilibrium

conditions. It is then reasonable to ask whether nonequilibrium fluctuations, far from equilibrium, also contain meaningful information. The answer turns out to be yes and results obtained in this area are gathered under the name “fluctuation theorems”.

There are few theoretical results valid for systems far away from equilibrium, and fluctuation theorems belong to this group. This is a vast topic, and there are numerous theorems applying to various nonequilibrium set-ups. For instance, transient fluctuation theorems are applicable to systems driven away from an initial state of equilibrium. The nonequilibrium work theorem of Jarzynski [11, 12] (Eq. 4.20) and Crooks’s fluctuation theorem [13–15] (Eq. 1.47), which deal with work fluctuations and play an important role in this dissertation, fall into this category. The former is an equality which relates the free energy difference between two equilibrium states to the ensemble average of the exponentiated negative work done on the system as it is driven arbitrarily far from equilibrium, and it can be derived from the latter. Steady state fluctuation theorems, like that of Gallavotti, Evans and Cohen [16–20] dealing with fluctuations in entropy production, concern nonequilibrium steady states. This theorem expresses a symmetry of the probability distribution of entropy production, by relating the probabilities of entropy generation and annihilation of equal amount [20]. The relationships between these various theorems have been studied. Close to equilibrium, fluctuation theorems can be used to recover linear response results [21]. Far away from equilibrium they provide us with valuable theoretical tools where most of the standard results of statistical mechanics are no longer applicable.

Small Systems. Thermodynamics describes physical processes at the macroscopic scale. For macroscopic systems fluctuations away from the expected behavior are often suppressed due to the law of large numbers. Ensemble equivalence follows from this “rule”, whereby the relative fluctuations of thermodynamic parameters that are not held fixed vanish. Phase transitions offer an important exception to this “rule” within thermodynamics. Second-order phase transitions are accompanied by diverging susceptibilities which in turn require there being long-range correlations inside matter. The fact that phase transitions can only occur (strictly speaking) in the limit of $N \rightarrow \infty$ $V \rightarrow \infty$ with $N/V = \text{fixed}$ is yet another way in which they exemplify deviations from the “rule” that fluctuations are suppressed in large systems.

With the recent advances in nanotechnology, it has become possible to make real-time observations of small systems, and even design and build structures and machines at the nano scale. For small systems fluctuations are both relatively larger and more frequent such that the standard thermodynamic description is not adequate [22]. Developing a “thermodynamics” of small systems is important both because of the need to explain the physics observed at small scales, but also to guide the engineers in their designs of nano devices like molecular motors [4]. Yet, it is not even clear whether a thermodynamical description of very small objects is possible. Fluctuation theorems are very useful at this frontier, since they can address large fluctuations under nonequilibrium conditions.

Small systems have some similarities to systems with long-range interactions [23, 24]. Whenever the size of the small system is comparable to the range of interactions

the energy becomes non-additive. In the absence of a thermodynamical limit, non-additivity can occur even for short-ranged interactions as long as the system size is comparable to the range of interactions.¹ Additivity is essential in the derivation of the canonical ensemble, and its absence manifests itself in surprising ways in systems with long-range interactions like inequivalence of ensembles, negative specific heat and susceptibilities, which we might expect to see in small systems as well (to the extent that the non-additivity is at the source of these effects). Although mathematically speaking phase transitions require the thermodynamical limit, they are also observed in small systems like clusters of atoms etc. which need to be considered from a more general point of view [23]. This suggests that phase transitions in small systems may show anomalies similar to those in systems with long-range interactions.

Strong coupling. Besides the lack of large number statistics, fluctuations are important in small systems due to the strong influence of the environment. Unlike macroscopic systems with short-range interactions, the interaction energy can become comparable to the system energy in small systems with any type of interactions. This translates into a strong influence by the environment in terms of dissipation and noise, which in turn induces strong fluctuations in the system dynamics. In order to describe this influence correctly we need an approach that fully incorporates the dynamic interplay between system and environment. The open quantum systems paradigm, which we will describe soon, meets this need.

¹In contrast, for long-range interactions non-additivity persists even in the thermodynamical limit.

An important aspect of the canonical distribution of equilibrium statistical mechanics is its simplicity. For instance the entire effect of the thermal bath on the system is encoded in one parameter, the temperature. The nature of the bath or the nature of the coupling between the system and the bath do not matter. This simplicity is a direct consequence of the weak coupling assumption. In the presence of non-vanishing coupling, temperature is not sufficient to specify the influence of the thermal bath on the equilibrium phase space distribution and the dynamics of the system. The effect on the phase space distribution is captured by the “Hamiltonian of mean force” [A.3](#) which replaces the isolated system Hamiltonian in the canonical distribution and depends on the details of the bath and the system-bath coupling. The dynamics of the system is also modified accordingly with the bath causing dissipation and decoherence in the quantum case. These effects can be adequately studied within the open systems framework which we will return to briefly.

The difficulty of dealing with small systems from the classical perspective is furthered by the fact that below a certain “size” quantum coherence needs to be included in ones consideration. Decoherence, the mechanism responsible for the suppression of quantum coherence, is, as a general rule, stronger on large scales and high temperatures, and quantum effects are more dominant at low temperatures and small scales. Yet under special circumstances they can survive at large scales and high temperatures as well. We will analyse this possibility closer when we talk about macroscopic quantum phenomena (MQP) later. Some quantum features are merely quantitative deviations from the expectations based on classical mechanics. Others involve qualitative effects that can not be accounted for at all in the paradigm of

classical mechanics. Among these effects *entanglement* is arguably the one most unique to quantum mechanics. [25]

Quantum Dissipative Systems. Most physical systems we encounter in nature as well as in the laboratory are not perfectly isolated from their surroundings. They display dissipative dynamics, whereby energy and other conserved quantities can be exchanged between the system and its environment. Such systems can be phenomenologically modelled in classical mechanics by going beyond the Hamiltonian formalism, for instance by adding a phenomenological friction and noise terms into the equations of motion of the system. Such additions to the dynamical rule do not in general violate any fundamental laws of classical mechanics. This is not true in quantum mechanics.

The standard formulation of quantum mechanics relies on Hamiltonians for the canonical quantization scheme. However, Hamiltonian systems cannot describe dissipative dynamics. Addition of phenomenological friction and noise terms is not as straightforward as in classical mechanics because quantum mechanics puts numerous restrictions (for instance via the uncertainty principle) on the types of dynamics allowed. Thus one has to be much more careful in describing dissipative dynamics in quantum mechanics.

Open Quantum Systems. There are two common approaches to overcome this difficulty. One approach is to resort to alternative quantization schemes. The other is through open quantum systems (OQS) paradigm [26–31], which is the approach used in this dissertation. In this approach one applies standard quantum mechanics to a closed system composed of the system of interest and an environment it interacts

with. When the environmental degrees of freedom are coarse grained, or “integrated over”, the resulting open system dynamics displays dissipation, fluctuations and decoherence in a way that respects all the rules of quantum mechanics by construction. The effect of the environment on the system is called the “back-action” and its inclusion in the open system dynamics requires careful treatment. The influence functional (IF) approach we use includes back-action in a self-consistent manner, which ensures the resultant open system dynamics satisfies basic conditions like positivity of the density matrix and the fluctuation dissipation relation.

The use of a microphysics model such as the quantum Brownian motion (QBM) model could provide a rigorous basis for any phenomenological description. It makes explicit any assumption made in the phenomenological models which enables one to clearly define the range of validity of the results derived from each model, as well as being able to provide the details in the derivations with or without these assumptions. Applying methods of nonequilibrium statistical mechanics such as the Zwanzig-Mori-Nakajima projection operator or the Feynman-Vernon influence functional formalism [26] to a microscopic model consummates the objectives of quantum open-system treatment. Using these methods one obtains a description of the open-system dynamics in terms of open system variables alone. With the back-action of the environment taken into account in a self-consistent manner, the dynamics of the open system will in general be non-Markovian as it contains memories, and the noise in the environment is generally colored, as it contains many time scales characterized by its spectral density and varies with temperature. Dissipation in the open system dynamics is controlled and balanced by the noise in

the environment as manifested in the existence of fluctuation-dissipation relations between these two sectors. In the QBM model, the deterministic component of the back-action is dissipation and the stochastic component is the noise, which is also responsible for decoherence.

Environment related quantities like heat can also be addressed within this framework. The IF is one of the key methods we use, since it has the advantage of including the back-action in a self-consistent manner and one can invoke field theory techniques (by way of the almost equivalent Schwinger-Keldysh closed time path formalism) to address nonequilibrium statistical mechanics issues. Moreover, the dissipative and decoherent effects due to the environment are neatly separated in this method, which allows us to draw conclusions about quantum-classical transition such as encountered in decoherent histories formalism.

Equilibration. At the intersection of equilibrium and nonequilibrium lies the process of equilibration which is closely related to the problem of dissipation and irreversibility. Recently, equilibration in quantum systems has attracted a lot of attention [32–37]. Unlike classical mechanics, where chaotic molecular dynamics provides an explanation for the emergence of irreversibility from underlying reversible dynamics [7], quantum dynamics is unitary and consequently arguments used in classical mechanics based on chaotic dynamics do not directly apply to quantum systems.² The quantum open systems paradigm described above provides a useful framework in which we studied the equilibrium states and the process of equilibra-

²Quantum chaos, which is quite different from the classical chaos, may one day play a similar role in quantum systems.

tion beyond weak coupling approximation.

Macroscopic Quantum Phenomena. We mentioned that in small systems quantum mechanical effects might become important. This statement implicates a belief that quantum effects are absent at macroscopic systems. Indeed, the traditional view is that only “small” objects necessitate a quantum description, while classical mechanics, a limiting case of quantum mechanics, is sufficient for the description of the macro world. In the face of new challenges from macroscopic quantum phenomena (MQP), viz, quantum features occurring in objects of “large” sizes, systems with many components or degrees of freedom, organized in some ways where they can be identified as macroscopic objects, this common belief now requires a much closer scrutiny, involving possible reformulations and/or reinterpretations. Faced with the challenge of MQP, even a naive and seemingly unequivocal notion, like what is meant by “macroscopic”, needs to be reconsidered.

This emerging field is ushered in by several categories of definitive experiments. A common example of MQP is superconductivity, where the Cooper pairs can extend to very large scales compared to interatomic distances, and Bose-Einstein condensate (BEC), where in the N-body ground state, a finite fraction of atoms occupy the same quantum state. Other important examples are in nanoelectromechanical devices [38–40], where the center of mass of a macroscopic object, the cantilever, is seen to obey a quantum mechanical equation of motion. Experimental proposals to detect the superposition between a mirror and the quantum field, and between two mirrors, have been proposed [41, 42] while the interference pattern formed when a large object composed of C^{60} molecules passing through two slits have been observed [43].

Likewise for experiments in quantum optomechanics, see e.g., [44, 45].

By contrast, this new field, which is rich in open issues at the foundation of quantum and statistical physics, remains little explored theoretically (with the important exception of the work of Leggett [46]). At first sight it might appear that MQP should be already covered under quantum statistical mechanics (QSM), however a close inspection reveals that the role played by quantum mechanics in QSM is rather limited. Only energy levels are used in the construction of the partition function which do not carry any information about the quantum coherence of the corresponding quantum states. The “quantum” in QSM also refers to effects due to spin-statistics, in the difference between bosons and fermions, and distinguishability: different combinatorics in distinguishable (classical feature) versus identical (quantum feature) particles. Quantum features over and beyond those contained in particle spin statistics and energy levels, such as large scale quantum coherence and entanglement, is what the “quantumness” in MQP highlights. QSM, although perfectly valid in its domain of validity, is limited in scope to mostly thermodynamic quantities. For example, based on the standard QSM treatment we would not be able to conclude that two BECs could interfere. The coherence properties necessary for this effect are not contained in the limited information entering the partition function and thus cannot be recovered from it.

In the following sections we provide some of the background material essential to the understanding of later chapters. We conclude this chapter with an outline of the dissertation and a summary of the contributions of this work to the field.

1.2 Quantum Mechanics - A Brief Review

First we briefly review some basic formulations of quantum mechanics that will be used in the analysis of QBM model.

1.2.1 Density Matrix Formalism

The most common formulation of quantum mechanics is in terms of pure states (or kets) $|\psi\rangle$, which are vectors living in a Hilbert space \mathcal{H} . In the conventional interpretation of quantum mechanics, the kets represent states which are known as well as is allowed by quantum mechanics [1]. In this sense, pure states correspond to *micro-states* of classical mechanics.³ The dynamics of the pure quantum states are governed by the Schrödinger Equation:

$$i\hbar \frac{\partial}{\partial t} |\psi\rangle = \mathbf{H} |\psi\rangle, \quad (1.1)$$

where \mathbf{H} is the Hamiltonian operator.

The concept of *micro-states* is not useful in describing macroscopic systems, where our knowledge of the system is incomplete. In statistical mechanics a macroscopic system is usually described by a *macro-state*, which is a probability distribution over the *micro-states* the system may in reality be in. This leads to a description

³An important difference between classical and quantum mechanical *micro-states* is that classical micro-states specify the state completely, with no probabilistic element whereas pure quantum states have probabilities built into them.

of the state of the system in terms of a density matrix (also called density operator)

$$\boldsymbol{\rho} = \sum_i p_i |\psi_i\rangle \langle \psi_i|, \quad (1.2)$$

where p_i is the probability that the *micro-state* is $|\psi_i\rangle$. The dynamics of the density matrix follows from that of the kets (1.1):

$$i\hbar \frac{d\boldsymbol{\rho}}{dt} = [\mathbf{H}, \boldsymbol{\rho}] \quad (1.3)$$

Observables are described by operators in quantum mechanics. The expectation value of an observable associated with an operator $\boldsymbol{\mathcal{O}}$ is given by:

$$\langle \boldsymbol{\mathcal{O}} \rangle = \text{Tr}[\boldsymbol{\rho} \boldsymbol{\mathcal{O}}] \quad (1.4)$$

1.2.2 Wigner Representation

It is not easy to establish a correspondence between classical and quantum mechanics using the density matrix representation described in the previous section. In classical mechanics phase space plays a central role, whereas the density matrix does not resemble anything like a phase space distribution. The Wigner representation is arguably the description of quantum systems that most resembles the classical phase space [47].

Consider a system of N particles. Let $\mathbf{x} = (\mathbf{x}_1, \dots, \mathbf{x}_N)$ and $\mathbf{p} = (\mathbf{p}_1, \dots, \mathbf{p}_N)$ be the position and momentum operators, respectively. Any operator $\boldsymbol{\mathcal{O}}$ can be

written as a linear combination of the following form [1]:

$$\mathcal{O} = \int dx dp \mathcal{O}_W(x, p) \Theta(x, p), \quad (1.5)$$

$$\Theta(x, p) = \left(\frac{1}{4\pi^2} \right)^N \int d\alpha d\beta e^{i\alpha \cdot (\mathbf{x} - x) + i\beta \cdot (\mathbf{p} - p)}, \quad (1.6)$$

$$\mathcal{O}_W(x, p) = (2\pi\hbar)^N \text{Tr}[\mathcal{O} \Theta(x, p)]. \quad (1.7)$$

In the above we used $x = (x_1, \dots, x_N)$, $p = (p_1, \dots, p_N)$, $\mathbf{x} = (\mathbf{x}_1, \dots, \mathbf{x}_N)$, $\mathbf{p} = (\mathbf{p}_1, \dots, \mathbf{p}_N)$ and “ \cdot ” indicates the inner product between vectors. In this section we use bold symbols exclusively for quantum mechanical operators to avoid confusion. Eqs. (1.5-1.7) define the *Wigner representation*. Being an operator, the density matrix also admits a representation of the form (1.7), which is referred to as the Wigner function:

$$\mathcal{W}(x, p) = (2\pi\hbar)^N \text{Tr}[\rho \Theta(x, p)] = \int dy e^{\frac{i}{\hbar} p \cdot y} \rho(x - y/2, x + y/2, t). \quad (1.8)$$

Quantum mechanical expectation values can be calculated in the Wigner representation as:

$$\langle \mathcal{O} \rangle = \frac{1}{(2\pi\hbar)^N} \int dx dp \mathcal{W}(x, p) \mathcal{O}_W(x, p) \quad (1.9)$$

Because of its appearance in phase space variables it, is often said that the Wigner function is the quantum correspondence of the classical phase space density [48], and the peak of the Wigner function coincides with the classical trajectory in phase space. This is an erroneous statement. The Wigner function is by construction equivalent to the density matrix and as such gives a complete quantum mechanical description of the *macro-state* of the system. For instance, although the density

matrix is a positive operator, the corresponding Wigner function can take negative values, which makes its interpretation as a real probability distribution impossible. That's why it is referred to as a quasi-probability distribution. Even in cases where the Wigner function is positive-definite everywhere in phase space, it should not be thought of as a classical phase space density [48, 49]. When we study the dynamics of the Wigner function later in this chapter we will encounter further differences between the classical phase space density and the Wigner function.

Under special conditions for Gaussian systems such as a free simple harmonic oscillator (closed system) or one which interacts bilinearly with an ohmic bath at high temperature (an open system), the Wigner function remains positive definite for all times. The quantum and classical dynamics have the same form in the equations of motion [50]. For more general conditions by including environmental influence the reduced Wigner function, defined in Sec. 1.3, may become positive definite at late times after the system has sufficiently been decohered. This can indeed be used as a criterion for the appearance of classicality. We will comment more on this issue in Chapter 3 when we study quantum fluctuation theorems using the decoherent histories conceptual framework.

1.2.3 Dynamics of the Wigner Function

If the dynamics of the quantum system is governed by a Hamiltonian as in (1.1), the corresponding dynamics of the Wigner function is governed by the Moyal

equation:

$$\frac{\partial \mathcal{W}(x, p; t)}{\partial t} = \{\{\mathbf{H}(x, p; t), \mathcal{W}(x, p; t)\}\}, \quad (1.10)$$

where the Moyal bracket is defined as:

$$\{\{g, h\}\} \equiv \frac{2}{\hbar} g(x, p) \sin \left(\frac{\hbar}{2} \sum_{i=1}^N \left(\overleftarrow{\partial}_{x_i} \overrightarrow{\partial}_{p_i} - \overleftarrow{\partial}_{p_i} \overrightarrow{\partial}_{x_i} \right) \right) h(x, p) \quad (1.11)$$

and the arrows indicate the direction the partial derivatives act. The Moyal bracket is a generalization of the Poisson bracket and reduces to it in the limit $\hbar \rightarrow 0$.

$$\{g, h\} \equiv \lim_{\hbar \rightarrow 0} \{\{g, h\}\} = \sum_{i=1}^N \left(\frac{\partial g}{\partial x_i} \frac{\partial h}{\partial p_i} - \frac{\partial g}{\partial p_i} \frac{\partial h}{\partial x_i} \right) \quad (1.12)$$

Eq. (1.10) is the quantum generalization of the Liouville equation, which determines the dynamics of the classical phase space density $f(x, p)$:

$$\frac{\partial f(x, p; t)}{\partial t} = \{H(x, p; t), f(x, p; t)\} \quad (1.13)$$

The Moyal equation (1.10) allows us to have an expansion in term of \hbar , thus identifying the corrections to classical dynamics due to quantum mechanics.

A special case, very relevant to the content of this dissertation, is that of linear systems. By linear systems we mean systems whose dynamics is governed by a Hamiltonian that is at most quadratic in phase space variables. For example: N harmonic oscillators that are coupled amongst each other via further harmonic potentials. For such systems it is easy to see from Eqs. (1.10, 1.11) that the terms of $O(\hbar)$ vanish identically. This means that the Moyal bracket when acting on an arbitrary Wigner function and a linear Hamiltonian, reduces to the classical Poisson bracket. This, on the other hand, means that the quantum dynamics of

a Wigner function generated by a linear Hamiltonian is identical to the classical dynamics, generated by the same Hamiltonian, of a phase space density. Thus, we can directly apply our understanding of the classical dynamics of linear systems to the dynamics of quantum systems. In the rest of this dissertation we will use statements like: “*for linear systems quantum and classical dynamics are identical*” to refer to this correspondence. The Wigner representation proves especially useful for linear systems exactly for this reason.

A complete description of a quantum system requires the specification of the Hamiltonian as well as the initial state. For linear systems all the quantum mechanical effects are contained in the initial state, because the quantum dynamics are identical to the classical dynamics. As an example, consider the thermal state of a harmonic oscillator with frequency ω at inverse temperature $\beta = 1/k_B T$ in classical and quantum mechanics:

$$\mathcal{W}(x, p) = \frac{2}{\coth(\beta\hbar\omega/2)} e^{-\frac{H_{\text{HO}}(x, p)}{\coth(\beta\hbar\omega/2)\hbar\omega/2}}, \quad (1.14)$$

$$\mathbf{f}(x, p) = \lim_{\hbar \rightarrow 0} \mathcal{W}(x, p) = \beta\hbar\omega e^{-\beta H_{\text{HO}}(x, p)} = \frac{e^{-\beta H_{\text{HO}}(x, p)}}{Z_{\text{HO}}(\beta)}, \quad (1.15)$$

where we adopted the normalization suggested by (1.9). As this trivial example shows, the quantum and classical thermal states differ, with the difference being more pronounced at low temperatures.

1.3 Open Quantum Systems (OQS)

We describe an open quantum system by starting with a closed system (C)⁴ comprised of the system of interest (S) and an environment (E). The Hilbert space of the closed system is a tensor product of the Hilbert spaces of the system of interest and the environment: $\mathcal{H}_C = \mathcal{H}_S \otimes \mathcal{H}_E$. The density matrix of the closed system obeys unitary dynamics governed by the equation of motion (1.3) with the closed system Hamiltonian given by

$$\mathbf{H}_C = \mathbf{H}_S + \mathbf{H}_E + \mathbf{H}_I + \mathbf{H}_R, \quad (1.16)$$

where \mathbf{H}_R contains all of the “renormalization” (R) effects. We shall comment on “renormalization” later.

The expectation value of an observable only on the system S can be calculated using the *reduced density matrix*, which is obtained from the full density matrix via a partial trace:

$$\rho_S = \text{Tr}_E [\rho_C] \quad (1.17)$$

$$\langle \mathcal{O}_S \rangle = \text{Tr}_S [\mathcal{O}_S \rho_S] \quad (1.18)$$

In the Wigner representation the partial trace corresponds to integration over phase space variables. If we denote the system variables by (X, P) and the environ-

⁴In the literature the adjective “isolated” is sometimes used to indicate no exchange of particles and energy and “closed” only refers to no exchange of particles but allowing for an exchange of energy. In this thesis, we use the adjective “closed” to indicate both no exchange of particles and energy.

ment variables by (x, p) we have

$$\mathcal{W}_S(X, P) = \int \frac{dx dp}{(2\pi\hbar)^{N_E}} \mathcal{W}_C(X, x, P, p) \quad (1.19)$$

where N_E is the number of degrees of freedom in the environment.

The time evolution of the open quantum system is in general governed by non-unitary dynamics. The equations governing the time evolution of the reduced density matrix and Wigner function are called *master equation* and *Fokker-Plank equation*, respectively. The theory of OQS is a highly developed field [30, 31]. In this chapter we will only go into the details of the exactly solvable QBM model. In Chapter 2 we will use perturbative results for general system and environments developed in Refs. [51, 52].

1.4 Quantum Brownian Motion (QBM) Model

In this section we introduce and solve the QBM that will be used to study various themes of this dissertation in following chapters. Our treatment follows closely that of Ref. [51]. The model consists of $N_S + N_E$ coupled harmonic oscillators. The system of interest consists of N_S oscillators, and N_E denotes the number of degrees of freedom that constitutes the environment.

1.4.1 The Hamiltonian

The Hamiltonian governing the unitary dynamics of the closed system dynamics is given by

$$\mathbf{H} = \mathbf{H}_S(\mathbf{X}, \mathbf{P}) + \mathbf{H}_E(\mathbf{x}, \mathbf{p}) + \mathbf{H}_I(\mathbf{X}, \mathbf{x}) + \mathbf{H}_R(\mathbf{X}), \quad (1.20)$$

$$= \frac{1}{2} (\mathbf{P}^T \mathbf{M}^{-1} \mathbf{P} + \mathbf{X}^T \mathbf{C} \mathbf{X}) + \frac{1}{2} (\mathbf{p}^T \mathbf{m}^{-1} \mathbf{p} + \mathbf{x}^T \mathbf{c} \mathbf{x}) - \mathbf{x}^T \mathbf{g} \mathbf{X} + \mathbf{H}_R(\mathbf{X}). \quad (1.21)$$

Since there is not much danger of confusing quantum mechanical operators and regular parameters in this section, bold symbols are used to indicate matrices and vectors of both types. Operators are not distinguished by a hat either, since that notation is reserved for the Laplace transform. In the rest of this dissertation, we will use bold symbols for operators only in places where confusion is likely or in places we want to emphasize the operator nature. Thus \mathbf{X} and \mathbf{P} are vectors of size N_S whose elements are the coordinates and momenta of the system oscillators. Similarly the environmental coordinates and momenta are denoted by x and p , respectively. These should be interpreted as operators in the quantum case and as ordinary numbers in the classical case. We assume that the spring constant matrices \mathbf{C}, \mathbf{c} as well as the mass matrices \mathbf{M}, \mathbf{m} are real and positive definite, and can be considered in general to be symmetric. If necessary, one can relax the positivity condition and even consider time-dependent mass matrices, spring constant matrices and system environment coupling matrix \mathbf{g} [51].

In general, the coupling to the environment modifies, or “renormalizes”, the dynamic frequency of the system oscillators. The frequency “renormalization” is a

well-understood effect, and it is often balanced by adding a counter-term $H_{\text{ren}}(\mathbf{X})$ to the Hamiltonian. This is especially useful if one wants to focus on other environmental effects like dissipation and decoherence.⁵ This motivates the choice of “renormalization” which is equivalent to inserting the entire system-environment interaction in the square of the potential:

$$\mathbf{H}_C = \frac{1}{2} (\mathbf{P}^T \mathbf{M}^{-1} \mathbf{P} + \mathbf{X}^T \mathbf{C} \mathbf{X}) + \frac{1}{2} \left(\mathbf{p}^T \mathbf{m}^{-1} \mathbf{p} + [\mathbf{x} - \mathbf{c}^{-1} \mathbf{g} \mathbf{X}]^T \mathbf{c} [\mathbf{x} - \mathbf{c}^{-1} \mathbf{g} \mathbf{X}] \right), \quad (1.22)$$

since this keeps the phenomenological system-system couplings from changing as will be seen in (1.33).

1.4.2 Solution of the QBM Model

We have seen in Sec. 1.2.3 that for quantum systems which have at most quadratic terms in their Hamiltonians, the dynamics in phase space representation is the same as the corresponding classical dynamics. This allows us to utilize the classical mechanical solution in terms of trajectories in phase space for both the classical and quantum models. This will be done in the next section. The difference between quantum and classical case will be manifest in the implementation of initial conditions, which will be briefly discussed in the section following the next. Further details will be provided in the individual chapters.

⁵An added technical advantage to this choice is that the spectrum of the resulting Hamiltonian is always bounded from below as can be seen in (1.22). Compare this with the treatment of Chapter 4, where we chose not to add the counter-term. As a result we had to make an extra assumption to rule out Hamiltonians without lower bound (see the paragraph following Eq. (4.10)).

1.4.2.1 Dynamics

The equations of motion can be obtained from the Hamiltonian (1.22) in the usual way by taking partial derivatives with respect to coordinates and momenta. This can be done while maintaining the compact matrix notation, although special care needs to be taken in maintaining the order of terms and in the interpretation of divisions. The resulting equations of motion are:

$$\mathbf{M}\ddot{\mathbf{X}}(t) + (\mathbf{C} + \delta\mathbf{C})\mathbf{X}(t) = \mathbf{g}^T\mathbf{x}(t), \quad (1.23)$$

$$\mathbf{m}\ddot{\mathbf{x}}(t) + \mathbf{c}\mathbf{x}(t) = \mathbf{g}\mathbf{X}(t), \quad (1.24)$$

$$\delta\mathbf{C} \equiv \mathbf{g}^T\mathbf{c}^{-1}\mathbf{g}, \quad (1.25)$$

We proceed by first solving the equation of motion (1.24) for the environmental degrees of freedom in terms of the system oscillators.

$$\mathbf{x}(t) = \left(\dot{\mathbf{f}}(t)\mathbf{m}\mathbf{x}_0 + \mathbf{f}(t)\mathbf{p}_0 \right) + (\mathbf{f} * \mathbf{g}\mathbf{X})(t), \quad (1.26)$$

$$\mathbf{f}(t) \equiv \mathbf{m}^{-\frac{1}{2}} \frac{\sin(\boldsymbol{\omega}t)}{\boldsymbol{\omega}} \mathbf{m}^{-\frac{1}{2}}, \quad (1.27)$$

$$\boldsymbol{\omega}^2 \equiv \mathbf{m}^{-\frac{1}{2}}\mathbf{c}\mathbf{m}^{-\frac{1}{2}}, \quad (1.28)$$

$$(A * B)(t) \equiv \int_0^t ds A(t-s)B(s), \quad (1.29)$$

where $*$ indicates the Laplace convolution, \mathbf{f} is the free Green's function of the bath positions and $\boldsymbol{\omega}$ is the free bath frequencies upon diagonalization. The solution (1.26) can then be substituted into (1.23) to yield the first form of the Langevin

Equation:

$$\mathbf{M}\ddot{\mathbf{X}}(t) + 2(\boldsymbol{\mu} \cdot \mathbf{X})(t) + (\mathbf{C} + \delta\mathbf{C}) \mathbf{X}(t) = \boldsymbol{\xi}(t), \quad (1.30)$$

$$\boldsymbol{\mu}(t) \equiv -\frac{1}{2}\mathbf{g}^T \mathbf{f}(t) \mathbf{g}, \quad (1.31)$$

$$\boldsymbol{\xi}(t) = \mathbf{g}^T \left(\dot{\mathbf{f}}(t) \mathbf{m} \mathbf{x}_0 + \mathbf{f}(t) \mathbf{p}_0 \right). \quad (1.32)$$

Here $\boldsymbol{\mu}(t, s)$ is called the dissipation kernel for reasons that will become clear later and $\boldsymbol{\xi}(t)$ is the noise acting on the system. Note that the dissipation kernel is independent of the environment's initial state, whereas the properties of noise are determined by it. In certain limits of physical interest, i.e. the Ohmic limit which we will encounter in Chapter 3, the dissipation kernel becomes ill-defined, and it is customary to integrate by parts the integral in which it appears. This amounts to trading the dissipation kernel $\boldsymbol{\mu}$ for what is called the damping kernel $\boldsymbol{\gamma}$ and in the process picking up a boundary term. The resulting Langevin equation is what we will use in the rest of this dissertation:

$$\mathbf{M}\ddot{\mathbf{X}}(t) + 2 \int_0^t ds \boldsymbol{\gamma}(t, s) \dot{\mathbf{X}}(s) + \mathbf{C} \mathbf{X}(t) + 2\boldsymbol{\gamma}(t) \mathbf{X}_0 = \boldsymbol{\xi}(t), \quad (1.33)$$

$$\boldsymbol{\gamma}(t, s) = \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\omega[t-s])}{2\omega^2} \mathbf{m}^{-\frac{1}{2}} \mathbf{g}, \quad (1.34)$$

The Langevin equation is a linear integro-differential equation. As such, it has two linearly independent solutions per degree of freedom.

Given that our damping kernel is stationary, the Langevin equation can be expressed in the Laplace domain as

$$[z^2 \mathbf{M} + 2z\hat{\boldsymbol{\gamma}}(z) + \mathbf{C}] \hat{\mathbf{X}}(z) = [z\mathbf{M}\mathbf{X}_0 + \mathbf{P}_0] + \hat{\boldsymbol{\xi}}(z), \quad (1.35)$$

where $\mathbf{P} = \mathbf{M}\dot{\mathbf{X}}$ and $(\mathbf{X}_0, \mathbf{P}_0)$ correspond to the initial values at $t = 0$, and the Laplace transform is defined as

$$\hat{f}(z) \equiv \int_0^\infty dt e^{-zt} f(t). \quad (1.36)$$

Formally, the solutions in the Laplace domain can be easily found by inversion:

$$\hat{\mathbf{X}}(z) = \hat{\mathbf{G}}(z)[z\mathbf{M}\mathbf{X}_0 + \mathbf{P}_0] + \hat{\mathbf{G}}(z)\hat{\boldsymbol{\xi}}(z), \quad (1.37)$$

$$\hat{\mathbf{G}}(z) = [z^2\mathbf{M} + 2z\hat{\boldsymbol{\gamma}}(z) + \mathbf{C}]^{-1}. \quad (1.38)$$

Note that since the damping kernel is symmetric, i.e. $\boldsymbol{\gamma}(t, s) = \boldsymbol{\gamma}(t, s)^T$, so are the propagator $\mathbf{G}(t, s)$ and its Laplace transform. It is also useful to consider the following representation:

$$\hat{\mathbf{G}}(z) = \mathbf{M}^{-\frac{1}{2}} \left[z^2 + 2z\hat{\boldsymbol{\lambda}}(z) + \boldsymbol{\Omega}^2 \right]^{-1} \mathbf{M}^{-\frac{1}{2}}, \quad (1.39)$$

$$\hat{\boldsymbol{\lambda}}(z) \equiv \mathbf{M}^{-\frac{1}{2}} \hat{\boldsymbol{\gamma}}(z) \mathbf{M}^{-\frac{1}{2}}, \quad (1.40)$$

$$\boldsymbol{\Omega}^2 \equiv \mathbf{M}^{-\frac{1}{2}} \mathbf{C} \mathbf{M}^{-\frac{1}{2}}, \quad (1.41)$$

where the eigenvalues of $\boldsymbol{\Omega}^2$ coincide with the squared frequencies of the normal modes of the free system. Back in the time domain we have

$$\mathbf{X}(t) = \dot{\mathbf{G}}(t)\mathbf{M}\mathbf{X}_0 + \mathbf{G}(t)\mathbf{P}_0 + (\mathbf{G} * \boldsymbol{\xi})(t), \quad (1.42)$$

This is the exact solution for the dynamics of the N_S system oscillators. Because the Langevin equation is a linear equation, its solutions consist of the sum of a homogeneous solution and a particular solution. The two sets of independent homogeneous

solutions satisfy the initial conditions:

$$\mathbf{M}\dot{\mathbf{G}}(0) = \mathbf{1}, \quad \mathbf{M}\ddot{\mathbf{G}} = 0, \quad (1.43)$$

$$\mathbf{G}(0) = 0, \quad \dot{\mathbf{G}}(0) = \mathbf{M}^{-1}. \quad (1.44)$$

$$(1.45)$$

1.4.2.2 On Initial Conditions

In the previous section we focused on the dynamics without worrying about initial conditions. The role of initial conditions of the system oscillators are easily seen in (1.42). As mentioned before, the damping kernel is independent of the initial conditions and as a result so are the homogenous solutions $\mathbf{G}(t)$ and $\mathbf{M}\dot{\mathbf{G}}(t)$. The initial conditions of the environmental oscillators effect the open system in two ways. First, dynamically the system is driven by the noise term ξ . Second, correlations may exist between system and environment initial conditions. We will consider three types of initial conditions for the BM model in this work: in Chapter 2 the thermal state of the closed system + environment , in Chapters 2 and 3 uncorrelated system and environment with the environment in a thermal state, and in Chapter 4 the classical microcanonical ensemble of the closed system + environment .

1.4.3 Significance and generality of the QBM model

Microscopic models, that make first principles calculations possible, allow one to examine all the assumptions entering the derivation rather than relying on phenomenology, and explore new horizons, like the low temperature or strong coupling

regimes. The QBM model is used extensively in the open quantum systems literature thanks to its exact solubility and its generality. The solubility is due to the linearity of the model. The generality may not be immediately obvious. Representing the environment by a set of simple harmonic oscillators might appear to be a serious restriction to weak influences on the system, because of its linearity. Yet it is known from the influence functional formalism [26], that such a model environment can emulate any source of Gaussian noise with proper choice of coupling. An argument for the generality of the model is given by Caldeira and Leggett [53]: “For most cases of interest, at least when the system variable is macroscopic, this assumption is physically reasonable; in that case the environment is usually also (geometrically) macroscopic and the interaction of the system with any one environmental degree of freedom is generally proportional to the inverse of the volume, while the characteristic energy of such a degree of freedom is volume-independent.” The applicability of the model is limited to cases where the influence of the system on each bath mode is weak. This does not imply that the influence of the bath as a whole on the system is weak as well. The Brownian particle interacts with a very large number of environmental degrees of freedom. The effect of these interactions can add up to yield strong dissipation, fluctuations and decoherence for the Brownian particle.

1.5 Fluctuation Theorems (FTs)

Recent advances in technology, like real-time monitoring and control of single molecules, enable experiments where small systems can be studied under nonequilib-

rium conditions [22]. Alongside these advances, there has been considerable progress in our theoretical understanding of the nonequilibrium statistical mechanics of small systems. The fluctuation theorems (FTs) of Jarzynski [11] and Crooks [14] are among these advances. For earlier work on entropy fluctuation theorems, such as by Cohen, Evans, Searles and others, see, e.g., [20].

FTs relate equilibrium thermodynamic quantities of a physical system, such as free energy differences, to the averages of mechanical quantities in nonequilibrium processes, like exponentiated work. For complex biological systems like proteins and DNAs, the free energy differences are difficult to calculate while the averages of work in nonequilibrium processes can be obtained from measurements in experiments or via careful numerical simulations.

1.5.1 Classical Fluctuation Theorems

In this dissertation we will be mostly concerned with the nonequilibrium work relation [11, 12],

$$\langle e^{-\beta W} \rangle = e^{-\beta \Delta F}, \quad (1.46)$$

and the closely related fluctuation theorem, due to Crooks [13–15],

$$\frac{P(W)}{\tilde{P}(-W)} = e^{\beta(W - \Delta F)}. \quad (1.47)$$

Both of these relate the statistical fluctuations in the work W performed on a system during a nonequilibrium process, to a free energy difference ΔF between two equilibrium states of the system. The angular brackets in Eq. (1.46) denote an

average over an ensemble of realizations of the process, and β specifies the inverse temperature at which the system is prepared prior to each realization. In Eq. (1.47) the numerator and denominator denote the distributions of work values corresponding to a conjugate pair of “forward” and “reverse” processes. Eqs. (1.46) and (1.47) have been derived by various means, using a variety of equations of motion to model the microscopic dynamics – see Ref. [54] for a review with extensive references – and have been confirmed experimentally [55–60].

Here we will focus our attention on the formulation of these results within the framework of Hamiltonian dynamics. The Hamiltonian for the system of interest is assumed to depend on a control parameter λ , whose time dependence over an interval $0 \leq t \leq \tau$ is specified by a schedule, or *protocol*, λ_t . The free energy difference ΔF refers to two different equilibrium states, corresponding to the initial and final parameter values, λ_0 and λ_τ . Work done during this process is defined as

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$$W = \int_0^\tau dt \frac{\partial H}{\partial \lambda} \dot{\lambda}(t), \quad (1.48)$$

where an overdot denotes derivative with respect to time. Although the Hamiltonian dynamics of the system is entirely deterministic, due to the probabilistic nature of the initial conditions that are sampled from the thermal phase space density, work is described by a probability distribution $P(W)$.

⁶For a discussion of various definitions of work, their relationship to each other and how that affects the content and context of the fluctuation theorems see [61].

1.5.2 Quantum Fluctuation Theorems

The main difficulty in formulating fluctuation theorems for quantum mechanics is defining work. Except for closed systems there is no agreement on a definition of work in quantum mechanics. For closed systems there is general agreement on the following operational definition [62,63]: 1) Measure the energy of the system using the Hamiltonian initially at $t = 0$ to be E_n^0 , thus “collapsing the wavefunction” to one of the eigenfunctions of the Hamiltonian at the initial time: $\mathbf{H}(0)|\psi_n^0\rangle = E_n^0|\psi_n^0\rangle$. 2) Let the system evolve under the time dependent Hamiltonian according to the prescribed protocol. 3) At the end of the protocol measure the energy of the system using the Hamiltonian at $t = \tau$ to be E_m^τ , thus collapsing the wavefunction to an eigenfunction of the Hamiltonian at τ : $\mathbf{H}(\tau)|\psi_m^\tau\rangle = E_m^\tau|\psi_m^\tau\rangle$. For this specified realization, work is defined as $W = E_m^\tau - E_n^0$. Since the system is closed, one can interpret the change in energy of the system as work performed on the system. In classical mechanics of isolated systems work acquires a probabilistic feature only due to the sampling of the initial conditions, since the dynamics is deterministic. In quantum mechanics work acquires an additional probabilistic feature from the dynamics:

$$\begin{aligned}
P(W) &= \frac{1}{2\pi} \int du e^{-iuW} \text{Tr}[e^{iu\mathbf{H}_H(\tau)} e^{-iu\mathbf{H}_H(0)} \boldsymbol{\rho}_\beta] \\
&= \sum_{m,n} \delta [W - (E_m^\tau - E_n^0)] |\langle \psi_m^\tau | \mathbf{U}(\tau, 0) | \psi_n^0 \rangle|^2 \langle \psi_n^0 | \boldsymbol{\rho}_\beta | \psi_n^0 \rangle \\
&= \sum_{m,n} \delta [W - (E_m^\tau - E_n^0)] p(n \rightarrow m) p_0(n), \tag{1.49}
\end{aligned}$$

where the subscript H indicates Heisenberg operators, $p_0(n)$ is the probability that the result of the first energy measurement is $|\psi_n^0\rangle$, and $p(n \rightarrow m)$ is the probability of transition from $|\psi_n^0\rangle$ to $|\psi_m^\tau\rangle$ during time evolution from 0 to τ . Jarzynski equality and Crooks's fluctuation theorem can be proven in a few lines for a closed system with this definition of work. In Chapter 3 we present our approach to quantum fluctuation theorems, which differs from the above formulation in that it does not involve energy measurements.

1.6 Summary of Major Findings

In chapter 2 we investigate the late-time steady states of open quantum systems coupled to a thermal reservoir in the strong coupling regime. In general such systems do not necessarily relax to a Boltzmann distribution if the coupling to the thermal reservoir is non-vanishing or equivalently if the relaxation time scales are finite. Using a variety of nonequilibrium formalisms valid for non-Markovian processes, we show that starting from a product state of the closed system = system + environment, with the environment in its thermal state, the open system which results from coarse graining the environment will evolve towards an equilibrium state at late times. This state can be expressed as the reduced state of the closed system thermal state at the temperature of the environment. For the quantum Brownian motion model, which is exactly solvable, we are able to show in a rigorous way that all multi-time correlations of the open system evolve towards those of the closed system thermal state. Multi-time correlations are especially relevant

in the non-Markovian regime, since they cannot be generated by the dynamics of the single-time correlations. For more general systems, which cannot be exactly solved, we are able to provide a general proof that all single-time correlations of the open system evolve to those of the closed system thermal state, to first-order in the relaxation rates. For the special case of a zero-temperature reservoir, we are able to explicitly construct the reduced closed system thermal state in terms of the environmental correlations.

In Chapter 3 we present a first-principles analysis of the nonequilibrium work distribution and the free energy difference of a quantum system interacting with a general environment (with arbitrary spectral density and for all temperatures) based on the quantum Brownian motion model. We use the decoherent history conceptual framework to explain how the notion of trajectories in a quantum system can be made viable and use the environment-induced decoherence scheme to assess the strength of noise which could provide sufficient decoherence to warrant the use of trajectories to define work in open quantum systems. From the solutions to the Langevin equation governing the stochastic dynamics of such systems we were able to produce formal expressions for these quantities entering in the FTs, and using these expressions prove explicitly the validity of the FTs at the high temperature limit. At low temperatures our general results would enable one to identify the range of parameters where FTs may not hold or need be expressed differently. We explain the relation between classical and quantum FTs and the advantage of this micro-physics open-system approach over the phenomenological modeling and energy-level calculations for substitute closed quantum systems.

In Chapter 4 we calculate the probability distribution of work for the exactly solvable classical Brownian motion model with a finite environment. The initial conditions of the combined system and environment are sampled from a microcanonical distribution and the system is driven out of equilibrium by changing the control parameter according to a prescribed protocol. In the limit of infinitely large environment, i.e. $N \rightarrow \infty$, we recover the nonequilibrium work theorem and Crooks's fluctuation theorem. Moreover, the microcanonical Crooks relation is verified for finite environments. The equivalence of multi-time correlation functions of the system in the infinite environment limit for canonical and microcanonical ensembles is proven. Our results support the hypothesis that for macroscopically large environments the sampling of the initial conditions from a canonical or microcanonical distribution is equivalent as far as system observables are concerned. Albeit restricted to one simple model, our results go beyond the standard results on *ensemble equivalence* in a number of ways. First of all, the system plus environment model considered in this chapter is not extensive and homogenous (like gas particles in a container). Second, the thermodynamic limit (of the environment) is taken with the system size fixed. Thus, the quantities we consider do not have to be macroscopic. Third, we consider multi-time averages taken over nonequilibrium processes. This implies that even fluctuations behave identically for canonical and microcanonical ensembles. Finally, we show that in the infinite environment limit, the Brownian particle relaxes to an equilibrium state which does not depend on whether the canonical or microcanonical ensemble is used for the environment.

In Chapter 5 we summarize our thoughts in attempting a systematic investi-

gation into some key foundational issues of quantum macroscopic phenomena, with the goal of ultimately revealing or building a viable theoretical framework. Three major themes discussed are the large N expansion [64], the correlation hierarchy [65] and quantum entanglement [66, 67]. We give a sketch of the first two themes. We then discuss several key issues in the consideration of macro and quantum. We point out that there exist many levels of structure in a composite body and only by judicious choice of an appropriate set of collective variables can one give the best description of the dynamics of a specific level of structure. To understand how the coupling pattern amongst the constituents of two macro objects (modelled by N harmonic oscillators), enters into the picture, we consider two types of coupling: each constituent particle is coupled to only one other particle (1-to-1) versus coupled to all particles (1-to-all). In the 1-1 case with pairwise interactions of equal strength, the entanglement is independent of the number of constituent particles N in the macroscopic object. In the 1-to-all case the relative coordinates are decoupled and the center of mass (CoM) coupling scales with N . Here we expect the entanglement between the CoM variables to increase with increasing size of the macroscopic objects and survive at higher temperatures. We provide a proof of the conditions whereby the CoM variable decouples, a prerequisite for the CoM variable to be protected from decoherence and play a special role in the entanglement between two such macroscopic objects. Similar qualitative behavior is found when the couplings between the constituents of the macroscopic objects are statistically independent Gaussian random variables.

Quantum entanglement is known to persist at high temperatures [68] and

large scales [69] under certain conditions, and may actually decrease with increased connectivity in a quantum network [70]. We pick out these somewhat counter-intuitive examples to show that there are blind spots worthy of our attention and issues which we need to analyze closer. Our purpose is to try to remove the stigma that quantum only pertains to micro, in order to make way for deeper probes into the conditions whereby quantum features of macroscopic systems are manifest.

Some technical details related to Chapter 2 have been provided in the Appendix A and those related to Chapter 4 are provided in Appendix B.

Chapter 2: Equilibrium states of open quantum systems in the strong coupling regime

2.1 Introduction

Equilibrium states are typically discussed and derived in one of three settings or scenarios. In the more-common equilibrium (Gibbs) perspective, originally based upon classical ensemble theory, the entire system consisting of a system of interest plus its environment is taken to have some well-defined state or set of states, and upon coarse graining the environment, the system can appear thermal [33,34]. In the less-common non-equilibrium perspective, the environment is taken to be initially thermal, whereas the open system is allowed to dynamically relax from an arbitrary initial state into an equilibrium state [29,71–73]. This approach is referred to as the Langevin paradigm [74]. Both scenarios described above apply to situations where there is a clear distinction and separation between the system and environment degrees of freedom. When there is no clear distinction or the separation is not physically justifiable, as in a molecular gas where each particle is identical, a very different set of physical variables and different kind of coarse graining measure need be considered. One can examine the behavior of the n -particle distribution functions

and perform the coarse graining (e.g., “slaving” in [74]) on the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy [8]. This approach is referred to as the Boltzmann paradigm.

The equilibrium and non-equilibrium perspectives can be made to complement each other rather naturally within the Langevin or open system paradigm. In the former case, Popescu et al. [33] have shown that for an overwhelming majority of pure states of the system + environment (within a narrow energy interval), the reduced density matrix is very close to the reduced density matrix corresponding to the microcanonical state of the system + environment (defined in the same energy interval). In their approach the comparison is done without explicitly determining an equilibrium state. The authors emphasize that for strong coupling, the equilibrium state is not of Boltzmann type, and yet their results are valid in this domain. It is important to note that dynamics does not play any role in their derivation; the entire argument is based on *kinematics*. The beauty of this approach is that one can explain the abundance of thermal-like states without referring to ensembles or time averages.

Linden *et al* [36] expands upon the approach of [33, 34] to demonstrate *dynamical* relaxation¹ under very weak assumptions. Specifically, they proved that any subsystem of a much larger quantum system will evolve to an approximately steady state. On the other hand Reimann [37] showed that the expectation value of

¹See Sec. 2.1.1 for the definition of the terms relaxation, equilibration and thermalization as used in this work. There we also describe the meaning of the term *equilibration* as used in Refs. [36, 37, 75, 76], which differ from our definition.

any “realistic” quantum observable will relax to an approximately constant value. ([75] gave a clear analysis and unification of these two results.) Finally [76] proves relaxation over a finite amount of time both in the sense of [36] and [37].

Dynamical relaxation of an open quantum system has been studied in the limit of vanishing coupling to the environment in [29, 71–73]. In this limit the equilibrium state is shown to be of Boltzmann form, i.e. described by a canonical ensemble. In this case the result is called thermalization, rather than just relaxation. In our work reported here, we derive the equilibrium state of an open system coupled to a thermal reservoir explicitly, even in the strong coupling regime. Moreover for the N oscillator quantum Brownian motion (N-QBM) model we are able to show the relaxation of multi-time correlations of the open system as well. To do so we need to restrict the environment to be in a thermal initial state.

Relaxation in quantum lattice systems have been studied in great detail in [77, 78]. These papers demonstrate local relaxation of the system to Gaussian states under dynamics generated by quadratic Hamiltonians with short-range interactions. Relaxation occurs irrespective of the initial state. However, the parameters of the asymptotic local Gaussian states do depend on it. By “local” it is meant that any “small” subset of lattice sites relaxes to a steady state whereby every part of the lattice acts as an environment for the other parts. Their proofs feature a non-commutative central limit theorem for non-i.i.d. random variables.

Recently, Pagel *et al.* [79] studied relaxation in the QBM model with a single system oscillator. Similar to [78] and unlike our work, [79] shows relaxation even for non-thermal initial states for the environment. They derived conditions for ther-

malization and a relation of the asymptotic temperature to the energy distribution in the initial bath state. The methodology of this work is the same as our treatment of the QBM model. Their derivation involves discarding rapidly oscillating terms at late times. This step has a similar effect to the use of the non-commutative central limit theorem in [78]; however, in our opinion it is mathematically not as rigorous. Neither of these works consider multi-time correlations.

Another difference between our work and [36, 37, 75–78] is in the methods and emphasis. We take the open quantum systems approach [26–31] of assuming an environment (E) which the system (S) interacts with, keeping some coarse grained information about the environment and accounting for its systematic influences on the system in a self-consistent manner. The time evolution of the open quantum system is in general governed by non-unitary dynamics. In contradistinction, [36, 37, 75–78] consider the unitary time evolution of the closed system (S + E) and then trace out the environment to get the system state. Both approaches are equally valid, each providing a different perspective into the physics with different emphasis. We will provide a more detailed comparison of our results to those in the literature in the discussion section at the end.

2.1.1 Relaxation, Equilibration and Thermalization

Before we present our approach, we want to define carefully what is meant by equilibration. To begin with let us consider a system in contact with two thermal

reservoirs² at different temperatures. The system relaxes into a late-time steady state, which can be described by a reduced density matrix. All expectation values of system operators will also be time-independent at late times. Yet there will be a steady heat flux from the hot reservoir to the cold reservoir through the system. This is an example of a non-equilibrium steady state.

In general we define steady states via time independent density matrices: $d\rho(t)/dt = 0$ and use the term *relaxation* to describe the generic convergence of the reduced density matrix to a fixed but arbitrary state in the late-time limit. If the density matrix is diagonal in the energy eigenbasis of the system, we call it a stationary state. An isolated stationary state is also a steady state, but this is not true for open systems with non-vanishing coupling to their environments.

In this work we reserve the term equilibrium for systems whose multi-time correlations can be derived from the thermal state of a possibly extended closed system which is governed by Hamiltonian dynamics. As a result of our definition, equilibration implies relaxation but the reverse is not true. The thermal reservoir distinguishes itself from other possible environments by the *universality* of its fluctuation-dissipation relation (FDR)³, detailed-balance conditions and

²We call an environment a reservoir if the environment has an infinite number of degrees of freedom, and a reservoir at constant temperature, a thermal reservoir.

³As long as the environment is modelled after a physical system, fluctuations will be related to dissipation; hence there will be a FDR. However for general environments this relation depends on the specifics of the system-environment coupling. Thermal environments are unique in that the FDR does not depend on the details of the system and the coupling to the system [80]. This is why our proof does not extend to non-equilibrium steady states arising from non-thermal environments

Kubo-Martin-Schwinger (KMS) relations. In the vanishing coupling limit thermal reservoirs lead to the thermalization of the system as defined below. However for non-vanishing coupling to a thermal reservoir the equilibrium state of the system does not need to be of the Boltzmann form

$$\rho_S(\beta) = \frac{e^{-\beta \mathbf{H}_S}}{\text{Tr}_S [e^{-\beta \mathbf{H}_S}]} \quad (2.1)$$

The asymptotic states we derive in this chapter in the strong coupling limit describe equilibration and not thermalization.

The term thermalization is reserved for the relaxation of the density matrix of a system to the Boltzmann form (2.1) *irrespective* of the initial state of the system. Thermalization defined in this sense can take place only if the system-environment coupling is vanishingly weak. To be specific, one requires (1) decaying environmental correlation functions, defined in Sec. 2.3, (2) an initially thermal reservoir and (3) vanishing relaxation rates⁴ or, equivalently, vanishing environmental correlation functions.

These conditions are customarily achieved by assuming short-range interactions and a relatively large system size, see Fig. (2.1). However this assumption is generally not justifiable for small systems as Fig. (2.2) suggests. In this chapter, we address the stationary state of open quantum systems in contact with a thermal

such as two thermal reservoirs at different temperatures. A treatment of non-thermal environments in QBM model can be found in [79].

⁴To see a simple example of a relaxation rate consider the N-QBM model of Sec. 1.4 for N=1. In the Markovian limit the damping kernel can be written as $\gamma(t, s) = \gamma_0 M \delta(t - s)$, where γ_0 acts as the damping rate.

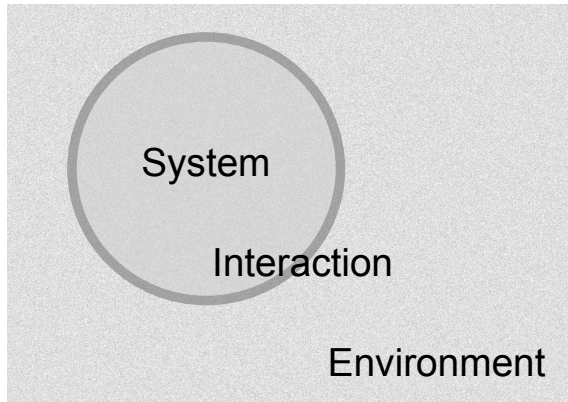


Figure 2.1: Depiction of a system embedded in its environment, with short-range interactions. The typical argument for neglecting the interaction energy is that in the macroscopic limit the boundary becomes immeasurable in relation to the bulk.

reservoir at temperature $k_B T = 1/\beta$, without the assumption of a vanishing interaction strength and allow for finite relaxation timescales. Relation (2.1) is known not to hold under these conditions [81]. Phenomenologically, one can estimate the corrections we describe by the ratio of the relaxation rates γ to the system’s energy level splittings Ω , or γ/Ω .⁵

As thoroughly discussed in Ref. [81], this fact is often overlooked in many circumstances, due to the effects of ancillary approximations such as the rotating-wave approximation, renormalization of environmentally-induced energy-level shifts

⁵A well-known example is the density of states for an atom or molecule, which is necessarily interacting with the electromagnetic field to a degree which cannot be ignored when considering the Lamb shift, black-body radiation shifts, etc.. For optical frequencies, the emission rates of atoms are very small relative to their transition frequencies, and so these corrections are very small. However in other systems, such as condensates, these corrections can be of considerable size.

and overly-simplistic models. As we explain in Appendix [A.1](#), this fact may also be overlooked due to its absence in the case of classical, Gaussian noise.

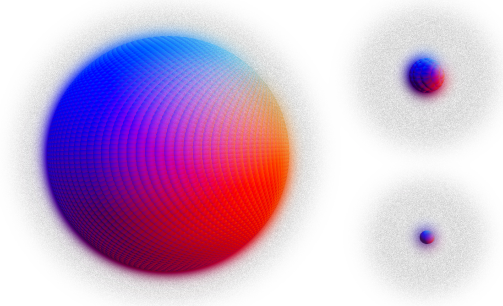


Figure 2.2: Depiction of systems of decreasing particle number. For systems consisting of a small number particles, the argument in Fig. [2.1](#) obviously does not apply. Furthermore, it is known that neglecting the interaction energy in these finite systems always results in infinite relaxation and thermalization times.

Finally, the term equilibrium is used in Ref. [\[36\]](#) to describe what in our terminology are steady states and in Ref. [\[37\]](#) to describe what in our terminology are stationary states. Both cases have been covered in Refs. [\[75, 76\]](#) with the single term equilibrium. These states do not necessarily meet our more stringent criteria of equilibrium described above. Here we refer to the result of these works using the terminology we defined above.

2.1.2 Model and Assumptions

In this chapter we use the open quantum system setup described in Sec. [1.3](#) which describes a system coupled to an environment. The interaction Hamiltonian \mathbf{H}_I in [\(1.16\)](#) generates environmental correlation functions, c.f. Eqs. [\(2.41\)](#), [\(2.45\)](#)),

and we assume these correlations to be decaying functions. This assumption allows for irreversible dynamics in the open system. Implicit in this assumption is that the environment contains a continuum of modes (e.g. infinite volume). This latter assumption can be satisfied by coupling the system directly to field degrees of freedom that are uncountably infinite, such as the electromagnetic field. Note, however, that we do not assume the interaction Hamiltonian to be negligible compared to the system Hamiltonian.

Finally, for mathematical simplicity we assume the initial state of the system and environment to be uncorrelated at $t = 0$ ⁶

$$\rho_{\text{C}}(0) = \rho_{\text{S}}(0) \otimes \frac{e^{-\beta \mathbf{H}_{\text{E}}}}{Z_{\text{E}}(\beta)}, \quad (2.2)$$

where the environment (a thermal reservoir) is in its isolated equilibrium state with partition function $Z_{\text{E}}(\beta) = \text{Tr}_{\text{E}}[e^{-\beta \mathbf{H}_{\text{E}}}]$, and the system (S) is in an arbitrary state. The proofs in this chapter depend crucially on the properties of thermal states as discussed before.

The assumption of a thermal state for the environment can be justified, for instance, by the approach of Popescu et al. [33] in the weak coupling limit, by giving the environment its own environment, without any restriction on the system-environment coupling strength. In this sense the work of Popescu et al., and those prior, serve as a pedagogical springboard for our analysis of strongly-coupled systems.

⁶The implication of initial correlations are considered in Ref. [82, 83]: Correlated initial states are more physical, particularly in the early time evolution, but they have essentially no bearing on the mathematical results we derive herein, which are focused upon the asymptotic time evolution.

2.1.3 Results

It is well known that in the limit of vanishing interaction strength, an open system coupled to a single thermal reservoir relaxes to its thermal state [29,52,71,81]

$$\lim_{\gamma \rightarrow 0} \lim_{t \rightarrow \infty} \boldsymbol{\rho}_S(t) = \frac{e^{-\beta \mathbf{H}_S}}{Z_S(\beta)}, \quad (2.3)$$

where $\boldsymbol{\rho}_S(t) = \text{Tr}_E[\boldsymbol{\rho}_C(t)]$ denotes the reduced density matrix and γ a generic relaxation rate of the open system. Note that all relaxation rates are, at minimum, second-order in the interaction, being primarily determined by the two-time correlations of the environment.

In Ref. [84], it was shown to second-order in the interaction, and for a single tensor-product coupling of system and environment operators, that an open system coupled to a single thermal reservoir can be confirmed to relax to the reduced closed system thermal state

$$\lim_{t \rightarrow \infty} \boldsymbol{\rho}_S(t) = \text{Tr}_E \left[\frac{e^{-\beta \mathbf{H}_C}}{Z_C(\beta)} \right]. \quad (2.4)$$

We extend this proof to general system-environment couplings. For zero-temperature environments we demonstrate agreement with the ground state obtained from the time-independent Schrödinger equation. Moreover, we give a non-perturbative proof of Eq. (2.4) for the exactly-solvable model of N -oscillator quantum Brownian motion (N-QBM), wherein the interacting system and environment are linear. In that model we are also able to rigorously prove that all multi-time correlations of the open system relax to those of the closed system thermal state with non-vanishing interaction. Correspondence of the multi-time correlations is an important consideration

as, outside of the Markovian regime, the dynamics of the multi-time correlations cannot be generated by the dynamics of the single-time correlations, as per the quantum regression theorem [85].

2.1.3.1 The *reduced*, closed system thermal state

It is important to emphasize that Eq. (2.4) pertains strictly to the open system S and not to the closed system $(S + E)$, as equilibration requires not only a reservoir and late-time limit, but also a degree of coarse graining. As we show in Sec. 2.2.5, if one considers any individual mode of the environment, then its dependence upon the initial state of the system never decays. In this sense, information pertaining to the system's past is encoded in the environment, but only when considering the state of the closed system $(S + E)$. However, upon coarse graining the environment by considering the time-evolution of a continuum of environment energies, and not one individual mode energy, then all dependence upon the initial state of the system is seen to decay away in time. In this sense, information pertaining to the system's past is only measurable for a finite span of time.

The above statement is based on the fact that, while the open system experiences irreversible dynamics: dissipation, diffusion and decoherence, the closed system $(S + E)$ experiences reversible dynamics. Consider, for instance, the coupling of a mixed state of the system to a zero-temperature reservoir. Given unitary dynamics, the joint state of the system and environment cannot relax from a mixed state into a pure state (the ground state of the interacting theory). However, the

environment is exceedingly large when compared to the system, and so the system’s entropy, when spread out over every mode of the environment, can become immeasurable. This is a general phenomena of environmentally-induced irreversible dynamics: conserved quantities such as energy and entropy can flow into the environment, and owing to the overwhelmingly large number of degrees of freedom, become difficult to track or retrieve.

The chapter is organized as follows: In Sec. 2.2 we derive the equilibrium state for the linear N-QBM model. In Sec. 2.3 we extend our analysis to nonlinear systems via perturbation theory. In Sec. 2.4 we summarize our results and compare them to relevant works in the literature and provide some new insights into the key issues. Some technical details have been provided and the notation is defined in the Appendices.

2.2 Linear systems

Calculation of the late-time steady state and multi-time correlations of an open quantum system requires the knowledge of and the ability to treat the dynamics with due consideration to initial state. As mentioned in the Sec. 1.2.3, the dynamics of quantum and classical linear systems are identical. As a result they can be solved exactly by finding the transformation which maps the system into a set of uncoupled harmonic oscillators (eigenmodes) undergoing undamped oscillations of a single eigenfrequency. This method, referred to as the “diagonalization of the Hamiltonian” or a generalized Bogoliubov transformation, gives the time evolution

of all oscillators as superpositions of the eigenmodes. In the limit of an infinite environment (and only then), the superposition for the system oscillators can result in dissipative and stochastic behavior at late times.

However as mentioned in Sec. 1.4.2.2 the initial conditions are different for thermal states in quantum mechanics versus classical mechanics, the difference being especially pronounced at low temperatures (for an example see Eq.(1.14)). This is the main source of non-triviality of our result Eq. (2.4) as it applies to linear systems. As has been detailed in Appendix A.1, it is relatively simple to account for the effects of a linear environment in classical mechanics. This is not so in quantum mechanics because the Wigner function of the thermal state is quite complicated (especially for systems with multiple degrees of freedom), and coarse graining the environmental degrees of freedom remains challenging.

In this section we adopt the open quantum system approach in following the dissipative dynamics of our system in the form of a Langevin equation wherein the noise terms fully incorporate the influence of the environmental degrees of freedom. This method produces the same result as the explicit diagonalization of the Hamiltonian of the closed system, yet it shifts the focus on the reduced system early in the derivation and is mathematically more straightforward. A trivial observation allows us to simplify the derivation even further. Since the thermal state of a closed Hamiltonian system is stationary, we can replace the closed system thermal state by its own late-time steady state (see Fig. 2.3). This way we have a symmetry between the quantities we want to compare and fewer terms to calculate overall.

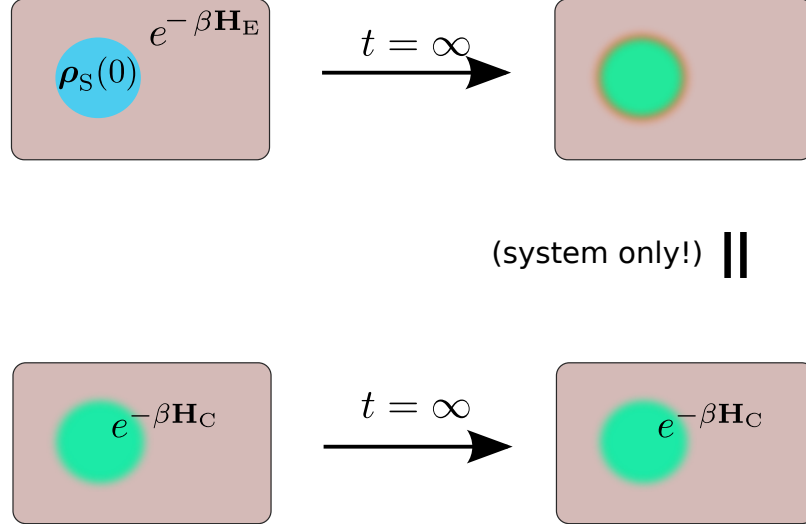


Figure 2.3: The thermal state of the combined system + environment is stationary, i.e. it does not change over time. Also the multi-time correlations are unchanged under time translations. Our goal is to compare this state with the late-time limit of the product initial state Eq. (2.2), as far as the system observables are concerned. Instead of doing this directly, we also take the late-time limit of the former, which does not change that state as argued above. The comparison of two late-time expressions prove to be much easier technically in the open system approach.

2.2.1 Noise

We consider the case in which the system and environment are uncorrelated at $t = 0$ and the environment is in its thermal state $e^{-\beta H_E}/Z_E(\beta)$. The noise has zero mean and the two time correlation is given by the noise kernel

$$\nu(t, t') = \langle \xi(t) \xi(t')^T \rangle_{\xi} = \mathbf{g} \mathbf{m}^{-\frac{1}{2}} \frac{\coth(\beta \hbar \omega / 2) \cos(\omega [t - t'])}{2\omega} \mathbf{m}^{-\frac{1}{2}} \mathbf{g}, \quad (2.5)$$

where the Gaussian average over the stochastic process ξ is equivalent to tracing over the environment degrees of freedom. The noise and damping kernels then satisfy

the fluctuation-dissipation relation (here in the Fourier domain)

$$\tilde{\boldsymbol{\nu}}(\omega) = \tilde{\kappa}(\omega)\tilde{\boldsymbol{\gamma}}(\omega), \quad (2.6)$$

$$\tilde{\kappa}(\omega) \equiv \hbar\omega \coth\left(\frac{\hbar\omega}{2k_{\text{B}}T}\right), \quad (2.7)$$

with the Fourier transform defined

$$\tilde{f}(\omega) \equiv \int_{-\infty}^{+\infty} dt e^{-i\omega t} f(t), \quad (2.8)$$

and where $\tilde{\kappa}$ is the (quantum) FDR kernel. Therefore, the problem is completely specified in terms of the damping kernel. For general Gaussian states of the closed system, for which the system and environment are correlated, the noise can be correlated with $(\mathbf{X}_0, \mathbf{P}_0)$ and the noise kernel modified. This is the case for the closed system thermal state given by the density matrix $e^{-\beta\mathbf{H}_c}/Z_C(\beta)$, as we will see in the following sections.

2.2.2 Single-time correlations in the closed system thermal state

In this section we calculate the single-time correlations in the closed system thermal state of the N-QBM model. The partition function for the N-QBM model has been derived in Appendix A.3, Eq. (A.41). In the rest of the chapter including associated appendices we suppress the dependence of the partition function on β for brevity of notation. As a first step we take the logarithm of the partition function and write it as:

$$\log Z_C = \log Z_E - \frac{1}{2}\text{Tr} \log \mathbf{M}^{-1} - \frac{1}{2}\text{Tr} \log \mathbf{C} - \sum_{r=1}^{\infty} \text{Tr} \log \left(\mathbf{M}^{-1} \hat{\mathbf{G}}(\nu_r)^{-1} \right) + \text{constant}, \quad (2.9)$$

where $\nu_r = 2\pi r/\hbar\beta$ are the Matsubara frequencies.

We begin by making a general observation. Consider the thermal state of a system described by a Hamiltonian where the momenta appear only in the kinetic energy term of the form $\sum_a p_a^2/2m$. Then all correlations between position and momentum operators vanish: $\langle x_a p_b \rangle = 0$. This can be seen by noting that all correlations are time-translation-invariant in equilibrium and forming the derivatives $\frac{d}{dt} \langle x_a(t) x_b(t) \rangle$ and $\frac{d}{d(t-t')} \langle x_a(t) x_b(t') \rangle \big|_{t=t'}$. This observation applies to N-QBM model.

Let angular bracket with the subscript C denote expectation values in the closed system thermal state. Expectation values corresponding to the uncorrelated initial state are denoted by attaching the subscript E to the bracket. For the purpose of partial differentiation, the partition function is to be regarded as a function of **C**, **M**, **c**, **m**, **g** and not (explicitly) of ω . With a straightforward application of Theorem 1, the reduced system correlations are given by:

$$\langle \mathbf{X}\mathbf{X}^T \rangle_C = -\frac{2}{\beta} \frac{\partial \log Z_C}{\partial \mathbf{C}}, \quad (2.10)$$

$$\langle \mathbf{X}\mathbf{P}^T \rangle_C = \langle \mathbf{P}\mathbf{X}^T \rangle = 0, \quad (2.11)$$

$$\langle \mathbf{P}\mathbf{P}^T \rangle_C = -\frac{2}{\beta} \frac{\partial \log Z_C}{\partial \mathbf{M}^{-1}}. \quad (2.12)$$

The position-position and position-momentum correlations between system and

reservoir modes are calculated similarly:

$$\begin{aligned}\langle \mathbf{X}\mathbf{x}^T \rangle_C &= \langle \mathbf{x}\mathbf{X}^T \rangle_C^T = \frac{1}{\beta} \frac{\partial \log Z_C}{\partial \mathbf{g}^T} + \langle \mathbf{X}\mathbf{X}^T \rangle_C \mathbf{g}^T \mathbf{c}^{-1}, \\ \langle \mathbf{X}\mathbf{p}^T \rangle_C &= \langle \mathbf{p}\mathbf{X}^T \rangle_C^T = 0,\end{aligned}\tag{2.13}$$

$$\langle \mathbf{P}\mathbf{x}^T \rangle_C = \langle \mathbf{x}\mathbf{P}^T \rangle_C^T = 0.\tag{2.14}$$

To calculate the momentum-momentum correlations between system and environment, we take the time derivative of $\langle \mathbf{X}(t)\mathbf{p}^T(t) \rangle_C$ and set it to zero. Since in the closed system thermal state all expectation values are time-independent, we know that there is in fact no dependence on time. Using the equations of motion it is straightforward to show that:

$$\langle \mathbf{P}\mathbf{p}^T \rangle_C = \mathbf{M} \langle \mathbf{X}\mathbf{x}^T \rangle_C \mathbf{c} - \mathbf{M} \langle \mathbf{X}\mathbf{X}^T \rangle_C \mathbf{g}^T.\tag{2.15}$$

The environment correlations can be calculated by direct differentiation of the partition function:

$$\langle \mathbf{x}\mathbf{x}^T \rangle_C = -\frac{2}{\beta} \frac{\partial \log Z_C}{\partial \mathbf{c}} + \mathbf{c}^{-1} \mathbf{g} \langle \mathbf{X}\mathbf{X}^T \rangle_C \mathbf{g}^T \mathbf{c}^{-1},\tag{2.16}$$

$$\langle \mathbf{x}\mathbf{p}^T \rangle_C = \langle \mathbf{p}\mathbf{x}^T \rangle_C = 0,\tag{2.17}$$

$$\langle \mathbf{p}\mathbf{p}^T \rangle_C = -\frac{2}{\beta} \frac{\partial \log Z_C}{\partial \mathbf{m}^{-1}}.\tag{2.18}$$

Now we are in a position to determine all the single-time correlations of the interacting theory in the closed system thermal state. Since the equilibrium state is stationary these single-time correlations are time-independent. The details for some of these formulae are provided in Appendix A.4. All the nonzero correlations are

given by:

$$\langle \mathbf{X}\mathbf{X}^T \rangle_C = \frac{1}{\beta} \hat{\mathbf{G}}(\nu_0) + \frac{2}{\beta} \sum_{r=1}^{\infty} \hat{\mathbf{G}}(\nu_r), \quad (2.19)$$

$$\langle \mathbf{P}\mathbf{P}^T \rangle_C = \frac{1}{\beta} \left(\mathbf{M} - \nu_0^2 \mathbf{M} \hat{\mathbf{G}}(\nu_0) \mathbf{M} \right) + \frac{2}{\beta} \sum_{r=1}^{\infty} \left(\mathbf{M} - \nu_r^2 \mathbf{M} \hat{\mathbf{G}}(\nu_r) \mathbf{M} \right), \quad (2.20)$$

$$\langle \mathbf{X}\mathbf{x}^T \rangle_C = \langle \mathbf{X}\mathbf{X}^T \rangle_C \mathbf{g}^T \mathbf{c}^{-1} - \frac{2}{\beta} \sum_{r=1}^{\infty} \nu_r \hat{\mathbf{G}}(\nu_r) \hat{\gamma}(\nu_r) \mathbf{g}^{-1}, \quad (2.21)$$

$$\langle \mathbf{P}\mathbf{p}^T \rangle_C = \mathbf{M} \langle \mathbf{X}\mathbf{x}^T \rangle_C \mathbf{c} - \mathbf{M} \langle \mathbf{X}\mathbf{X}^T \rangle_C \mathbf{g}^T, \quad (2.22)$$

$$\langle \mathbf{p}_0 \mathbf{p}_0^T \rangle_C = \langle \mathbf{p}_0 \mathbf{p}_0^T \rangle_E - \frac{2}{\beta} \sum_{r=1}^{\infty} \nu_r^2 \mathbf{m} \hat{\mathbf{f}}(\nu_r) \mathbf{g} \hat{\mathbf{G}}(\nu_r) \mathbf{g}^T \hat{\mathbf{f}}(\nu_r) \mathbf{m}, \quad (2.23)$$

$$\begin{aligned} \langle \mathbf{x}_0 \mathbf{x}_0^T \rangle_C &= \langle \mathbf{x}_0 \mathbf{x}_0^T \rangle_E + \mathbf{c}^{-1} \mathbf{g} \langle \mathbf{X}_0 \mathbf{X} \rangle_C \mathbf{g}^T \mathbf{c}^{-1} - \frac{2}{\beta} \sum_{r=1}^{\infty} \nu_r^2 \left(\mathbf{c}^{-1} \mathbf{m} \hat{\mathbf{f}}(\nu_r) \mathbf{g} \hat{\mathbf{G}}(\nu_r) \mathbf{g}^T \hat{\mathbf{f}}(\nu_r) \right) \\ &\quad - \frac{2}{\beta} \sum_{r=1}^{\infty} \nu_r^2 \left(\hat{\mathbf{f}}(\nu_r) \mathbf{g} \hat{\mathbf{G}}(\nu_r) \mathbf{g}^T \hat{\mathbf{f}}(\nu_r) \mathbf{m} \mathbf{c}^{-1} \right) \\ &\quad - \frac{2}{\beta} \sum_{r=1}^{\infty} \nu_r^4 \left(\mathbf{c}^{-1} \mathbf{m} \hat{\mathbf{f}}(\nu_r) \mathbf{g} \hat{\mathbf{G}}(\nu_r) \mathbf{g}^T \hat{\mathbf{f}}(\nu_r) \mathbf{m} \mathbf{c}^{-1} \right), \end{aligned} \quad (2.24)$$

where $\hat{\mathbf{f}}$ is the Laplace transform of the free reservoir propagator given by Eq. (1.27).

2.2.3 Equivalence of single-time correlations for the open system

In this subsection we show that the single-time correlations of system variables for the uncorrelated initial state are asymptotically identical to the single-time correlations corresponding to the closed system thermal state. We start by calculating the variances for the closed system thermal state. The requirement that $\mathbf{G}(t)$ is a decaying function means that the Laplace transform $\hat{\mathbf{G}}(z)$ is analytic in the right half-plane. Hence $\hat{\mathbf{G}}(-i\omega)$ is analytic in the upper-half plane. On the other hand $\coth(\beta\hbar\omega/2)$ has simple poles on the imaginary axis at the Matsubara frequencies ν_r . The summations over r in Eq. (2.19) can be written as a contour integral using

Cauchy's theorem:

$$\langle \mathbf{X}\mathbf{X}^T \rangle_C = \frac{\beta\hbar/2}{2\pi i} \times \frac{2}{\beta} \int_C dz \coth(\beta\hbar z/2) \hat{\mathbf{G}}(-iz). \quad (2.25)$$

The contour of integration is chosen to encircle the upper-half plane in a counter-clockwise direction. The poles on the imaginary axis at Matsubara frequencies ν_r for $r \geq 1$ are encircled, but only half of the pole at the origin is enclosed. The arc of the contour does not contribute to the integral when the radius is taken to infinity. Hence we can write this expression as an integral on the real line. Furthermore, by the symmetry of the integrand, the real part vanishes and the integral is given by:

$$\langle \mathbf{X}\mathbf{X}^T \rangle_C = \frac{\hbar}{2\pi} \int_{-\infty}^{+\infty} d\omega \coth(\beta\hbar\omega/2) \text{Im}[\hat{\mathbf{G}}(-i\omega)]. \quad (2.26)$$

A similar argument can be used to derive:

$$\langle \mathbf{P}\mathbf{P}^T \rangle_C = \frac{\hbar}{2\pi} \int_{-\infty}^{+\infty} d\omega \omega^2 \coth(\beta\hbar\omega/2) \text{Im}[\hat{\mathbf{G}}(-i\omega)]. \quad (2.27)$$

Eqs. (2.26,2.27) are identical to the results obtained by [86] for the asymptotic values of variances corresponding to an uncorrelated initial state. Therefore, we have proven that the single-time correlations of the open system relax to those of the closed system thermal state.

2.2.4 Equivalence of multi-time correlations

In this section we generalize the results of the previous section to include multi-time correlations. We begin by calculating the two-time correlation function $\langle \mathbf{X}(t)\mathbf{X}(t')^T \rangle_C$ using the trajectories obtained from the Langevin equation. Note that for the closed system thermal state this quantity is stationary. To simplify the

proof we make use of this observation and take the late-time limit of the closed system thermal state as well without loss of generality. This trick makes the comparison of the two cases easier and reduces the amount of computation.

The dynamics of the system is given by the solution (1.42) of the Langevin equation, which is valid for any initial state. The dependence on initial state is hidden in the correlations between \mathbf{X}_0 , \mathbf{P}_0 and $\boldsymbol{\xi}(t)$. The two-time position correlation is given by

$$\begin{aligned}
\langle \mathbf{X}(t)\mathbf{X}(t')^T \rangle &= \dot{\mathbf{G}}(t)\mathbf{M} \langle \mathbf{X}_0\mathbf{X}_0^T \rangle_C \mathbf{M}\dot{\mathbf{G}}(t') + \mathbf{G}(t) \langle \mathbf{P}_0\mathbf{P}_0^T \rangle_C \mathbf{G}^T(t') \\
&+ \dot{\mathbf{G}}(t)\mathbf{M} \int_0^{t'} ds' \langle \mathbf{X}_0\boldsymbol{\xi}(s')^T \rangle_C \mathbf{G}(t'-s') + \int_0^t ds \mathbf{G}(t-s) \langle \boldsymbol{\xi}(s)\mathbf{X}_0^T \rangle_C \mathbf{M}\dot{\mathbf{G}}(t') \\
&+ \mathbf{G}(t) \int_0^{t'} ds' \langle \mathbf{P}_0\boldsymbol{\xi}(s')^T \rangle_C \mathbf{G}(t'-s') + \int_0^t ds \mathbf{G}(t-s) \langle \boldsymbol{\xi}(s)\mathbf{P}_0^T \rangle_C \dot{\mathbf{G}}(t') \\
&+ \int_0^t ds \int_0^{t'} ds' \mathbf{G}(t-s) \langle \boldsymbol{\xi}(s)\boldsymbol{\xi}(s')^T \rangle_C \mathbf{G}(t'-s'). \tag{2.28}
\end{aligned}$$

As mentioned earlier, unlike the uncorrelated initial state, the terms in the second and third lines do not vanish in the closed system thermal state. We consider the case where

$$\lim_{t \rightarrow \infty} \mathbf{G}(t) = \lim_{t \rightarrow \infty} \boldsymbol{\gamma}(t) = 0. \tag{2.29}$$

These are the criteria for dissipative dynamics. Under these assumptions the first two terms in Eq. (2.28) vanish in the late-time limit for any initial state. The terms in the second and third lines have one factor of $\mathbf{G}(t)$ or $\dot{\mathbf{G}}(t)$, that goes to zero in the late-time limit, multiplied by a convolution integral. In Appendix A.4 we show that these convolution integrals are finite. Hence, the terms in second and third lines also vanish asymptotically. Finally we show the equivalence of the term in the last

line for the uncorrelated and thermal initial states at late times in Appendix. [A.5](#).

The comparison of more general multi-time correlations can be done similarly using the trajectories of the Langevin equation. The above example demonstrates how in the late-time limit the effects of initial conditions of the system die out and the noise statistics of both preparations converge. The equivalence at the level of trajectories ensures that all the multi-time correlations will be identical.

Let us reiterate the result we just obtained: a linear system linearly coupled to a linear thermal reservoir (with uncountably many degrees of freedom) at inverse temperature β does relax to the equilibrium state described by [\(2.4\)](#). This state is different from the Boltzmann state given by [\(2.3\)](#) whenever the interaction between the system and environment is not negligible. Moreover the multi-time correlations of system observables also relax to their corresponding values in the closed system thermal state.

2.2.5 The effect of coarse graining

Up until this point we only focused on the system degrees of freedom. Now we turn our attention to the environment. Following Ref. [\[51, 86\]](#), the trajectories of the environment oscillators, as driven by the system oscillators, are given by

$$\mathbf{x}(t) = \left[\dot{\mathbf{f}}(t)\mathbf{m}\mathbf{x}(0) + \mathbf{f}(t)\mathbf{p}(0) \right] + \mathbf{f}(t) * \mathbf{g}\mathbf{X}(t), \quad (2.30)$$

in terms of their free propagator $\mathbf{f}(t)$ given by Eq. [\(1.27\)](#). Into Eq. [\(2.30\)](#) we substitute the system trajectories, which are damped oscillations driven by noise for the

continuum environment:

$$\mathbf{X}(t) = \left[\dot{\mathbf{G}}(t) \mathbf{M} \mathbf{X}(0) + \mathbf{G}(t) \mathbf{P}(0) \right] + \mathbf{G}(t) * \boldsymbol{\xi}(t). \quad (2.31)$$

We then find the environmental dependence upon the initial state of the system to be

$$\mathbf{x}(t) = \mathbf{f}(t) \mathbf{g} * \left[\dot{\mathbf{G}}(t) \mathbf{M} \mathbf{X}(0) + \mathbf{G}(t) \mathbf{P}(0) \right] + \dots, \quad (2.32)$$

with all additional terms only dependent upon the initial state of the environment. The system-dependent terms correspond to a convolution of harmonic oscillations of the environment with non-locally damped oscillations of the system. Resolving these integrals leads to some terms which oscillate with environment frequencies ω and do not decay.

As a simple example, consider the local damping of a single system oscillator.

The open-system propagator or Green's function is given by

$$G(t) = \frac{\sin(\tilde{\Omega}t)}{M\tilde{\Omega}} e^{-\gamma_0 t}, \quad (2.33)$$

$$\tilde{\Omega} = \sqrt{\Omega^2 - \gamma_0^2}. \quad (2.34)$$

The environment's dependence upon the initial state of the system is given by

$$x_k(t) = \left\{ X(0) \frac{d}{dt} + \frac{P(0)}{M} \right\} \left\{ \frac{d^2}{dt^2} - 2\gamma_0 \frac{d}{dt} + \Omega^2 \right\} h_k(t) + \dots, \quad (2.35)$$

$$h_k(t) \equiv \frac{g_k f_k(t)}{(\omega_k^2 - \Omega^2)^2 + 4\gamma_0^2 \omega_k^2}, \quad (2.36)$$

plus terms that decay exponentially and the terms which depend upon the initial state of the environment. The function $h_k(t)$ oscillates forever, the same as $f_k(t)$, and therefore the environment retains information pertaining to the initial state of

the system forever. However, this information is not measurable forever. The system only interacts with the integrated trajectories, which resolve to a convolution of the damping kernel and open-system propagators.

$$\mathbf{g}^T \mathbf{x}(t) = -2\dot{\gamma}(t) * \left[\dot{\mathbf{G}}(t) \mathbf{M} \mathbf{X}(0) + \mathbf{G}(t) \mathbf{P}(0) \right] + \dots, \quad (2.37)$$

and, upon integrating over a continuum of environment frequencies (here performed by multiplication with the infinite matrix \mathbf{g}^T), the oscillatory terms decay in time. Thus, the late-time limit and coarse graining together are responsible for the erasure of all information pertaining to the initial state of the system.

2.3 General systems

Here we consider the single-time correlations of a discrete or nonlinear quantum system with arbitrary (linear or nonlinear) coupling to a quantum thermal environment, but under the assumption that the influence of the environment on the open system may be treated perturbatively. First we derive the second-order steady state, as much as is possible, from the second-order master equation. Second we derive the reduced thermal state directly from the closed-system thermal state. Finally we derive the reduced thermal state via canonical perturbation theory, for the case of zero temperature. All of these formalisms will be shown to produce equivalent results where valid.

2.3.1 Steady state

The time-evolution of the reduced density matrix of the open system can be generated by a perturbative master equation

$$\dot{\rho}_S(t) = \mathcal{L}(t)\{\rho_S(t)\}, \quad (2.38)$$

where the Liouville operator can be expanded in terms of the interaction Hamiltonian by a variety of methods [52, 87–89].

$$\mathcal{L}(t) = \mathcal{L}_0 + \mathcal{L}_1(t) + \mathcal{L}_2(t) + \cdots, \quad (2.39)$$

$$\mathcal{L}_0\{\rho\} = -i[\mathbf{H}_S, \rho], \quad (2.40)$$

In general, $\mathcal{L}_1(t)$ can be absorbed into the system Hamiltonian, and so we will primarily concern ourselves with the second-order term. For simplicity we will assume there is no degeneracy or near-degeneracy in the system energy spectrum; generalization to degenerate or nearly-degenerate systems is straightforward.

Expanding the interaction Hamiltonian in terms of system \mathbf{L}_n and environment \mathbf{l}_n operators

$$\mathbf{H}_I = \sum_n \mathbf{L}_n \otimes \mathbf{l}_n, \quad (2.41)$$

the multivariate master equation can be represented [52]

$$\mathcal{L}_2\rho = \sum_{nm} [\mathbf{L}_n, \rho(\mathbf{A}_{nm} \diamond \mathbf{L}_m)^\dagger - (\mathbf{A}_{nm} \diamond \mathbf{L}_m)\rho], \quad (2.42)$$

where the \mathbf{A} operators and \diamond product define the second-order operators

$$(\mathbf{A}_{nm} \diamond \mathbf{L}_m)(t) \equiv \int_0^t ds \alpha_{nm}(t, s) \{\mathcal{G}_0(t, s) \mathbf{L}_m(s)\}, \quad (2.43)$$

in terms of the zeroth-order (state) propagator of the system

$$\mathcal{G}_0(t, s)\{\boldsymbol{\rho}\} = e^{-\imath(t-s)\mathbf{H}_S} \boldsymbol{\rho} e^{+\imath(t-s)\mathbf{H}_S}, \quad (2.44)$$

and the (multivariate) environmental correlation function

$$\alpha_{nm}(t, s) \equiv \langle \mathbf{l}_n(t) \mathbf{l}_m(s) \rangle_E. \quad (2.45)$$

The second-order operator can be expressed as the Hadamard product

$$\langle \omega_i | \mathbf{A}_{nm} \diamond \mathbf{L}_m | \omega_{i'} \rangle = A(\omega_i - \omega_{i'}) \langle \omega_i | \mathbf{L}_m | \omega_{i'} \rangle, \quad (2.46)$$

where $|\omega_i\rangle$ are the energy eigenbasis for the isolated system. In the late-time limit, the second-order coefficients resolve

$$A_{nm}(\omega) = \frac{1}{2} \tilde{\alpha}_{nm}(\omega) - \imath \mathcal{P} \left[\frac{1}{\omega} \right] * \tilde{\alpha}_{nm}(\omega), \quad (2.47)$$

where $\tilde{\alpha}(\omega)$ denotes the Fourier transform of the stationary environment correlation function $\boldsymbol{\alpha}(t - s) = \boldsymbol{\alpha}(t, s)$, \mathcal{P} the Cauchy principal value and $*$ the appropriate Fourier convolution.

With the multivariate master equation detailed, we can prove relation (2.4) to second-order in the interaction. This generalizes the univariate proof in Ref. [84], which considered a single tensor-product interaction between the system and environment. As the proof is straightforward in either case, we will give an outline and focus upon differences which arise in the multivariate treatment.

We are looking for the stationary state $\boldsymbol{\rho}_\beta$, such that

$$\mathcal{L}\{\boldsymbol{\rho}_\beta\} = 0, \quad (2.48)$$

we know from detailed balance that the zeroth-order stationary state is the thermal state (2.3), e.g. see [52]. Second-order corrections can be generated from the second-order master equation via canonical perturbation theory. More explicitly, we have

$$\langle \omega_i | \boldsymbol{\rho}_\beta | \omega_j \rangle_{i \neq j} \propto e^{-\beta \omega_i} \delta_{ij} - i \frac{\langle \omega_i | \mathcal{L}_2 \{ e^{-\beta \mathbf{H}} \} | \omega_j \rangle}{\omega_i - \omega_j}, \quad (2.49)$$

but only for the denoted off-diagonal perturbative corrections (in the energy basis $|\omega\rangle$). As explained in Ref. [90], due to unavoidable degeneracy, specifically that the diagonal elements are all stationary to zeroth-order, the second-order master equation cannot determine the second-order corrections to the diagonal elements of the density matrix. Calculating these second-order diagonal terms would require knowledge of the fourth-order master equation, and, unfortunately, the general fourth-order master equation has never been rendered to the degree of tractability that the second-order master equation has.

By a simple application of the multivariate master equation to Eq. (2.49), we easily obtain these second-order corrections to the thermal state of the system. Corrections to the steady state can be represented

$$\langle \omega_i | \boldsymbol{\delta \rho}_\beta | \omega_j \rangle = \sum_{nmk} \frac{C_{ijk}^{nm}}{Z_0(\beta)} \langle \omega_i | \mathbf{L}_m | \omega_k \rangle \langle \omega_k | \mathbf{L}_n | \omega_j \rangle, \quad (2.50)$$

where $Z_0(\beta)$ is the partition function of the free system and with the off-diagonal (and non-resonant) coefficients given by

$$\begin{aligned} C_{ijk}^{nm} |_{\omega_i \neq \omega_j} = & \text{An} \left[e^{-\beta \omega_k} \frac{A_{nm}(\omega_{ik}) - A_{nm}(\omega_{jk})}{\omega_i - \omega_j} \right] \\ & + \text{An} \left[\frac{e^{-\beta \omega_i} A_{mn}(\omega_{ki}) - e^{-\beta \omega_j} A_{mn}(\omega_{kj})}{\omega_i - \omega_j} \right], \end{aligned} \quad (2.51)$$

where $\omega_{ij} = \omega_i - \omega_j$ and $A_{nm}(\omega)$ are the second-order master equation coefficients in (2.47). “An” denotes the anti-Hermitian part; the Hermitian and anti-Hermitian parts are defined

$$\text{He}[Q_{nm}] \equiv \frac{1}{2}(Q_{nm} + Q_{mn}^*), \quad (2.52)$$

$$\text{An}[Q_{nm}] \equiv \frac{1}{2}(Q_{nm} - Q_{mn}^*), \quad (2.53)$$

and for univariate noise (one collective coupling to the reservoir) the Hermitian and anti-Hermitian parts are simply the real and imaginary parts. In either case the anti-Hermitian part of (2.47) is the second term.

2.3.2 Equilibrium state

We wish to compare the straightforward expansion of (2.49) to the reduced closed system thermal state at second-order, and so we require a perturbative expansion of (2.4). There exists such a perturbative expansion of exponential matrices utilizing the identity

$$\frac{d}{d\epsilon} e^{\mathbf{A} + \epsilon \mathbf{B}} = e^{\mathbf{A} + \epsilon \mathbf{B}} \int_0^1 du e^{-u(\mathbf{A} + \epsilon \mathbf{B})} \mathbf{B} e^{+u(\mathbf{A} + \epsilon \mathbf{B})}, \quad (2.54)$$

to obtain an operator-Taylor series in the perturbation $\epsilon \mathbf{B}$. After a fair amount of simplification, one can determine the second-order stationary state to be

$$\rho_{\mathcal{B}} \propto e^{-\beta \mathbf{H}_S} + e^{-\beta \mathbf{H}_S} \int_0^\beta d\beta' \int_0^{\beta'} d\beta'' \langle \underline{\mathbf{H}}_I(-i\beta') \underline{\mathbf{H}}_I(-i\beta'') \rangle_{\mathbf{E}}, \quad (2.55)$$

in terms of the complex-time operators

$$\underline{\mathbf{H}}_I(-i\beta) \equiv e^{+\beta(\mathbf{H} + \mathbf{H}_E)} \mathbf{H}_I e^{-\beta(\mathbf{H} + \mathbf{H}_E)}, \quad (2.56)$$

where the noise average is taken with respect to the free thermal state of the environment, and factors inside the environmental trace have been written to suggest their correspondence with the environmental correlation function evaluated at imaginary times. Finally, note that the weak coupling expansion of the thermal state has the potential for secular behavior in β , due to the fact that a factor of β necessarily accompanies every factor of the interaction. Therefore, some terms in the expansion will only be accurate in the high-temperature regime if they retain polynomial dependence in β after integration.

The double integrals in Eq. (2.55) reduce to

$$\sum_{nm} \int_0^\beta d\beta' \int_0^{\beta'} d\beta'' \alpha_{nm}(-\imath\beta', -\imath\beta'') \underline{\mathbf{L}}_n(-\imath\beta') \underline{\mathbf{L}}_m(-\imath\beta''), \quad (2.57)$$

in terms of the complex-time operators

$$\underline{\mathbf{L}}(-\imath\beta) \equiv e^{+\beta \mathbf{H}_S} \mathbf{L} e^{-\beta \mathbf{H}_S}. \quad (2.58)$$

After a Fourier expansion of the complex-time correlation functions, expressions (2.49) and (2.57) can be compared term-by-term in the energy basis wherein the imaginary-time integrals of Eq. (2.57) can be resolved as the master equation operators were. Though the two expressions will then be composed of the same objects, they will not immediately appear to be equivalent. The final step is to apply the relevant multivariate Kubo-Martin-Schwinger (KMS) relations (also found in [52])

$$\tilde{\boldsymbol{\alpha}}(+\omega) = \tilde{\boldsymbol{\alpha}}^\text{T}(-\omega) e^{-\beta\omega} = \tilde{\boldsymbol{\alpha}}^*(-\omega) e^{-\beta\omega}, \quad (2.59)$$

and then one can see that the two expressions are equivalent in their off-diagonal elements. Moreover, as can be seen in (2.51), the off-diagonal expressions are free

of behavior secular in β and are, therefore, valid in the low-temperature regime.

Whereas the second-order diagonal corrections to the steady state could not be obtained from the second-order dynamics due to unavoidable degeneracy, there is no such obstruction for the equilibrium state here. As studied in [91], these terms can be obtained by analytic continuation

$$C_{iik}^{mm} = \lim_{\omega_i \rightarrow \omega_j} C_{ijk}^{mm}, \quad (2.60)$$

$$C_{iik}^{mm} = \frac{d}{d\omega_i} \text{An}[e^{-\beta\omega_k} A_{nm}(\omega_{ik}) + e^{-\beta\omega_i} A_{mn}(\omega_{ki})]. \quad (2.61)$$

However, notice that the second term will contain a $(d/d\omega)e^{-\beta\omega} = -\beta e^{-\beta\omega}$, and therefore this term is secular in β . So whereas the diagonal corrections of the second-order steady state could not be determined from the second-order master equation, here they can be determined, but they are only generally valid at high temperature. Despite this, [91] reported good agreement for a harmonic oscillator at low temperature.

2.3.3 Zero-Temperature Analysis

Though correspondence was established where valid, the previous analysis was seen to be insufficient for the complete calculation of low-temperature equilibrium states of the open system. However, as we shall now show, at least for zero-temperature noise, it is still possible to easily construct the reduced closed system thermal states in terms of the same environmental correlation functions which occurred in the previous analysis. The following relations were applied towards the inspection of two-level atoms interacting via a zero-temperature quantum field

in [92].

In the zero-temperature regime we can apply mundane perturbation theory to derive the stationary-state perturbations. One merely considers the perturbed ground state of the system + environment

$$\boldsymbol{\psi} = \boldsymbol{\psi}_0 + \boldsymbol{\psi}_1 + \boldsymbol{\psi}_2 + \cdots, \quad (2.62)$$

$$\boldsymbol{\psi}_0 \equiv |0\rangle \otimes |0\rangle_{\text{E}}, \quad (2.63)$$

and then traces out the environment

$$\boldsymbol{\rho}_\beta = |0\rangle\langle 0| + \left\langle \boldsymbol{\psi}_2 \boldsymbol{\psi}_0^\dagger + \boldsymbol{\psi}_1 \boldsymbol{\psi}_1^\dagger + \boldsymbol{\psi}_0 \boldsymbol{\psi}_2^\dagger \right\rangle_{\text{E}} + \cdots, \quad (2.64)$$

where we neglect the first moment of the reservoir as previously discussed. Without loss of generality let us set the ground-state energy of the system to zero. The calculation of the reduced state is then a straightforward application of canonical perturbation theory with some coarse graining. In doing this we obtain the same off-diagonal corrections (2.51), however for the diagonal (and similarly, resonant) corrections we obtain

$$C_{iik}^{nm} = \text{An} \left[e^{-\beta\omega_k} \frac{d}{d\omega_i} A_{nm}(\omega_{ik}) + e^{-\beta\omega_i} \frac{d}{d\omega_i} A_{mn}(\omega_{ki}) \right], \quad (2.65)$$

where the Boltzmann weights are guessed, as these relations have only been derived here at zero temperature. Note that the second term here is different from its analytically-continued value in (2.61). Whereas the analytically-continued values may diverge in the zero-temperature limit, obviously these values cannot. Therefore Eq. (2.65) is exact for zero-temperature and our best guess for the positive-temperature coefficients: it has the correct functional dependence upon the Boltz-

mann weight and fourth-order master equation coefficients. At worst this is an interpolation of the zero and high-temperature states.

2.4 Discussion

In this chapter we investigated the equilibrium states of open quantum systems from dynamics / non-equilibrium point of view. We show that starting from a product state (2.2) the open system which results from coarse graining the environment will evolve to a late-time steady state. This state can be expressed as the reduced state of the closed system thermal state at the temperature of the environment, i.e. Eq. (2.4). This result is important when the system-environment coupling is not negligible⁷, or alternatively, when relaxation rates are not insignificant in relation to the system frequencies. In this case the stationary state of the system (2.4) differs from the canonical Boltzmann state (2.3), although we have not focused on this difference in this chapter (see Hilt *et al.* [93] for an example). One might argue that this state is the closest one can get to thermalization in the strong coupling regime.⁸ However in this work we use the term equilibrium state for Eq. (2.4) and reserve the term thermal state to the standard Boltzmann form (2.3).

Our proof is exact for the linear model and to second-order in interaction

⁷Based on the discussion of Fig. 2.2, we expect our results to be most relevant to small systems.

⁸Alternatively one could define this state to be *the* thermal state in the strong coupling regime. However this state depends on the specifics of the reservoir and the coupling to the reservoir. Hence it is not specified by the system parameters alone and referring to it as *the* thermal state is, in our opinion, misleading.

strength for nonlinear models. Moreover, for the exactly solvable linear case we prove the equivalence of multi-time correlations. The issue of multi-time correlations in the context of equilibration/thermalization seems to be mostly ignored in the literature. We argue that multi-time correlations are important outside the Markovian regime, as was pointed out in [94]. For instance, the relaxation of multi-time correlations cannot be deduced from the relaxation of the reduced density matrix of the system, neither can the explicit value of the multi-time correlations be derived from the equilibrium state, if the dynamics is non-Markovian. In this respect our analysis of the linear N-QBM model provides insight into equilibration phenomena beyond the density matrix formalism.

An essential ingredient of our proofs is the continuum limit for the environment. For a finite environment the $t \rightarrow \infty$ limit of the reduced state does not exist within the formalism presented here and another ingredient is necessary to ensure relaxation to equilibrium. Having classical molecular dynamics in mind [7], we entertain the possibility that quantum chaos might be one avenue to explore [35].

On the other hand we can consider a large but finite environment. It can be argued that for any relevant times $t > 0$ the effect of an infinite reservoir can be approximated arbitrarily closely by a large but finite reservoir. Then equilibration is observed for the time-interval between the relaxation time and the recurrence time. Note that this interval is huge for a large environment, since the recurrence time grows very rapidly with the number of degrees of freedom. As a result the system stays close to its equilibrium most of the time. This interpretation helps us touch base with the results of [36, 37, 76] where relaxation in finite systems is proven for

time averaged quantities.

2.4.1 Comparison with recent literature

To put this work in developmental context, here we compare more specifically our results to that of Linden et al. [36], Reimann [37], and Short and Ferrelly [76]⁹. All these works have in common with us the set-up of a small system coupled to a large environment and relaxation is achieved dynamically via time-evolution. A major difference is the choice of initial conditions: they allow for any initial state, which is spread over sufficiently many energies, whereas we restrict our environment to be in a thermal state. In turn we can derive the form of the equilibrium state explicitly.

Unlike what is done here these authors all make the assumption of non-degenerate energy gaps (this assumption is relaxed to a certain degree in [76]) and assume finite dimensional Hilbert spaces. The linear model we solved exactly here has infinitely degenerate energy gaps and we considered a reservoir consisting of an infinite number of degrees of freedom. Ref. [36] considers only pure states for the closed system (in the spirit of [33, 34]). Finally they all define relaxation in terms of time averaged quantities, i.e. systems behave as if they are in their steady state most of the time. Ref. [76] also provides upper limit for the relaxation time.

The proofs of [36, 37, 75, 76] rely on the much greater dimensionality of the Hilbert space of the environment compared to that of the system. The system +

⁹See Sec. 2.1.1 for the clarification of the different use of the term *equilibration* in the literature and here.

environment state is propagated as a whole using unitary dynamics. The fact that the environment is large is utilized in the tracing out of the environment at the end of time evolution. In this derivation the effect of the environment on the system dynamics is not so easily accessible.

In our proof, the fact that the environment consists of a large number of degrees of freedom manifests itself in the form of its decaying correlations. These correlations in turn determine the non-unitary aspects of the open system dynamics. We use this non-unitary open system dynamics to evolve the reduced state of the system to its equilibrium state. In particular we do not refer to the state of the closed system explicitly¹⁰. Our derivation is more in the idioms of open quantum systems paradigm, where the influence of the environment on the system dynamics can be continuously monitored and explicitly expressed (e.g., consistent back-reaction from the environment is fully embodied in the influence functional [26]).

Relaxation is demonstrated in [36,37,76] for very general Hamiltonians, including strong coupling between the system and the environment. In their derivation the strong coupling regime does not present any extra difficulty. In the open system approach we adopted in this dissertation strong coupling is difficult to handle. On the other hand, as a benefit of our method we can describe the nature of the equilibrium state, i.e. Eq. (2.4), besides proving its existence and uniqueness.

¹⁰Except for Sec. 2.2.5, where we do look at the individual environmental modes just to make the point that the closed system (S + E) does not equilibrate.

Chapter 3: Quantum fluctuation theorems

3.1 Introduction

Compared to equilibrium statistical physics, few theorems of generality are established for nonequilibrium systems. Hence any valid statement with a broad spectrum of implications and wide range of applications is of great value. The fluctuation theorems of Jarzynski [11] and Crooks [14] in nonequilibrium statistical mechanics are of such a nature and have stimulated intense research interest and activities in the past decade and a half. Both theorems were already introduced in Sec. 1.5.1 but we recite them here for convenience. The fluctuation theorem of Jarzynski:

$$\langle e^{-\beta W} \rangle = e^{-\beta \Delta F}. \quad (3.1)$$

The fluctuation theorem of Crooks:

$$\frac{P(W)}{\tilde{P}(-W)} = e^{\beta(W - \Delta F)}. \quad (3.2)$$

Fluctuation theorems were originally formulated for classical systems and their derivation relied heavily on the concept of a phase space, wherein the system evolution can be described as a trajectory. Quantities of interest for thermodynamical considerations appearing in these theorems like work, heat, entropy etc. were given

microscopic definitions based on trajectories. There are even detailed fluctuation theorems about distributions of trajectories themselves [54,95,96].

In standard quantum mechanics, states are described by vectors living in a Hilbert space. Observables, including the Hamiltonian, are associated with Hermitian operators. Trajectories in phase space are not part of this description.¹ This mismatch between classical mechanics and the standard formulation of quantum mechanics hindered a straightforward derivation of quantum fluctuation theorems based on the classical derivation. It is then natural to ask if FTs hold for quantum systems, and if not, under what conditions would they fail, and whether there exist quantum fluctuation theorems (QFTs) different in form and content from the classical FTs. If this is not possible, can one identify corrections due to quantum mechanical effects?

How to define work in quantum physics? In the derivation of fluctuation theorems of Jarzynski and Crooks, henceforth referred to as FTs in short, the main conceptual obstacle is how to make sense of work in a quantum setting. To begin with, work is not an observable [97], and as such, treating it as a quantum mechanical operator [98,99] is largely a computational convenience. Thus the foremost task is to find a physically meaningful definition and an operationally feasible way to calculate it. We will address this issue with a new approach described below.

Let us try to appreciate the content of this pivotal point. In classical mechanics *exclusive work* [61] imparted to a system, say a particle, is defined as the integration

¹Although the Wigner representation can cast quantum mechanics in a phase space picture, this analogy is not perfect as argued in section 1.2.2.

of applied force on the system with displacement along a path. The force is exerted by an external agent which causes the system to move along a trajectory. Once one knows the trajectory, work can be calculated, but the difficulty for quantum system is that particles don't follow trajectories, they are described by a wave function which is a very different notion and entity from paths. The key challenge is to make sense of trajectories in quantum physics. We mention several approaches below and then present our own.

Closed versus open quantum system. If one restricts one's attention to closed quantum systems, i.e., isolated quantum systems having no interaction with any of their environments, one can define work via two-time energy measurements discussed in Sec. 1.5.2, and general agreement seems to be reached. However this is merely an idealization of realistic physical systems which are more often open. The influence of their environments which the systems of interest interact with need be accounted for in the open system's evolution. Even in the simplest cases when one talks about temperature or refers to (equilibrium) thermodynamic quantities a heat bath (canonical ensemble) or a particle reservoir (grand canonical ensemble) is implicitly assumed, which are open-system setups.

Since for closed quantum systems fluctuation theorems can be easily derived, one can think of the system + environment as a closed system and work out the QFTs. This was done in [63, 100, 101]. However, this formulation has the innate shortcoming that the work defined therein requires the energy measurement of the *combined* system at two times while the FTs refer to the work done on the *open* system of interest. This shortcoming is remedied in our treatment by adopting the

open quantum systems approach. Before going into the details of our treatment we give a brief account of the literature on FTs in open quantum systems (OQS).

Open quantum system fluctuation theorems in the literature. Using microscopic models and open system dynamics several suggestions for trajectories have been made. For example, De Roeck [102] used the unraveling of the open system master equation and compared his results to that of the closed system approach. Deffner et al. [103] used the quantum Smoluchowski equation (QSE), which was derived from taking the high friction and high temperature limit in [104], as a starting point. They considered the solution to the QSE in terms of classical path integrals and interpreted these paths as trajectories. But these trajectories are difficult to interpret physically, being more in the nature as devices (to help solve a differential equation) than actual physical entities. By making the assumption that the reduced dynamics of a driven open quantum subsystem is described by a quantum master equation Esposito and Mukamel [105] recast its solution in a representation which takes the form of a birth-death master equation (BDME) with time-dependent rates and used it to define “quantum” trajectories. But these QSE and BDME, just as the Pauli master equation, govern transition probabilities, are equivalent to a reduced density matrix with only diagonal elements, and thus contain no quantum phase information ².

²This may be viewed as the completely decohered end product of a decoherent history or environment-induced decoherence process (complete diagonalization of the reduced density matrix) but as we shall explain in more detail below, it corresponds to the case of very strong noise acting on the subsystem, which is possible for high temperatures, and thus it falls under the parameter regime where the classical FTs are valid. In fact for Gaussian systems, the QFT derived under

Alternatively Crooks [106] proved the Jarzynski equality by considering the Markovian dynamics of a quantum system in the following setting: Instead of measuring the system, generalized measurement superoperators were used to represent measurements of heat flow. If the quantum environment is assumed to be large, to have rapidly decohered and always remain at thermal equilibrium, plus being uncorrelated and unentangled with the system, then the change in energy of the bath can be measured without further disturbing the dynamics of the system.

Horowitz [107] adopted the strategy of Crooks to study a model similar to ours: a one-dimensional forced harmonic oscillator weakly coupled to a thermal reservoir. Unlike the QBM model, there the environment is engineered by weakly coupling the harmonic oscillator to a sequence of two-level systems. The evolution of the harmonic oscillator is monitored and the amount of energy exchanged with the environment is inferred by measuring the state of each two-level system after its interaction with the oscillator. Quantum trajectories are obtained from the stochastic Schrödinger equation, which governs the evolution of the harmonic oscillator conditioned on the measurement outcomes. Consistent trajectory-dependent definitions are introduced for work, heat, and entropy and a detailed fluctuation theorem is proven.

The treatment of [107] has the advantage of possible experimental realizations in cavity quantum electrodynamics. Its validity is limited to weak coupling regime and Markovian dynamics. The approach we develop in this chapter, on the other hand, will allow us to go beyond the weak coupling regime and study non-Markovian

these conditions have exactly the same form as the classical FTs.

effects as well. A correspondence between trajectories obtained from a stochastic Schrödinger equation and those emerging within decoherent histories framework has been established in Ref. [108, 109].

In comparison with earlier work our approach is closest in spirit to that of Chernyak and Mukamel [98]. However our methods (they use superoperators in Liouville space) and interpretations (they use von Neumann’s wave function collapse for quantum measurement) are different. We will detail the differences after we have a chance to describe our approach.

3.1.1 Our approach and findings

For the sake of conciseness we just state what we do and name the ingredients in our approach here, leaving more detailed explanations to the next section.

In this chapter we analyze the fluctuation theorems (FTs) using the exactly solvable microscopic quantum Brownian motion (QBM) model of a quantum harmonic oscillator coupled to a heat bath of N_E quantum harmonic oscillators with arbitrary spectral density function and for all temperatures (see Sec. 1.4 with $N_S = 1$ for details). This is referred to as a “general” environment in [28] where an exact master equation for these full ranges was obtained and where our discussions in the application of this model to QFTs are based upon ³. The low temperature results are of special interest for the derivation of QFTs since this is the regime where

³We advise against calling this a *non-Markovian environment*, because non-Markovian refers to stochastic *processes*, not systems. Instead, use, e.g., *colored noise environments*, which can engender non-Markovian dynamics in the open subsystem.

deviations from the classical FTs might be observed.

Decoherent history approach to define trajectories for quantum systems.

We resort to the conceptual framework of decoherent [110] or consistent [111, 112] histories (*dechis*) and the key notion of decoherence for understanding the process of quantum to classical transition. We believe this is the most faithful and intuitive way of defining trajectories or explaining how they arise from quantum mechanics. To be more precise, these trajectories are actually stochastic classical paths in a quasi-classical domain as a result of decoherence in the histories. They arise by the action of noise which is defined as variations in neighboring histories. (For a succinct explanation of the first point see e.g., [111, 113] and [114] on the second point.)

Environment-induced decoherence for explicit computations.

While the decoherent history paradigm is conceptually clear for explaining the origin and mechanisms in the emergence of classical stochastic trajectories, it is less versatile in actual computations. The environment-induced decoherence (*envdec*) approach can be of more practical use. Here, the approximate diagonalization of the reduced density matrix of the reduced or open system with respect to some basis is used as a signifier of decoherence of the quantum system in transit to classicality, whereby the notion of trajectory becomes viable. But which basis? This is the physically relevant issue. The quantum system is more readily decohered in the so-called “pointer basis” [115], which is affected by the form of interaction between the system and its environment. Here, with an explicit environment specified, it is easier to see how noise arises and its nature (colored, multiplicative [116]) than

in the *dechis* approach. The connection between these two approaches is discussed in [117]. An explicit model calculation (the QBM model) was given in the *dechis* approach [118] where one can compare these two approaches in operational details.

Significance of stochastic regime between quantum and classical.

In reference to trajectories of quantum origin, we notably attach the word “stochastic” to classical. This is because there is a stochastic component to them after the quantum histories decohere. They are described by a probability distribution function.⁴ Each such trajectory is a realization of this distribution. Taking the stochastic average of an ensemble of such trajectories will yield the unique classical path which is a solution of a deterministic classical equation of motion.

Decoherence is due to noise, quantum or thermal or both. In the *envdec* scheme, one can see this explicitly from the stochastic equations governing the open (reduced) system. Noise is responsible for quantum diffusion which brings forth decoherence. The stronger the noise, the more complete the decoherence process and the more classical the trajectories. In fact for the QBM model there are two diffusion terms: a normal diffusion dominates at high temperatures and an anomalous diffusion which dominates at low temperature. The latter is what one should focus on in marking the difference between the classical and the quantum FTs. Therefore

⁴ The stochastic component does not necessarily arise from the use of an ensemble. Even for a pure state of the universe, there is a stochastic component to the decoherent set of histories due to coarse graining which is related to the fact that the reduced state of a pure state is in general mixed. This is a fundamental difference between classical and quantum mechanics regarding the origin of probabilities in each theory.

the behavior of a system in the stochastic regime actually holds the key to quantum-classical transition or correspondence. It is particularly suitable for the exploration of FTs in open systems as they are also cast in a stochastic framework in terms of the probability distribution of work.

Our findings. In this chapter we present a first-principles analysis of the nonequilibrium work distribution and the free energy difference of a quantum system interacting with a general environment (with arbitrary spectral density and for all temperatures) based on a well-understood micro-physics (quantum Brownian motion) model under the conditions stipulated by the Jarzynski equality and Crooks' fluctuation theorem (FTs). We use the decoherent history conceptual framework to explain how the notion of trajectories in a quantum system can be made viable and use the environment-induced decoherence scheme to assess the strength of noise which could provide sufficient decoherence to warrant the use of trajectories to define work in open quantum systems. From the solutions to the Langevin equation governing the stochastic dynamics of such systems we were able to produce formal expressions for these quantities entering in the FTs, and from them prove explicitly the validity of the FTs at the high temperature limit. At low temperatures our general results could lead to the identification of a parameter range where FTs may not hold or need to be expressed differently. We explain the relation between classical and quantum FTs and the advantage of this micro-physics open-system approach over the phenomenological modeling and energy-level calculations for substitute closed quantum systems.

3.2 Key points and main ideas

We are seeking a derivation of quantum fluctuation theorems in nonequilibrium physics by applying concepts and practices in quantum foundation and measurement theory via decoherent histories and environment-induced decoherence with its ensuing classical stochastic equations. Classical and quantum fluctuation theorems have been introduced in Sec. 1.5.1 and Sec. 1.5.2, respectively. In this chapter we will present our formulation of quantum fluctuation theorems in terms of trajectories. To understand how trajectories can emerge in quantum mechanics we review the decoherent histories and environment-induced decoherence approaches to quantum-classical transition. For good reviews on this subject we mention [111, 112, 119].

3.2.1 Trajectories in classical mechanics

Consider a classical harmonic oscillator, without a bath. Initial position and momentum of the oscillator are sampled from the thermal phase space density. The rest of the trajectory is entirely determined by the protocol of how the external force is applied. Work is calculated using this deterministic trajectory according to eq.(1.48). However, deterministic trajectory is strictly a classical notion and cannot be applied to a general quantum mechanical system. A state that is sampled from the thermal density matrix in general does not have a well-defined position and momentum. Furthermore, the time evolution usually causes the wavefunction to spread further. We cannot talk about the quantum oscillator being at one point in space, having a certain velocity, and moving in a deterministic continuous trajectory

as a function of time.

Next consider the same classical model with a heat bath. For each realization of the protocol, the initial data for both the system oscillator and the bath are sampled from the initial phase space density. The initial data for the bath determines the noise for that particular realization. The system oscillator follows a trajectory determined by a combined action of the deterministic force $f(t)$ and the stochastic force $\xi(t)$. Although the noise is stochastic, each realization of the experiment corresponds to a unique noise and hence a unique trajectory. The definition of work in terms of trajectories is unaffected.

It is a simple yet subtle and deep point how the interaction with a bath would help to define a trajectory for a quantum particle. To understand this conceptually we adopt the decoherent or consistent histories viewpoint of quantum mechanics as described below.

3.2.2 Trajectories in quantum mechanics

Trajectories which are well-defined in classical mechanics are generally ill defined in quantum mechanics except under certain conditions. We shall spell out these conditions here. Let us begin with something simple, such as a quantum particle in motion. In a closed quantum system S , namely, a system subjected to no outside (environmental) influence except for its own quantum fluctuations, the closest entity to its “trajectory” is a wave packet moving with a certain group velocity but which also spreads in time due to the Heisenberg uncertainty relation between

the variance of the canonical variables, position and momentum in this case. The same system at a finite temperature is no longer closed because for it to exist at finite temperature it must be or have been in contact with a source with energy exchange or a bath B. The influence of the environment E (we call an E a bath B if it is infinitely large and is described by a thermal density matrix with inverse temperature β) has complicated and interesting consequences. This is the subject of open quantum systems.

There are at least two major effects an environment brings in “opening up” a closed quantum system: a) it turns the original Hamiltonian (unitary) dynamics to dissipative (nonunitary) dynamics – this refers to energy flow from the system to the environment, b) fluctuations in the environment decoheres the quantum system – this refers to quantum phases of the system being dispersed into the environmental variables. The latter is responsible for shaping the notion of trajectories in quantum system and there are precise conditions pertaining to the features of the environment (e.g. high temperature) whereby they become physically well-defined in a measurement. One way is to construct the reduced density matrix of the open quantum system and look at whether and how quickly its off-diagonal elements decay in time, leaving the system’s statistical state describable by an approximately diagonal density matrix with respect to some physically meaningful basis (related to measurement instruments and interaction, such as Zurek’s “pointer basis” [115]). This time, called decoherence time, marks the appearance of classical features, because after it is effectively decohered this open system is adequately described by probabilities rather than amplitudes, its quantum phase information is lost (more

accurately, dispersed into or shared by the multitude of environmental degrees of freedom). This process is captured by the stochastic equations, the most common forms are the master equation, the Langevin and the Fokker-Planck equations.

What distinguishes these equations is the presence of noise or fluctuations in the environment, and dissipative dynamics of the open (reduced) system, depicting the two distinct features of open system dynamics. In general two kinds of noise exist in any quantum system, the intrinsic quantum noise entering in the Heisenberg relation which exists for all systems including closed ones, and noise from a finite temperature bath ⁵. Both contribute to decohering a quantum system, although the thermal noise usually dominates at high temperatures.

There are many ways to characterize a quantum system as approaching its classical limit. The familiar cases are the correspondence principle, the Bohr-Sommerfeld rules in quantum mechanics, the description of Maxwell-Boltzmann distribution as limits of the Fermi-Dirac and the Bose-Einstein distributions, or the more simplistic $\hbar \rightarrow 0$ or “at high temperature” stipulations. One can show that the coherent state is the “most classical” of quantum states [120]. One can derive an uncertainty function at finite temperature [121] or equivalently calculate the entropy function and be able to demarcate the transition from the quantum noise- dominated regime to the thermal noise- dominated regime. There have been significant advances in the last two decades in our understanding of the quantum

⁵The noise due to bath itself has two components: one due to intrinsic quantum fluctuations in the bath, which are present even at zero temperature, and the other due to regular thermal fluctuations.

classical correspondence (see e.g., [122]). Decoherence is at the heart of the quantum to classical transition issue, and the main cause of it is noise of all forms, either in the fluctuations of the environment or in the separation of neighboring coarse grained histories, and in the precision of the measurement devices and procedures (see Sec. 3.3.1 for details). We will use the decoherent or consistent history [110–112] viewpoint for conceptual clarity, especially pertaining to the issue of trajectories but adopt the environment-induced decoherence (*envdec*) scheme for computations, as it is technically easier to manipulate.

3.2.2.1 Decoherence Functional in *Dechis* and Influence Functional in *Envdec*

The main idea of *dechis* approach is to define a history α by a set of projection operators $P_\alpha(t_k)$ acting at times t_k . As a special case we consider projections in position basis. These kind of histories are naturally implemented in the path integral approach. The projectors are represented by window functions $w_\alpha[x(t_k)]$, which take on unit value if the instantaneous configuration satisfies the requirement of the history α , and vanish otherwise. As a limiting case we mention a fine-grained history, for which the path is specified exactly at all times and is assigned an amplitude $\exp(iS/\hbar)$ as usual. It is useful to define the decoherence functional of two histories

α and $\tilde{\alpha}$ by [114]:

$$\mathcal{D}[\alpha, \tilde{\alpha}] = \int Dx Dy e^{i(S[x] - S[y])/\hbar} \rho(x(t_i), y(t_i); t_i) \left\{ \prod_k w_\alpha[y(t_k)] \right\} \left\{ \prod_l w_{\tilde{\alpha}}[x(t_l)] \right\}. \quad (3.3)$$

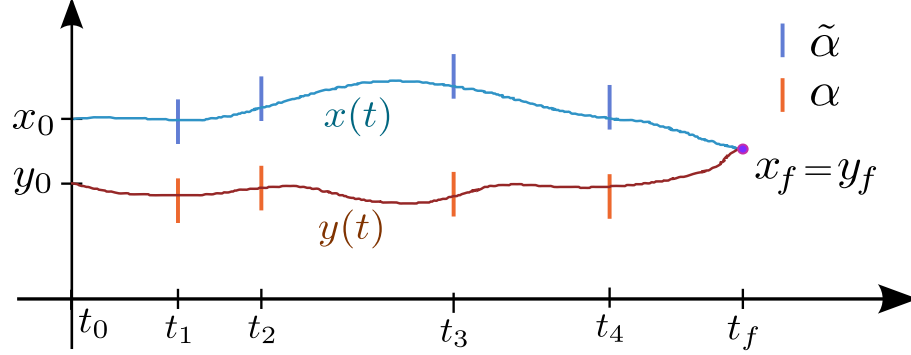


Figure 3.1: Depiction of a pair of paths in Eq. (3.3). Note that the paths have the endpoints at t_f common as required by the trace function.

The product over k and l can be discrete or continuous as is the case in Section (3.3.1). The probability of a given history α is given by the diagonal element of the decoherence functional: $P[\alpha] = \mathcal{D}[\alpha, \alpha]$. For classical trajectories it is required that the probability of a coarse grained history to be the sum of its constituents. For an arbitrary set of histories quantum interference effects lead to a violation of the probability sum rule: $P[\alpha \vee \tilde{\alpha}] = \mathcal{D}[\alpha, \alpha] + \mathcal{D}[\tilde{\alpha}, \tilde{\alpha}] + 2\text{Re}\mathcal{D}[\alpha, \tilde{\alpha}] \neq P[\alpha] + P[\tilde{\alpha}]$. If a set of histories can be identified for which the real part of the off-diagonal elements of the decoherence functional vanishes (or are much smaller than the diagonal elements for approximate decoherence) for *all pairs* of trajectories $\alpha, \tilde{\alpha}$ in the set, probabilities can be assigned to individual histories. The challenge is to identify the conditions under which, and to what extent, the decoherence condition is satisfied.

Technically the environment-induced decoherence (*envdec*) program is easier to implement, the relation between these two programs are explained or illustrated in [117, 118]. This is what we will do by way of the QBM model presented in the next section. We will argue that for histories obtained by coarse graining the environment sufficiently, and the system of interest to some extent (determined by the strength of noise), an approximate decoherence condition can be satisfied to the desired degree. At the other end, if quantum interference between particle histories continues to play a dominant role, decoherence is not consummated, the classical world is not reached and the concept of trajectories is ill-defined.

The quantum open system formulation, via the influence functional, provides one with a clear perspective in the organic relation between the processes of fluctuations / noise, correlation, decoherence and dissipation and how they enter in the transition from the quantum to the classical world with the intermediate stochastic and semiclassical regimes. While it is useful to explain this with the aid of stochastic equations which we will derive below, the key idea can be put succinctly: The stronger the effect of noise in the environment, the more efficiently it decoheres the quantum system and the clearer the classical notion of trajectory can be defined and used for the description of a quantum particle. The important new understanding is the existence of a stochastic regime between the quantum and the classical, and how quantum features are expressed in terms of classical stochastic variables ⁶.

⁶A famous case is the transcription of Gaussian quantum fluctuations in the environment as classical noise via the Feynman-Vernon identity [26].

3.2.2.2 Worldline Influence Functional Formalism

Thus far we learned that the decoherence of a quantum system due to the noise arising from a coarse grained environment is instrumental to the emergence of a classical world. How strongly the system is coupled to its environment(s), the nature of the noise from the environment and its temperature all enter in determining how completely the system is decohered, and there is always a stochastic component in the open system's dynamics governed by a Langevin equation or its (near) equivalent master or Fokker-Planck equations. Almost complete decoherence is a necessary condition for a classical description which, in this context, is what trajectories are predicated upon. Under this condition a powerful approach called the worldline (WL) influence functional (IF) formalism has been used effectively for more than two decades in nuclear / particle physics communities, see e.g., [123]. We shall only mention its key features so as to bring out its relevance to the present problem but skip all the details.

The influence functional technique of Feynman and Vernon [26], or the closely related closed-time-path effective action method of Schwinger [124] and Keldysh [125] are initial value (in-in) formulations which are particularly suitable for exploring the time evolution of many-body systems, unlike the S-matrix (in-out) formulation used for calculating scattering processes. In general this yields a nonlocal and nonlinear coarse grained effective action (CGEA) for the system's motion. The CGEA may be used to treat the nonequilibrium quantum dynamics of interacting particles. Take for example the QBM model: When the particle trajectory becomes

largely well-defined as a result of effective decoherence due to interactions with the environment, with some degree of stochasticity caused by noise, the CGEA can be meaningfully transcribed into a stochastic effective action, describing stochastic particle motion. The evolution propagator for the reduced density matrix of the open system is dominated by the particle trajectory given by the extremal solution of the real part of the CGEA. Stochastic fluctuations around the decohered semiclassical trajectories are described by the imaginary part of the CGEA. For further technical details, see [126, 127].

When the back-action of the environment is taken into account the dynamics of the open system will in general be non-Markovian as it contains memories, and the noise in the environment is generally colored, as it contains many time scales characterized by its spectral density and vary with temperature. Dissipation in the open system dynamics is controlled and balanced by the noise in the environment as manifested in the existence of fluctuation-dissipation relations between these two sectors. What is more important, because the influence action includes the back-action of the environment in a self-consistent manner, the worldline is not merely a prescribed classical entity, or a simple solution to an equation of motion at the tree level (in truth, with an ever-present stochastic component), but rather, a dynamical one, as the result of constant negotiation between the open system and its environments at all times. This is the special beauty of the IF method.

3.3 The QBM model

QBM model has been analyzed in Sec. 1.4 using the Wigner representation. In this section we provide an alternative derivation of the dynamics of the Wigner function due to [94] partly because of its elegance but also because of the importance of the techniques used in this derivation. Here the density matrix is propagated using configuration space path integrals and the back-action of the environment is encoded in the influence functional. Techniques similar to those used in this section are also used to derive the explicit form of the decoherence functional (3.20).

A closed quantum system can be partitioned into several subsystems according to the relevant physical scales. If one is interested in the details of one such subsystem, call it the distinguished, or relevant system, which interacts with the other subsystems comprising the environment, the details of which are not of interest, one can coarse grain the information in the environment but keep its overall influence on the distinguished subsystem of interest, thereby rendering it an open system. This influence is best captured by the influence functional technique of Feynman and Vernon [26] which we use here. We consider the QBM model with a time dependent external driving force $f(t)$:

$$H_S = \frac{P^2}{2M} + \frac{1}{2}M\Omega^2 X^2 - f(t)X, \quad (3.4)$$

Note that we did not use bold symbols for the position and momentum operators as well as mass and coupling constants above as opposed to Chapters 1 and 2. This is to emphasize the fact that for $N_S = 1$, regular QBM model, these operators are

not vectors and the constants are not matrices. This convention will be used in this chapter and the next.

The renormalization of the potential is the one shown in Eq. (1.22), which preserves the physical (observed) frequency of the system oscillator for any system-bath coupling. Moreover, without the renormalization term the potential might have no minimum and the thermal state might become ill-defined.

The combined system being closed, the Hamiltonian H_C gives rise to unitary evolution, its density operator ρ_C obeys the von-Neumann equation (1.3). However what we are interested in is how the system S behaves under the influence of its environment, in this case a heat bath B at temperature $1/\beta$. The state of the open system at any one time is completely specified by the reduced density matrix ρ_S defined in (1.17), which is obtained from the density matrix of the combined system by integrating out the bath degrees of freedom.

Because it incorporates the back-action of the environment the time evolution of the reduced density matrix of the open system is nonunitary and in general non-Markovian. The reduced density matrix of the system oscillator in position representation at t_f can be obtained from the reduced density matrix at some earlier time t_i via [128]:

$$\rho_S(x_f, x'_f, t_f) = \int dx_i dx'_i J(x_f, x'_f, t_f; x_i, x'_i, t_i) \rho_S(x_i, x'_i, t_i), \quad (3.5)$$

where J is the propagator. If the system and the bath are initially uncorrelated and the bath is in a Gaussian state the propagator J can be calculated exactly:

$$J(x_f, x'_f, t_f; x_i, x'_i, t_i) = \int_{x(t_i)=x_i}^{x(t_f)=x_f} Dx \int_{x'(t_i)=x'_i}^{x'(t_f)=x'_f} Dx' e^{\frac{i}{\hbar}(S_S[x] - S_S[x'] + S_{IF}[x, x'])}. \quad (3.6)$$

Above $\int Dx$ denotes Feynman's configuration space path integral. Now introduce the following notation: for functions $A(s), B(s)$ and kernel $K(s, s')$ define

$$A \cdot K \cdot B \equiv \int_{t_i}^{t_f} ds \int_{t_i}^{t_f} ds' A(s) K(s, s') B(s'). \quad (3.7)$$

In terms of the new variables the exponent appearing in Eq.(3.6) can be written as:

$$S_S[x] - S_S[x'] = -M\dot{X}(t_f)y_f + M\dot{X}(t_i)y_i + y \cdot (L_0 \cdot X - f), \quad (3.8)$$

$$S_{\text{IF}}[x, x'] = -y \cdot \mu \cdot X + \frac{i}{2} y \cdot \nu \cdot y, \quad (3.9)$$

where $L_0(t, t') = M(\frac{d^2}{dt^2} + \Omega^2)\delta(t - t')$ and $S_S[x]$ is the action associated with the trajectory x and the system Lagrangian. The kernels $\mu(s, s')$ and $\nu(s, s')$ are the dissipation and noise kernels, respectively. The former has been defined in (1.34) and the latter for a thermal environment in (2.5).

In the equivalent description in terms of the Wigner function one defines a reduced Wigner function \mathcal{W}_S in terms of the reduced density matrix formally in the same way as in Eq. (1.7). Using Eqs.(1.7-3.6) it can be shown that the reduced Wigner function evolves from time t_i to a later time t_f via

$$\begin{aligned} \mathcal{W}_S(X_f, P_f, t_f) &= \frac{1}{2\pi\hbar} \int dy_f e^{\frac{i}{\hbar} P_f y_f} \int dx_i dx'_i \int_{x(t_i)=x_i}^{x(t_f)=X_f-y_f/2} Dx \int_{x'(t_i)=x'_i}^{x'(t_f)=X_f+y_f/2} Dx' \\ &\times e^{\frac{i}{\hbar}(S_S[x]-S_S[x'] + S_{\text{IF}}[x, x'])} \int dp_i e^{-\frac{i}{\hbar} P_i (x'_i - x_i)} \mathcal{W}_S\left(\frac{x_i + x'_i}{2}, P_i, t_i\right). \end{aligned} \quad (3.10)$$

First we perform a functional change of variables from the variables $x(t), x'(t)$ to $X(t) = (x'(t) + x(t))/2$, $y(t) = x'(t) - x(t)$. We also perform a regular change of variables from x_i, x'_i to $X_i = (x'_i - x_i)/2$, $y_i = x'_i - x_i$. The Jacobian determinant for both change of variables is one. Then we use eqs.(3.8,3.9) and define $L = L_0 - \mu$

to obtain:

$$\begin{aligned} \mathcal{W}_S(X_f, P_f, t_f) &= \int dX_i \int dP_i \mathcal{W}_S(X_i, P_i, t_i) \frac{1}{2\pi\hbar} \int dy_f e^{\frac{i}{\hbar} P_f y_f} \int dy_i e^{-\frac{i}{\hbar} P_i y_i} \\ &\quad \times \int_{y(t_i)=y_i}^{y(t_f)=y_f} Dy \int_{X(t_i)=X_i}^{X(t_f)=X_f} DX e^{\frac{i}{\hbar} (-M\dot{X}(t_f)y_f + M\dot{X}(t_i)y_i + y \cdot (L \cdot X - f) - \frac{1}{2} y \cdot \nu \cdot y)}. \end{aligned} \quad (3.11)$$

The functional integral over y is Gaussian and can be evaluated formally to give:

$$\int_{y(t_i)=y_i}^{y(t_f)=y_f} Dy e^{\frac{i}{\hbar} (y \cdot (L \cdot X - f) - \frac{1}{2} y \cdot \nu \cdot y)} = \sqrt{\frac{1}{\det(\nu/2\pi\hbar)}} e^{-\frac{1}{2\hbar} (L \cdot X - f)^T \cdot \nu^{-1} \cdot (L \cdot X - f)}. \quad (3.12)$$

For the type of noise kernels displayed in Eq. (2.5) the outcome of this functional integral is independent of the endpoints y_i and y_f , irrespective of the distribution of bath frequencies. As a result the integral over y_i and y_f is trivial and gives $(2\pi\hbar)^2 \delta(M\dot{X}(t_i) - p_i) \delta(M\dot{X}(t_f) - p_f)$. We have

$$\begin{aligned} \mathcal{W}_S(X_f, P_f, t_f) &= \frac{2\pi\hbar}{\sqrt{\det(\nu/2\pi\hbar)}} \int dX_i \int dP_i \mathcal{W}_S(X_i, P_i, t_i) \\ &\quad \times \int_{X(t_i)=X_i}^{X(t_f)=X_f} DX (2\pi\hbar)^2 \delta(M\dot{X}(t_i) - P_i) \delta(M\dot{X}(t_f) - P_f) e^{-\frac{1}{2} (L \cdot X - f)^T \cdot \nu^{-1} \cdot (L \cdot X - f)}. \end{aligned} \quad (3.13)$$

Next we do another functional change from $X(t)$ to $\xi(t)$ where

$$X(t) \rightarrow \{X_i = X(t_i), P_i = M\dot{X}(t_i), \xi(t) = (L \cdot X - f)(t)\}. \quad (3.14)$$

For linear change of variables the Jacobian functional determinant is independent of ξ . To ensure that the boundary condition at t_f is satisfied, we need to place a delta function inside the new path integral. The net effect of the functional change of variables is:

$$\int_{X(t_i)=X_i}^{X(t_f)=X_f} DX \delta(M\dot{X}(t_i) - P_i) \rightarrow \int D\xi \delta(X_\xi(t_f) - X_f), \quad (3.15)$$

where $X_\xi(t)$ is the solution of the Langevin equation $(L \cdot X_\xi)(t) = f(t) + \xi(t)$ with the initial conditions (X_i, P_i) . After this functional change we obtain:

$$\begin{aligned}
\mathcal{W}_S(X_f, P_f, t_f) &= \int dX_i \int dP_i \mathcal{W}_S(X_i, P_i, t_i) \\
&\quad \times \int \frac{D\xi}{\sqrt{\det(2\pi\hbar\nu)}} e^{-\frac{1}{2}\xi^T \cdot \nu^{-1} \cdot \xi} \delta(M\dot{X}_\xi(t_f) - P_f) \delta(X_\xi(t_f) - X_f) \\
&\equiv \int dX_i \int dP_i \mathcal{W}_r(X_i, P_i, t_i) \int D\xi P_\xi[\xi] \\
&\quad \times \delta(M\dot{X}_\xi(t_f) - P_f) \delta(X_\xi(t_f) - X_f) \\
&= \left\langle \delta(M\dot{X}_\xi(t_f) - P_f) \delta(X_\xi(t_f) - X_f) \right\rangle_{(X_i, P_i), \xi}. \tag{3.16}
\end{aligned}$$

Here $\xi(t)$ is a random noise with Gaussian statistics and is characterized by its mean and variance:

$$\langle \xi(t) \rangle_\xi = 0. \tag{3.17}$$

$$\langle \xi(t) \xi(t') \rangle_\xi = \nu(t, t'). \tag{3.18}$$

Furthermore since the system and bath are assumed to be uncorrelated initially:

$$\langle X_i \xi(t) \rangle_\xi = \langle p_i \xi(t) \rangle_\xi = 0. \tag{3.19}$$

For the sake of brevity we will drop the subscript ξ on the angular brackets as well as in X_ξ in the rest of this chapter. These properties of the random noise are direct consequences of the fact that the environment is composed of harmonic oscillators and that it is initially in a thermal state. Eq.(3.16) has a clear interpretation. The dynamics of the reduced Wigner function is identical to the dynamics of the phase space density of a stochastic classical system described by the Langevin equation

$(L \cdot X)(t) = f(t) + \xi(t)$. This conclusion is of course the same as the one obtained in Sec. 1.4 using a completely different approach.

As argued in [94] the Langevin equation provides a more detailed description of the dynamics than the master and Fokker-Planck equations, in the sense that the class of quantum correlation functions which may be retrieved from the Langevin equation is larger than the corresponding class for the master or Fokker-Planck equations unless the dynamics is Markovian. Work as defined in Eq.(3.29) is an example of this kind of quantity, since its statistics requires the calculation of multi-time correlations. The non-triviality of multi-time correlations for non-Markovian dynamics has been emphasized in Chapter 2.

It is important to realize that this method gives exact quantum mechanical results at any parameter regime, including arbitrarily low temperatures. The fact that solutions $X(t)$ of a classical Langevin equation are used in eq.(3.16) should not be conjured as having made a semiclassical approximation as was done in e.g. [98].

The effect of environment-induced decoherence at work which validates the notion of a physical trajectory is implicitly contained in this method (depending on the temperature of the bath and its spectral density), not extrinsically introduced by hand. Since these processes are dynamically and self-consistently determined, no semiclassical approximation has been made specifically in the derivation. The real challenge is in the interpretation of the physical variables in light of quantum measurement theory, as we discussed previously. In the following section we discuss under what conditions physical trajectories emerge from the dechis formalism.

3.3.1 Decoherence Functional

We consider histories where the system variable $X(t)$ is specified to follow a trajectory $\chi(t)$ with a given precision $\sigma(t)$, while the environment variables are left completely unspecified. For technical reasons it is convenient to use Gaussian, rather than sharp, window functions $w_\chi[x(\cdot)]$. In the path integral this roughly corresponds to using $\exp \left\{ - \int dt \frac{(x(t) - \chi(t))^2}{2\sigma^2(t)} \right\}$ in eq.(3.3). Furthermore we introduce window functions at every instance of time rather than at discrete time intervals. The set of Gaussian window functions with this property acts as a noise term in the influence action. This can be seen in eq.(3.20) where the noise kernel always occurs in the combination $\nu + (2\sigma^2)^{-1}$. There is some error introduced due to the overlap of projectors defined as above. As a result we will be talking about approximate decoherence. In addressing the diagonal and off-diagonal elements of the decoherence functional it is convenient to define $U = (\chi' + \chi)/2$ and $u = \chi' - \chi$. In [74] it is shown that the decoherence functional for two histories $\chi(t)$ and $\chi'(t)$ defined via these projectors is approximately given by:

$$|\mathcal{D}[U - u/2, U + u/2]| \sim \exp \left\{ -\frac{1}{2}(L \cdot U)^T \cdot (\nu + (2\sigma^2)^{-1})^{-1} \cdot (L \cdot U) - \frac{1}{2}u \cdot (\nu^{-1} + 2\sigma^2)^{-1} \cdot u \right\}. \quad (3.20)$$

Here we again used the compact notation defined in (3.7). The off-diagonal elements vanish as a Gaussian for $u \gtrsim \sqrt{\nu^{-1} + 2\sigma^2}$. Hence an approximately consistent set of histories can be obtained by picking histories that differ by at least this amount. However, if the Langevin noise is weak such that $\nu^{-1} \gg \sigma^2$, we run into trouble.

Because now the decoherence condition requires $u \gtrsim \sqrt{\nu^{-1}} \gg \sigma$. Histories of precision σ in a set do not interfere with each other only if they are separated by a distance much larger than σ . This suggests that we cannot account for all probabilities within such a set.

We conclude that the precision should be adjusted to the noise level by: $\sigma^2 \sim \nu^{-1}$. Then the decoherence condition requires that $u \gtrsim \sigma$. Now we can have a set of histories which decohere approximately and for which the resulting probabilities add up to one. “A picture of the system evolution based on actual nearly-classical trajectories may only result from a compromise whereby the precision of observations is adjusted to the noise level, $\sigma^2 \sim \nu^{-1}$, where σ is the precision at which the trajectories are defined. Larger noise for a given σ means more decoherence but less predictability; for a weaker noise, predictability is only limited by the Heisenberg bounds, but individual trajectories will not decohere.⁷ If we are satisfied with predictability within the limits imposed by the Langevin equation, then in the strong noise limit we may consider individual trajectories as depicting physical reality.”⁸ This condition is ordinarily satisfied at temperatures high enough that the quantum and classical trajectories agree, even for non-Markovian dynamics, as we will see below.

For a given precision σ the higher the temperature, the stronger the noise and the more effectively it brings about decohering histories into trajectories. Thus, even at moderate temperatures and for relatively weak noise, by judicious choices of the

⁷We will continue the exploration of this regime in future work.

⁸Quotation is from [74], p. 89.

coarse graining measure σ , decoherence can be effective enough to warrant the notion of trajectories. It is in this regime that deviations from FTs can be identified using this method. At even lower temperatures no reasonable set of histories decohere and the notion of trajectories lacking, we cannot say the FTs are violated (even though it appears reasonable to doubt their validity) because the contents of FTs may be phrased without invoking trajectories. For completeness of technical presentation, we provide a low temperature expansion in section (3.5.1.2).

3.4 Solutions of the Langevin Equation

We rewrite the Langevin equation $(L \cdot X)(t) = f(t) + \xi(t)$ (also given by (1.33) but this time including the external force $f(t)$ and allowing for arbitrary initial time t_i):

$$M\ddot{X}(t) + 2 \int_{t_i}^t ds \gamma(t-s) \dot{X}(s) + M\Omega^2 X(t) = f(t) - 2\gamma(t-t_i)X(t_i) + \xi(t), \quad (3.21)$$

Note that we have placed the “slip-term” $2\gamma(t-t_i)X_i$ to the right hand side of the equality contrary to what we did in (1.33). A formal solution of the Langevin equation without the external force $f(t)$ has been obtained in Sec. 1.4.2.1. The effect of the external force can be incorporated trivially. But, for technical reasons we will use a slightly different solution in this chapter and the next. The difference is due to the interpretation of the “slip-term” as a driving force as opposed to being part of the homogenous equation, as indicated by being placed to the right of the equality sign in (3.21). Let us call the two linearly independent homogenous (with

respect to (3.21)) solutions $K(t)$ and $G(t)$ such that:

$$K(0) = M\dot{G}(0) = 1; \quad \dot{K}(0) = G(0) = 0. \quad (3.22)$$

The formal solution of the Langevin equation is then:

$$X(t) = X(t_i)K(t-t_i) + P(t_i)G(t-t_i) + \int_{t_i}^t dt' G(t-t') [f(t') - 2X(t_i)\gamma(t' - t_i) + \xi(t)]. \quad (3.23)$$

$K(t)$ and $G(t)$ can be calculated using the Laplace transforms:

$$\hat{K}(z) = \frac{2\hat{\gamma}(z)/M + z}{z^2 + 2z\hat{\gamma}(z)/M + \Omega^2}, \quad \hat{G}(z) = \frac{1/M}{z^2 + 2z\hat{\gamma}(z)/M + \Omega^2}. \quad (3.24)$$

Note that $K(t)$ differs from $\dot{G}(t)M$ (which is the second homogenous solution in Sec. 1.4.2.1) whereas $G(t)$ stays the same since the “slip-term” vanishes for the initial conditions $G(0) = 0$. Eq. (3.24) shows the relation between the two linearly independent homogenous solutions:

$$\begin{aligned} s\hat{K}(s) &= 1 - M\Omega^2\hat{G}(s), & sM\hat{G}(s) &= \hat{K}(s) - 2\hat{\gamma}(s)\hat{G}(s), \\ \dot{K}(t) &= -M\Omega^2G(t), & M\dot{G}(t) &= K(t) - 2 \int_0^t ds \gamma(t-s)G(s). \end{aligned} \quad (3.25)$$

3.4.1 Initial State Preparation

The derivation of classical mechanical FTs for closed systems requires the closed system to be in a thermal state initially. As pointed out earlier our derivation of the Langevin equation (3.21) assumes an uncorrelated initial state in which the bath is in the thermal state of its own Hamiltonian H_E . Such a state is obviously not the thermal state of the combined system and it is not stationary for any choice

of the system's initial state. For this reason the uncorrelated initial state is not appropriate for applications to FTs. This observation is valid even for the classical Brownian motion model and is therefore not due to a quantum mechanical effect.

Assume that the bath oscillator frequencies form a continuum. It is customary to define the spectral density of the bath as

$$J(\omega) \equiv \sum_n \frac{g_n^2}{2m_n\omega_n} \delta(\omega - \omega_n) \quad (3.26)$$

and interpret $J(\omega)$ as a continuous function. Here g_n denotes the coupling constant to the n 'th bath oscillator with mass m_n and frequency ω_n . As long as the spectral density is non-vanishing near the system frequency the resulting Langevin dynamics is truly dissipative, in the sense that $\lim_{t \rightarrow \infty} G(t), \gamma(t) = 0$. Physically, true dissipation corresponds to a positive average heat rate at all times. If the spectrum of bath frequencies is discrete, the resulting damping kernel is oscillatory. This is the case even for an infinite but countable number of discrete frequencies. As a result after some (possibly very long) time, there may be average heat flow from the bath into the system. By true dissipation we mean a definite arrow of time for all times. Under these assumptions it has been shown in Chapter 2 that if the uncorrelated initial state is prepared at the infinite past, for times $t > 0$ the dynamics of the system oscillator is indistinguishable from that of a combined system + bath thermal state preparation. In other words the effect of a thermal initial state can be achieved by allowing the uncorrelated system to equilibrate for an infinite amount of time. At $t=0$ the system density matrix is Gaussian. Means and variances of position and momentum are equal to those of the combined thermal state of QBM

given in [30]:

$$\sigma_{xx} = \frac{1}{M\beta} \sum_{r=-\infty}^{\infty} \frac{1}{\Omega^2 + \nu_r^2 + 2\nu_r\hat{\gamma}(\nu_r)/M} \quad (3.27)$$

$$\sigma_{pp} = \frac{M}{\beta} \sum_{r=-\infty}^{\infty} \frac{\Omega^2 + 2\nu_r\hat{\gamma}(\nu_r)/M}{\Omega^2 + \nu_r^2 + 2\nu_r\hat{\gamma}(\nu_r)/M} \quad (3.28)$$

where $\nu_r = 2\pi r/\hbar\beta$ are the bosonic Matsubara frequencies. These variances differ from those corresponding to a Boltzmann distribution with respect to the system Hamiltonian alone. The differences start at second-order in the coupling strength between the system and the bath. In the literature ignoring these differences is sometimes referred to as the *weak coupling approximation*. The results of this chapter *do not* depend upon the *weak coupling approximation* in this sense.

It is worth emphasizing that the equivalence of ensemble preparations is not just on the level of reduced density matrices, which can give only single-time correlations for general non-Markovian dynamics. FTs require multi-time correlations and equivalence of preparations at this level has been shown by proving the equivalence of corresponding trajectories for $t > 0$. This means that any quantum mechanical correlation function involving only the open system variables and times larger than zero will be identical in both preparations.

As a result the trajectories we obtained in the previous section can be used to describe a thermal state as long as we take $t_i \rightarrow -\infty$ and assume a continuous spectrum for the bath frequencies which does not vanish near the system frequency. The thermal state preparation procedure is as follows: at the infinite past the system and bath are in a product state: the bath is in the thermal state, and the system is in an arbitrary state. The combined system evolves in time under the Hamiltonian

(1.16) with H_S given by (3.4) with $f(t) = f(0)$ for $t < 0$. At $t = 0$ the force protocol is started as usual. Although the formulation of FTs is independent of the value of f after $t = \tau$, it proves convenient to define $f(t) = f(\tau)$ for $t > \tau$.

3.5 Probability Distribution of Work and the FTs

With these conceptual and technical preparations, we now can define work performed on the system in the time interval $[0, \tau]$ in the QBM model using the trajectories given by the solutions of the Langevin equation (3.23) as:

$$W = \int_0^\tau dt \frac{\partial H_C}{\partial t} = - \int_0^\tau dt \dot{f}(t) X(t) = - \int_{-\infty}^\infty dt \dot{f}(t) X(t) \equiv -\dot{f}^T \cdot X, \quad (3.29)$$

where the superscript T stands for transpose. In the last equality we utilized the notation of Eq.(3.7), and the integration limits have been set to plus and minus infinity. This change does not introduce any error since $\dot{f}(t)$ vanishes outside the interval $(0, \tau)$ due to the extended definition given at the end of the previous section. We will adopt this convention about integration limits for the rest of the chapter.

We define the retarded Green's function as $G_{ret}(t - t') = G(t - t')\theta(t - t')$.

Then for positive times:

$$X(t) = [G_{ret} \cdot f](t) + [G_{ret} \cdot \xi](t), \quad (3.30)$$

$$\langle X(t) \rangle = [G_{ret} \cdot f](t), \quad (3.31)$$

$$\sigma_{xx}(t, t') \equiv \langle X(t)X(t') \rangle - \langle X(t) \rangle \langle X(t') \rangle = [G_{ret} \cdot \nu \cdot G_{ret}^T](t, t'), \quad (3.32)$$

$$W = -\dot{f}^T \cdot G_{ret} \cdot f - \dot{f}^T \cdot G_{ret} \cdot \xi. \quad (3.33)$$

That $\sigma_{xx}(t, t')$ is a function of $t - t'$ only will be verified explicitly later.

Work defined in Eq.(3.33) is linear in $\xi(t)$ and $\xi(t)$ is a Gaussian random process. Thus, W itself is a Gaussian random variable. As a result the first two moments of W specify its entire statistics given by:

$$P(W) = \frac{1}{\sqrt{2\pi\sigma_W^2}} e^{-(W-\langle W \rangle)^2/2\sigma_W^2}. \quad (3.34)$$

The mean of work is given by:

$$\langle W \rangle = -\dot{f}^T \cdot G_{ret} \cdot f. \quad (3.35)$$

Integrating this by parts and defining $\Delta F = -(f(\tau)^2 - f(0)^2)/2M\Omega^2$ we get:

$$\langle W \rangle = \Delta F + \frac{\dot{f}^T \cdot K_e \cdot \dot{f}}{2M\Omega^2}, \quad (3.36)$$

where we have defined $K_e(t, t') \equiv K(|t-t'|)$ and used the symmetry of the integrand.

The standard deviation of work is calculated as:

$$\sigma_W^2 = \langle W^2 \rangle - \langle W \rangle^2 = \dot{f}^T \cdot \sigma_{xx} \cdot \dot{f}. \quad (3.37)$$

The Jarzynski equality states that:

$$\langle e^{-\beta W} \rangle = \int dW \mathcal{P}(W) e^{-\beta W} = e^{-\beta(\langle W \rangle - \beta\sigma_W^2/2)} = e^{-\beta\Delta F_C}, \quad (3.38)$$

where ΔF_C is the difference in free energy of the combined system for two different values of the external force f calculated quantum mechanically. Due to the linearity of the QBM model ΔF_C has the same form as ΔF defined earlier, which is the classical result. Note that this is only true for the difference of the free energies, since the quantum and classical free energies themselves are different even for the simple harmonic oscillator. The quantum mechanical free energy in the case of $f = 0$

is given by:

$$F_C = F_E - \frac{1}{\beta} \log \left(\frac{1}{\beta \hbar \Omega} \prod_{r=-\infty}^{\infty} \frac{\nu_r^2}{\Omega^2 + \nu_r^2 + 2\nu_r \hat{\gamma}(\nu_r)/M} \right) \quad (3.39)$$

where F_E is the free energy of the isolated bath. The corresponding free energy in the classical model is simply the sum of the free energies of the isolated system and bath. The equality of the difference of free energies due to a driving force in classical and quantum mechanics can be understood easily by noting that the main effect of the linear driving force is to shift the energy levels.

As a consequence Jarzynski equality is satisfied if and only if:

$$\dot{f} \cdot \sigma_{xx} \cdot \dot{f} = \frac{\dot{f} \cdot K_e \cdot \dot{f}}{\beta M \Omega^2}. \quad (3.40)$$

Note that this equality should hold for any $\dot{f}(t)$. This condition can be stated mathematically as

$$\frac{\delta}{\delta \dot{f}(s)} \int_0^{t_f} dt \int_0^{t_f} dt' \dot{f}(t) \left[\frac{K(|t-t'|)}{\beta M \Omega^2} - \sigma_{xx}(t-t') \right] \dot{f}(t') = 0, \quad (3.41)$$

$$\int_0^{t_f} dt \dot{f}(t) \left[\frac{K(|t-s|)}{\beta M \Omega^2} - \sigma_{xx}(t-s) \right] = 0. \quad (3.42)$$

This equation should also be valid for any $\dot{f}(t)$. Differentiating one more time with respect to $\dot{f}(s')$ we get the condition:

$$\sigma_{xx}(s-s') = \frac{K(|s-s'|)}{\beta M \Omega^2}. \quad (3.43)$$

For Crooks's fluctuation theorem we need to consider the reverse process which corresponds to a reversed force protocol and an initial state with the force value $f(\tau)$. We will use a subscript R for the quantities associated with the reversed process

and no subscript for forward process.

$$f_R(t) = f(\tau - t); \quad \Delta F_R = -\Delta F. \quad (3.44)$$

The corresponding work distribution is again specified by its first two moments, which can be shown to be:

$$\langle W \rangle_R = \langle W \rangle - 2\Delta F, \quad (3.45)$$

$$(\sigma_W^2)_R = \sigma_W^2.$$

Note that the standard deviation of work is the same for the forward and reverse protocols. The probability distribution of work in the reversed process is given by:

$$P_R(W) = \frac{1}{\sqrt{2\pi(\sigma_W^2)_R}} e^{-(W - \langle W \rangle_R)^2 / 2(\sigma_W^2)_R} = \frac{1}{\sqrt{2\pi\sigma_W^2}} e^{-(W - (\langle W \rangle - 2\Delta F))^2 / 2\sigma_W^2}. \quad (3.46)$$

Consider the ratio:

$$\frac{P_F(W)}{P_R(-W)} = e^{\frac{(\langle W \rangle - \Delta F)}{\beta\sigma_W^2/2} \beta(W - \Delta F)}. \quad (3.47)$$

Crooks's fluctuation theorem is satisfied if

$$\langle W \rangle - \Delta F = \beta\sigma_W^2/2. \quad (3.48)$$

This condition is equivalent to the condition (3.43) for the validity of Jarzynski equality. In general these two conditions need not be the same since Crooks's fluctuation theorem is more general than Jarzynski equality. However, this is the case for Gaussian distributed work.

Let us now try to understand the nature and meaning of condition (3.43). H and G are solutions to the homogenous Langevin equation. As such they do depend on the damping kernel but not on the noise kernel. σ_{xx} on the other hand

depends on both the damping kernel via G and on the noise kernel. For this equality to hold there has to be a relation between the noise and dissipation kernels. The same conclusion can be reached by studying Eq.(3.48). The average of work is independent of the noise kernel, but depends on the damping kernel. On the other hand the standard deviation of work does depend on both kernels.

There is indeed such a relationship between the damping and noise kernels for a thermal bath: the fluctuation dissipation relation (FDR) (2.6). However, the quantum mechanical FDR does not lead to condition (3.43), and thus the FTs do not need to hold. To see this note that the damping kernel is independent of \hbar . As a result the homogenous solutions of the Langevin equation, $K(t)$ and $G(t)$, do not depend on \hbar . On the other hand σ_{xx} in general is a function of arbitrarily large powers of \hbar via the coth term in the noise kernel. FTs are satisfied if \hbar is set to zero. Corrections to FTs is expected at $O(\hbar^2)$.

3.5.1 High and Low Temperature Regimes

As described in the previous subsection, the noise kernel is the only place where quantum effects are manifest, as can be seen by the appearance of \hbar . Assumptions made about the properties of the bath render the quantum features associated with the initial state of the system oscillator forgotten completely. In FTs the noise kernel appears only in the standard deviation of work σ_W^2 . In this subsection we will investigate this term in the high and low temperature regimes.

Using the Fourier transform one can show from Eq.(3.37) that:

$$\sigma_W^2 = (2\pi)^2 \int_{-\infty}^{\infty} d\omega \tilde{f}_d(\omega) \tilde{\sigma}_{xx}(\omega) \tilde{f}_d(-\omega), \quad (3.49)$$

where $\tilde{f}_d(\omega)$ denotes the Fourier transform of $\dot{f}(t)$. Recall that in our convention $\dot{f}(t)$ vanishes outside the interval $[0, \tau]$, thus the Fourier transform is well-defined.

Using the FDR (2.6) it can be shown that:

$$\tilde{\sigma}_{xx}(\omega) = \hbar\omega \coth\left(\frac{\beta\hbar\omega}{2}\right) \frac{\tilde{K}_e(\omega)}{2M\Omega^2}. \quad (3.50)$$

3.5.1.1 High temperature expansion

For frequencies satisfying $\beta\hbar\omega < 1$, \coth can be expanded into a Laurent series:

$$\coth\left(\frac{\beta\hbar\omega}{2}\right) = \frac{2}{\beta\hbar\omega} + \sum_{k=1}^{\infty} \frac{2^{2k} B_{2k}}{(2k)!} \left(\frac{\beta\hbar\omega}{2}\right)^{2k+1}, \quad (3.51)$$

$$\tilde{\sigma}_{xx}(\omega) = \frac{\tilde{K}_e(\omega)}{\beta M \Omega^2} + \sum_{k=1}^{\infty} \frac{2^{2k} B_{2k}}{(2k)!} \left(\frac{\beta\hbar\omega}{2}\right)^{2k+2} \frac{\tilde{K}_e(\omega)}{\beta M \Omega^2}, \quad (3.52)$$

where B_n is the n 'th Bernoulli number. If we assume that either $\tilde{K}_e(\omega)$ or $\tilde{f}_d(\omega)$ decreases sufficiently fast for large frequencies such that $\beta\hbar\omega \geq 1$, the Laurent series is a good expansion. Hence, the characterization of “high” temperature depends on two time scales: the intrinsic time scale of the oscillator (determined by its interaction with the bath as well as its natural frequency) and the time scale of the driving force. It is reasonable to assume that $\tilde{K}_e(\omega)$ vanishes for frequencies larger than the bath cutoff. Usually this is taken to be very large. We will assume that $\tilde{f}_d(\omega)$ becomes negligible at frequencies much smaller than this cutoff frequency, denoted as ω_h . This is expected to be a reasonable assumption for typical driving forces. Then, the high temperature expansion is strictly valid for $\beta\hbar\omega_h \ll 1$. However, this

condition is too conservative since not all environmental modes have equal influence on the system dynamics. The high temperature expansion is good whenever the temperature is high with respect to the most relevant environmental modes, which might be significantly smaller than ω_h . The relevant frequency range of the environment can be read directly from the spectral density (3.26).

If we keep only the first term in the expansion (3.52) we see that condition (3.40) for the validity of FTs is satisfied. Deviations from FTs to all orders of \hbar can be calculated to be:

$$\frac{1}{\beta M \Omega^2} \sum_{n=1}^{\infty} \frac{2^{2n} B_{2n}}{(2n)!} \left(\frac{\beta \hbar}{2} \right)^{2n+2} \dot{f} \cdot K_e^{(2n+2)} \cdot \dot{f}. \quad (3.53)$$

The superscript on K_e denotes the order of derivatives taken with respect to its argument. The correction term can also be written as:

$$\frac{1}{\beta M \Omega^2} \sum_{n=1}^{\infty} \frac{2^{2n} B_{2n}}{(2n)!} \left(\frac{\beta \hbar}{2} \right)^{2n+2} f^{(n+2)} \cdot K_e \cdot f^{(n+2)}. \quad (3.54)$$

Note that the knowledge of the homogenous solution to the Langevin equation, a purely classical object, is enough to calculate the correction term to all orders of \hbar .

3.5.1.2 Low temperature expansion

Below we present the form of the standard deviation of work in a low temperature expansion but we won't go into the details of the low temperature expansion because the notion of trajectories will ultimately break down at sufficiently low temperatures. For high frequencies the following expansion of \coth is more suitable than

Eq.(3.51):

$$\coth\left(\frac{\beta\hbar\omega}{2}\right) = \text{sgn}(\omega) \left[1 + 2 \sum_{k=1}^{\infty} e^{-k\beta\hbar|\omega|} \right], \quad (3.55)$$

$$\tilde{\sigma}_{\text{xx}}(\omega) = \frac{\hbar}{2M\Omega^2} |\omega| \tilde{K}_e(\omega) \left[1 + 2 \sum_{k=1}^{\infty} e^{-k\beta\hbar|\omega|} \right]. \quad (3.56)$$

This expansion is convergent for all frequencies. However convergence is fastest for $\beta\hbar\omega \gg 1$. If we assume that either $\tilde{K}_e(\omega)$ or $\tilde{f}_d(\omega)$ decreases sufficiently fast for $\omega \rightarrow 0$ such that $\beta\hbar\omega \leq 1$, expansion (3.56) is a good one to use for Eq.(3.49). Hence the characterization of low temperature depends on two time scales: the intrinsic time scale of the oscillator and the time scale of the driving force. It is reasonable to assume that $\tilde{K}_e(\omega)$ vanishes for frequencies lower than the lowest bath frequency. Usually this is taken to be very small. We will assume that $\tilde{f}_d(\omega)$ becomes negligible at frequencies much higher than the lowest bath frequency.⁹ Let us denote this frequency by ω_l . Then, the low temperature expansion is strictly valid for $\beta\hbar\omega_l \gg 1$. However, this condition is too conservative since not all environmental modes have equal influence on the system dynamics. The low temperature expansion is good whenever the temperature is low with respect to the most relevant environmental modes, which might be significantly larger than ω_l . The relevant frequency range of the environment can be read directly from the spectral density (3.26).

⁹This condition could be violated by a very slowly changing driving force. However, this case is not interesting for the study of FTs since the system would be evolving adiabatically and the work distribution would be sharply peaked around $W = \Delta F$.

3.5.1.3 High temperature conditions and Markovian Dynamics

An important special case is the Ohmic bath characterized by the spectral density:

$$J(\omega) = \frac{2\gamma_0}{\pi}\omega. \quad (3.57)$$

Without a high frequency cutoff, the damping kernel becomes local in time. The Langevin equation at late times takes on the form:

$$M\ddot{X}(t) + 2\gamma_o\dot{X}(t) + M\Omega^2 X(t) = f(t) + \xi(t). \quad (3.58)$$

Physically one would like to have a high frequency cutoff, which in turn makes the damping kernel nonlocal in time. The high frequency cutoff also cures the pathologies of the noise kernel that occur in the Ohmic case without cutoff. A large cutoff Λ ensures that the damping kernel is strongly peaked around zero. If the driving force $f(t)$ doesn't change significantly on time scales of order $1/\Lambda$, the Markovian approximation can be justified.

However, Markovian dynamics is not the criterion for FTs to be satisfied, high temperature is. This is because even at high temperature if the bath is non-Ohmic the dynamics of the open system can be non-Markovian.

3.6 Relation between Classical and Quantum FTs

In section 1.4 we have established that the dynamics of the QBM model is in fact classical. One may wonder if FTs are satisfied in classical dynamics, with

the above observation, what is it then that causes the possible violation of QFTs at low temperatures? Although the dynamics is the same for classical and quantum models, initial conditions are not. The thermal state, as characterized by the phase space density and the Wigner function, respectively, is different at low temperatures. The damping kernel does not depend on the initial conditions and thus is the same for both quantum and classical models. The noise kernel on the other hand depends on the initial state of the bath. As a result it is the noise kernel that is different and could give rise to deviations from FTs.

In the previous section we have seen how the classical limit is reached at high temperatures. We identified high temperatures as the ones such that all the relevant bath modes are multiply occupied. As is well known from elementary quantum mechanics, multiply-occupied harmonic oscillators act classically. In this classical limit FTs are satisfied.

Alternatively one can solve the classical version of the QBM model exactly, which is possible due to the linearity of the model. Moreover, in the classical model one can use the thermal state of the combined system (also allowing for a finite environment) instead of resorting to the infinite time preparation¹⁰. The result is a Langevin equation in which the noise is correlated with the initial conditions of the system oscillator. One can define a new noise by $\xi(t) - 2X(t_i)\gamma(t - t_i) \rightarrow \xi(t)$ which is uncorrelated with the initial conditions of the system oscillator and obeys the standard FDR. This redefinition also gets rid of the slip term [30, 129] in the

¹⁰The equivalence of both preparations for the classical model follows directly from the $\hbar \rightarrow 0$ limit of the quantum result obtained in Chapter 2. This point is also made in Appendix A.

Langevin equation and one obtains the familiar form:

$$M\ddot{X}(t) + 2 \int_0^t ds \gamma(t-s) \dot{X}(s) + M\Omega^2 X(t) = f(t) + \xi(t), \quad (3.59)$$

$$\langle \xi(t) \rangle_{\text{cl}} = 0, \quad (3.60)$$

$$\langle \xi(t) \xi(t') \rangle_{\text{cl}} = \frac{2}{\beta} \gamma(t-t'), \quad (3.61)$$

$$\langle X_i \xi(t) \rangle_{\text{cl}} = \langle P_i \xi(t) \rangle_{\text{cl}} = 0, \quad (3.62)$$

where the initial conditions of the open system are sampled from the reduced phase space density of the system that is obtained from the thermal phase space density of the combined system by integrating out the bath degrees of freedom. This reduced phase space density *happens to be* the thermal state with respect to the system Hamiltonian, as seen below in Eq. (3.63).

Eqs.(3.59-3.62) are the beginning point of the analysis of Mai and Dhar [130]. They start with the phenomenological Langevin equation that is identical to (3.59). They further assume a Gaussian noise with the classical FDR (3.61). Finally they assume that the initial values of the system oscillator coordinates are sampled from the classical phase space density $f_S(X_i, P_i, t_i) \propto \exp[-\beta H_S(X_i, P_i, t_i)]$. This last point can be justified from the microphysics model:

$$\begin{aligned} f_S(X_i, P_i, t_i) &= \prod_{n=1}^N \int dx_{ni} \int dp_{ni} f_C(X_i, P_i; \{x_{ni}\}, \{p_{ni}\}; t_i) \\ &\propto \exp[-\beta H_S] \prod_{n=1}^N \int dx_{ni} \int dp_{ni} e^{-\beta \sum_{n=1}^N \left[\frac{p_{ni}^2}{2m_n} + \frac{1}{2} m_n \omega_n^2 \left(x_{ni} - \frac{g_n}{m_n \omega_n^2} X_i \right)^2 \right]} \\ &\propto \exp[-\beta H_S(X_i, P_i, t_i)]. \end{aligned} \quad (3.63)$$

Similarly the change in free energy that appears in Jarzynski equality and Crooks's

fluctuation theorem is that of the combined system. However, the structure of the coupling and renormalization terms make it coincide with that of the isolated system oscillator. This clever scheme notwithstanding, we point out that in their phenomenological approach [130] the free energy difference is mistakenly interpreted as that of the free oscillator, since there is not enough information to track down its origin. Similarly initial conditions are sampled from the system thermal state with uncorrelated noise. Our microscopic model shows that none of these assumptions are individually justified, yet their combination somehow conspires to validate the fluctuation theorems. This kind of ambiguity and disconnectedness often found in the phenomenological models in the literature heightens the importance and advantage of using a first-principles approach based on micro-physics models, as is adopted here.

Starting from a microscopic model we were able to recover all the features of the phenomenological Langevin equation. From there on, using the same analysis as in [130] leads to the verification of FTs. However, it is crucial to make the following distinction: In the phenomenological theory there is no a priori reason why FTs should hold because the open system dynamics is not Hamiltonian. As a result one needs to show the validity of FTs explicitly. In our formalism, on the other hand, we start with a closed (system + bath) Hamiltonian system in a thermal state (of the combined system). Hence all the premises of the FTs are satisfied and one expects that they should hold. What needs to be done is to verify them from explicit calculations.

One might object to this claim by noting that an uncountably infinite bath is

required for the preparation described in section (3.4.1). The proof of FTs for close Hamiltonian systems utilizes the Liouville theorem, for which we have seen only proofs for finite number of degrees of freedom. In this sense our model, with infinite preparation time also doesn't trivially satisfy FTs, and needs the explicit verification. On the other hand for finite baths one can use the thermal state of the combined system at $t=0$ and then the FTs follow trivially. This second procedure is very easy for the classical model though somewhat complicated yet still straightforward for the quantum model. For the quantum model one would use the correlations of initial condition with noise and the noise kernel derived for Chapter 2 in Appendix A. The important point is that the infinite time preparation is only introduced for technical convenience. It can be argued that for any relevant times $t > 0$ the effect of an infinite bath can be approximated arbitrarily closely by a large but finite bath. Hence our results are insensitive to the unphysical assumptions about the bath we made in our derivation.

It is worth mentioning that Speck and Seifert [131] have shown that the Jarzynski relation holds for general classical ergodic systems governed by stochastic dynamics including non-Markovian processes. Ohkuma and Ohta [132] studied classical systems described by a non-linear, non-Markovian Langevin equation with Gaussian colored noise. Both of these works are more general than our work when applied to classical systems because they are not restricted to linear models. On the other hand both adopt a phenomenological approach without an underlying microscopic model, as we do.

3.7 Discussion

3.7.1 Comparison to previous work

As mentioned in sections 1.5.2 and 3.1 there seems to be a consensus on how to define work in closed quantum systems [63]. Work is defined as the difference of the energy of the closed system measured at two different times. This method is less attractive when applied to open systems (treating the system+environment as the closed system) since it involves measuring the energy of the combined system. Furthermore, work is restricted to the open system, and it is only a part of the total energy which involves also heat exchange with the bath. This can lead to big errors if one calculates the work of the combined system since work is the difference of two large numbers.

In this chapter we used the decoherent history conceptual framework to explain how the notion of trajectories in a quantum system can be made viable and use them to define work for open quantum systems. These quantities are likely to be more easily accessible than the energy levels for practical purposes related to experiments, especially for open quantum systems. The classical mechanical definition of work in terms of trajectories is used in the formulation of FTs.

The work operator is another route taken [62] but there is no satisfactory definition of work as an operator [97]. Besides, the work operator approach does not place any limit on the range of validity of its predictions. Using the environment-induced decoherence scheme we can assess how strong the noise in the environment

needs to be to provide sufficient decoherence to warrant the use of trajectories so as to be able to define work in open quantum systems. The question of whether deviations from FTs can be observed in low temperature experiments at all, and if so in which parameter range, requires more quantitative analysis. We intend to address this question in the future.

Compared with previous work in the literature the approach of Chernyak and Mukamel [98] is closest to ours in spirit. However, in substance our approach differs from theirs in several important ways, as numerated below. Foremost a theoretical justification of the use of and the derivation of the range of validity of the trajectory concept in quantum mechanics are necessary in the formulation of FTs. To this end the authors of [98] invoke continuous measurements and wave function collapse together with taking the semi-classical limit. We point out the key conceptual and procedural steps which we believe [98] are flawed.

Conceptual flaws.

It is said in [98] that “the classical limit can be reproduced by using the Wigner function”. Also, “ Q_+ (Our X) is a classical coordinate variable and Q_- (our y) is a quantum coordinate”

These wrong statements stem from, we believe, a lack of understanding of the central issues in quantum decoherence. Misconceptions like these were common but were addressed and clarified in the 90s. See e.g., [48, 110].

Quantitative differences.

The range of validity is not stated clearly in [98] and the generating functional of work given in their Eq.(9) is said to be valid at arbitrary temperature. We believe

this is an overclaim.

In the *dechis* or *envdec* formalism trajectories emerge due to the influence of the environment, in particular, the strength of noise: The stronger the noise, the more pronounced trajectories take shape; the weaker the noise, the more quantum features prevail. These conditions of classicality can be quantified clearly and from them one obtains the criteria for determining the range of validity of quantum FTs as we discussed in an earlier section.

Eq.(13) of [98] gives the lowest order in \hbar correction to the Jarzynski equality. We provide the corrections to arbitrary orders of \hbar in our Eq.(3.54) in terms of the homogenous solutions to the Langevin equation. Furthermore we show that these corrections apply to Crooks's fluctuation theorem as well. At the classical level we derive Crooks's fluctuation theorem and Jarzynski's equality for the Brownian motion model.

3.7.2 New issues brought forth

The *dechis* and *envdec* approach bring forth a number of new issues which were not so clearly noted before. We name three here.

Initial state preparation.

Initial state preparation is an important aspect of FTs. Most of the literature on FTs for closed systems is usually clear on this aspect. However a certain level of ambiguity exists in open system treatments. In this work we considered an initial thermal state for the closed system made up of the system of interest plus its envi-

ronment. However, for computational ease and clarity of exposition we developed an equivalent initial state preparation method based on product initial states for the system and the bath. This equivalence is proven in Chapter 2. Our initial state preparation replaces the system's dependence on the initial state by the properties of noise statistics. As a result our preparation method has only one probabilistic element as opposed to two. This makes the analysis clearer and the identification of quantum effects easier.

On the meaning of the average in Eq.(3.1).

The averages that are calculated using the statistics of noise can alternatively be expressed in terms of expectation values of quantum mechanical operators. The important point is that products of position and momentum operators need to be symmetrized owing to the properties of Wigner function, which is used in the averaging process. In the specific case of the Jarzynski equality, we observe that the average over noise realizations can also be obtained by taking the expectation value of the quantum mechanical operators as:

$$\begin{aligned}\langle e^{-\beta W} \rangle &= \int D\xi P_\xi[\xi] \int dX_i dP_i W_S(X_i, P_i, 0) e^{\beta \int_0^\tau dt \dot{f}(t) X_\xi(t)} \\ &= \text{Tr}_{S+E} \left[e^{\beta \int_0^\tau dt \dot{f}(t) \mathbf{X}_H(t)} \hat{\rho}_\beta \right],\end{aligned}\tag{3.64}$$

where the subscript H indicates the Heisenberg picture. In this special case symmetrization is achieved by the exponential function together with the fact that the dynamics is linear and work itself is a linear function of position. Consequently we don't need to impose the symmetrization procedure explicitly. It is in this strict sense that the results that are obtained using the work operator

$\mathbf{W} \equiv -\int_0^\tau dt \dot{f}(t) \mathbf{X}_H(t) = \mathbf{H}_H(\tau) - \mathbf{H}_H(0)$ for Jarzynski equality agree with our results obtained via trajectories.

How to decide if possible violations to FTs can be observed?

The formulation of FTs involves averages over noise realizations, with idealized situations where trajectories are perfectly well resolved for each realization of noise. But of course in an experiment, even classically, there is only finite resolution. Let us assume that the resolution of the experiment is independent of temperature. This introduces an error to the FTs obtained from this data that is independent of the temperature.

In the quantum case the condition $\sigma^2 \sim \nu^{-1}$ suggests that for stronger noise we can resolve the trajectory to a higher precision. As the noise weakens, such as at decreasing temperature, the stochastic features of classical trajectories are enhanced due to the decoherence condition and measurement results on a particle's trajectory becomes less precise. Further weakening the noise we will get to a point in which quantum or “Heisenberg” noise dominates [121]. Here lies a fundamental difference between classical and quantum. In quantum mechanics the ability of resolving trajectories is not only determined by the precision of the measurement device but also by the temperature. As a result the error, or inaccuracy, in FTs introduced by the resolution of trajectories increases constantly as the temperature is lowered, unlike in classical mechanics. Below a certain temperature, upon entering the quantum dominated regime, the imprecision in measurements will become too large to render any free energy calculations using FTs meaningless.

The properties of noise acting on the quantum Brownian particle are differ-

ent from the noise in the corresponding classical model, as was shown above. This introduces a deviation from FTs which is independent of the error introduced by the limited precision of measurements (discussed in the previous paragraph). The quantum corrections to the noise kernel become larger at lower temperatures. As a result we expect to observe deviations from FTs at low temperatures. However, the precision of trajectories diminish with temperature which introduces increasing errors to the calculation of averages. Given these competing effects, it is not clear if there is a parameter range where quantum corrections to the noise kernel are large enough to cause deviations from FTs observable beyond the error, or inaccuracy, introduced by measurement imprecision imposed by the decoherence condition. Further quantitative analysis is necessary to establish the domain of validity of our approach and the magnitude of possible violations to FTs within this domain as a function of temperature.

Chapter 4: Microcanonical work and fluctuation relations for an open system: an exactly solvable model

4.1 Introduction

In the previous chapter we have studied quantum fluctuation theorems. Now we turn our attention to classical fluctuation theorems, in particular the nonequilibrium work relation due to Jarzynski [11, 12],

$$\langle e^{-\beta W} \rangle = e^{-\beta \Delta F}, \quad (4.1)$$

and the closely related fluctuation theorem, due to Crooks [13–15],

$$\frac{P(W)}{\tilde{P}(-W)} = e^{\beta(W - \Delta F)}, \quad (4.2)$$

both of which were already introduced in Sec. 1.5.1.

Ref. [11] presents Hamiltonian derivations of Eq. (4.1) for two different scenarios. In the first, a system of interest is prepared in equilibrium by being placed in weak contact with a thermal reservoir, which is then removed. In this case it is natural to treat the initial conditions of the system of interest as a random sample from the canonical distribution (reflecting the method of preparation), and also to use Hamiltonian dynamics to model the subsequent evolution of the thermally isolated system as the control parameter is varied ($0 \leq t \leq \tau$).

In the second scenario considered in Ref. [11], the system remains in weak thermal contact with the reservoir throughout the process. In this derivation, initial conditions for the combined system *and reservoir* were assumed to be sampled from a canonical distribution, and then Hamilton’s equations were used to model evolution in the full phase space. In Ref. [133] this approach was extended to a system in strong thermal contact with a reservoir, again assuming canonically sampled initial conditions in the full phase space.

In both derivations described in the previous paragraph, the use of Hamilton’s equations to model the dynamics in the full phase space implies that the combined system of interest and reservoir are being treated as a large, thermally isolated system. The assumption of a canonical distribution of initial conditions for this combined system renders the derivation of Eq. (4.1) (as well as Eq. (4.2)) straightforward. However, from a conceptual perspective this assumption is somewhat problematic, as the equilibrium state of an isolated system is typically associated with the *microcanonical* ensemble. It is therefore natural to wonder whether Eqs. (4.1) and (4.2) remain valid when initial conditions are sampled microcanonically rather than canonically. In this chapter we will address this question through the exact analysis of the classical Brownian motion model 1.4, involving a harmonic oscillator (the system of interest) coupled strongly to a bath of N_E other harmonic oscillators (the thermal reservoir). This model has previously been studied by Hasegawa [134], who considered initial conditions sampled from the canonical ensemble. More generally, the study of model systems for which exact results can be obtained has illustrated and illuminated a variety of issues related to Eqs. (4.1) and (4.2). [135–156]

It is widely believed that in the thermodynamic limit, the average thermodynamic properties of a physical system are independent of the choice of the ensemble. This is the idea of *ensemble equivalence* [1]. However, the situation is quite different when fluctuations are considered [157]. (As a simple example note that the variance of the total energy is proportional to heat capacity in the canonical ensemble, but vanishes identically in the microcanonical ensemble.) This suggests that the validity of Eqs. (4.1) and (4.2), for microcanonically sampled initial conditions, does not follow immediately from the equivalence of ensembles, even when the thermal reservoir is assumed to be macroscopic. This issue is especially relevant since large fluctuations with very small probabilities play a dominant role in the nonequilibrium work theorem [158] whereas standard ensemble equivalence results do not make any claim about or depend on such low probability events. Moreover, the work W is not simply a function of the phase space variables, but rather a functional of the phase space trajectory, and its fluctuations may be more complex than that of typically considered phase space functions.

For a system interacting with a large environment it has been suggested in Ref. [96], using heuristic arguments, that the validity of the nonequilibrium work theorem may be insensitive to the particular distribution used and that the canonical ensemble should be viewed primarily as a computational convenience. A more detailed argument supporting this claim has been developed in Ref. [159]. In Ref. [160] the following microcanonical version of the Crooks fluctuation relation was derived:

$$\frac{P_E(W)}{\tilde{P}_{E+W}(-W)} = \frac{\Sigma_f(E+W)}{\Sigma_i(E)}, \quad (4.3)$$

where $P_E(W)$ stands for the probability density of doing work W during the forward process and $\tilde{P}_{E+W}(-W)$ stands for the probability density of doing work $-W$ during the time reversed process. The subscript indicates the energy of the microcanonical distribution from which the initial conditions are sampled. The right-hand side is the ratio of two densities of states at different energies and associated with initial and final Hamiltonians. (Note that Ref. [160] uses Ω to denote the density of states, which we reserve for the system frequency. Thus we opted to use Σ for the density of states instead). It was then argued in Ref. [160] that in the appropriate thermodynamic limit, one recovers Eq. (4.2). To the best of our knowledge, our work is the first to explore this issue using a model system for which the work distributions can be computed exactly.

The organization of this chapter is as follows. The model is introduced in Sec. 4.2. Exact expressions for the left-hand side of Eq. (4.1) are obtained in Sec. 4.3.1 and for the probability distribution of work in Sec. 4.4.1. The validity of nonequilibrium work relation in the limit of an infinite environment is proven in Sec. 4.3.2. The validity of microcanonical Crooks relation is shown in Sec. 4.4.2. Ensemble equivalence in its most general form is shown in Sec. 4.5.2. Some technical details of the derivation are provided in Appendix B.

4.2 The Model

We consider the classical Brownian motion model with $N_E + N_S \equiv \mathcal{N}$ degrees of freedom. N_S is the number of degrees of freedom of the system of interest and

$N_E \gg N_S$ is the number of degrees of freedom of the environment. The Hamiltonian governing the dynamics of this closed system is of the form (1.20) with the renormalization Hamiltonian set to zero. For explicit calculations we consider the case where the system consists of a single harmonic oscillator, i.e. $N_S = 1$:

$$\begin{aligned}
H_C &= H_S(\mathbf{Z}; \lambda_t) + H_I(\mathbf{Z}, \mathbf{z}) + H_E(\mathbf{z}) \\
H_S(\mathbf{Z}; \lambda_t) &= \frac{P^2}{2M} + \frac{1}{2}M\Omega^2(X - \lambda_t)^2 + \alpha X, \\
H_I(\mathbf{Z}, \mathbf{z}) &= - \sum_{n=1}^N g_n x_n X, \\
H_E(\mathbf{z}) &= \sum_{n=1}^N \left(\frac{p_n^2}{2m_n} + \frac{1}{2}m_n\omega_n^2 x_n^2 \right). \tag{4.4}
\end{aligned}$$

where $\mathbf{Z} = \{X_1, P_1, \dots, X_{N_S}, P_{N_S}\}$, $\mathbf{z} = \{x_1, p_1, \dots, x_N, p_N\}$, λ_t is a time-dependent parameter determined by the protocol, α is a constant and we let $N \equiv N_E$ to simplify the notation. This system Hamiltonian can be realized by the physical system depicted in Fig. 4.1. A mass on a slope is attached to a spring. The support of the spring is moved according to a time-dependent protocol; λ_t denotes the position of the support at time t . To recover the Hamiltonian (4.4) one identifies $Mg \sin \theta \equiv \alpha$. Friction is modelled via linear coupling to N harmonic oscillators that constitute the environment. Generalization to more than one system oscillator and allowing for interactions among environmental oscillators can be achieved by adopting the matrix notation of Chapter 1. However, such a general treatment is not necessary for the purpose of this work.

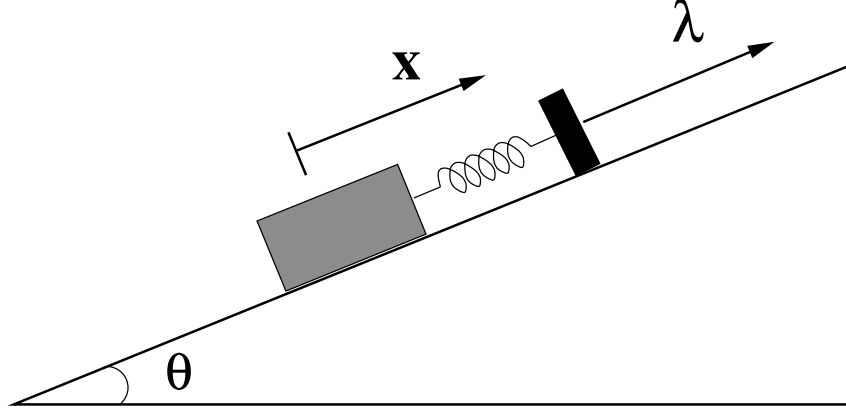


Figure 4.1: A mass on a slope is attached to a spring. The support of the spring is moved according to a time-dependent protocol; λ_t denotes the position of the support at time t . To recover the Hamiltonian (4.4) one identifies $Mg \sin \theta \equiv \alpha$. Friction is modelled via linear coupling to N harmonic oscillators that constitute the environment.

4.2.1 The Solution

It will prove convenient to define

$$f(\lambda_t) \equiv M\Omega^2\lambda_t - \alpha, \quad (4.5)$$

$$\mathcal{J}(\lambda_t) \equiv \frac{1}{2}M\Omega^2\lambda_t^2. \quad (4.6)$$

Then the system Hamiltonian can be written as:

$$H_S(\mathbf{Z}; \lambda_t) = \frac{P^2}{2M} + \frac{1}{2}M\Omega^2 X^2 - f(\lambda_t)X + \mathcal{J}(\lambda_t). \quad (4.7)$$

The corresponding Langevin equation is the same as (3.21) except for the renormalized frequency:

$$M\ddot{X}(t) + 2 \int_0^t ds \gamma(t-s) \dot{X}(s) + M\bar{\Omega}^2 X(t) = f(\lambda_t) - 2\gamma(t)X(0) + \xi(t), \quad (4.8)$$

$$\gamma(t) \equiv \sum_{n=1}^N \frac{g_n^2}{2m_n\omega_n^2} \cos(\omega_n t), \quad (4.9)$$

$$\bar{\Omega}^2 \equiv \Omega^2 - 2\gamma(0), \quad (4.10)$$

$$\xi(t) \equiv \sum_{n=1}^N g_n \left(x_n(0) \cos(\omega_n t) + \frac{p_n(0)}{m_n\omega_n} \sin(\omega_n t) \right). \quad (4.11)$$

The system-environment coupling is required to satisfy $\Omega^2 \geq 2\gamma(0)$ for the dynamics to be stable and we will make this assumption henceforth.

The solution to Eq. (4.8) can be written as

$$X(t) = X(0)K(t) + P(0)G(t) + \int_0^t ds G(t-s) [f(\lambda_s) - 2\gamma(s)X(0) + \xi(s)], \quad (4.12)$$

where $K(t)$ and $G(t)$ are the homogenous solutions of Eq. (4.8) with the right hand side set equal to zero. For details of this solution refer to Sec. 3.4.

4.3 Nonequilibrium Work Relation

We assume a protocol λ_t in the time interval $[0, \tau]$. This corresponds to a function $f(\lambda_t)$ via Eq. (4.5). The work associated with the Hamiltonian (4.7) for the duration of the protocol $\Delta t = \tau$ is given by

$$W = \int_0^\tau dt \dot{\lambda} \frac{\partial H_C}{\partial \lambda} = - \int_0^\tau dt \dot{f}(\lambda_t) X(t) + \Delta \mathcal{J}. \quad (4.13)$$

The dot over a function indicates time derivative, and $\Delta \mathcal{J} \equiv \mathcal{J}(\lambda_\tau) - \mathcal{J}(\lambda_0)$. This definition of work is motivated by the observation $dW = d\lambda \frac{H_C}{d\lambda} = \text{displacement} \times$

force. For a discussion of alternative definitions of work and various fluctuation theorems they lead to see Ref. [61].

For the nonequilibrium work relation (4.1) the initial state is sampled from the *canonical* ensemble at inverse temperature β using the Hamiltonian $H_C(\mathbf{Z}, \mathbf{z}; \lambda_0)$. The free energy difference of the combined system + environment is defined via $\Delta F_C \equiv \Delta F \equiv F_C(\lambda_\tau) - F_C(\lambda_0)$. In our model the free energies can be calculated explicitly.

$$Z_\beta(\lambda) = e^{-\beta F_C(\lambda)} = \int d\mathbf{Z} d\mathbf{z} e^{-\beta H_C(\mathbf{Z}, \mathbf{z}; \lambda)}. \quad (4.14)$$

Here $Z_\beta(\lambda)$ is the partition function associated with the Hamiltonian $H_C(\lambda; \mathbf{Z}, \mathbf{z})$.

The integral over the environmental degrees of freedom gives:

$$\int d\mathbf{z} e^{-\beta(H_I(\mathbf{Z}, \mathbf{z}) + H_E(\mathbf{z}))} \propto e^{\beta X^2 \sum_n \frac{g_n^2}{2m_n \omega_n^2}} = e^{\beta \gamma(0) X^2}. \quad (4.15)$$

Irrelevant constants that will eventually cancel out in the expression for ΔF have been omitted in the above expression. We use the definition (A.3) of the Hamiltonian of mean force as [133, 161]:

$$H^*(\mathbf{Z}; \lambda_t) = H_S(\mathbf{Z}; \lambda_t) - \gamma(0)X^2 = \frac{P^2}{2M} + \frac{1}{2}M\bar{\Omega}^2 X^2 - f(\lambda_t)X + \mathcal{J}(\lambda_t), \quad (4.16)$$

which amounts to shifting the frequency from Ω to $\bar{\Omega}$ in the original system Hamiltonian. Then Eq. (4.14) becomes (up to some irrelevant constants):

$$e^{-\beta F_C(\lambda)} \propto \int d\mathbf{Z} e^{-\beta H^*(\mathbf{Z}; \lambda)} \propto e^{\beta \frac{f(\lambda)^2}{2M\bar{\Omega}^2} - \beta \mathcal{J}(\lambda)}. \quad (4.17)$$

The free-energy difference is not an extensive quantity and is given by

$$\Delta F = -\frac{f(\lambda_\tau)^2 - f(\lambda_0)^2}{2M\bar{\Omega}^2} + \Delta \mathcal{J} \equiv -\mathcal{G} + \Delta \mathcal{J}, \quad (4.18)$$

where

$$\mathcal{G} \equiv \frac{f(\lambda_\tau)^2 - f(\lambda_0)^2}{2M\bar{\Omega}^2}. \quad (4.19)$$

Note that an overall shift in $f(\lambda_0)$ simply changes the equilibrium positions and one is tempted to set $f(\lambda_0) = 0$ in order to simplify the calculation. However, in the analysis of some fluctuation theorems, where both forward and reverse processes are considered, this would cause a loss of generality. Unless $f(\lambda_0) = f(\lambda_\tau)$, or equivalently $\Delta F = 0$, the reverse process is necessarily described with nonzero $f(\tilde{\lambda}_0)$.

In the next section we will consider the quantity:

$$\langle e^{-\bar{\beta}W} \rangle_{\text{mc}} = \frac{\int d\mathbf{Z}d\mathbf{z} \delta(H_C(\mathbf{Z}, \mathbf{z}; \lambda_0) - E) \exp \left[\bar{\beta} \left(\int_0^\tau dt \dot{f}(t) X(t; \mathbf{Z}, \mathbf{z}) - \Delta \mathcal{J} \right) \right]}{\int d\mathbf{Z}d\mathbf{z} \delta(H_C(\mathbf{Z}, \mathbf{z}; \lambda_0) - E)} \quad (4.20)$$

which represents the average of $\exp(-\bar{\beta}W)$, over an ensemble of trajectories with microcanonically sampled initial conditions in the full phase space. We will obtain an exact expression for this average, Eq. (4.22) below, valid for any positive value of the parameter $\bar{\beta}$. We will then show that in the thermodynamic limit, $N \rightarrow \infty$, Eq. 4.1 emerges when the value of $\bar{\beta}$ is set equal to the inverse temperature β_{mc} associated with the microcanonical energy E (see Eq. 4.31). That is:

$$\lim_{N \rightarrow \infty} \langle e^{-\beta_{mc}W} \rangle_{mc} = e^{-\beta_{mc}\Delta F}. \quad (4.21)$$

Although we obtain this result for the case of a single system oscillator, it is easily generalized to any number N_S of system oscillators, provided the limit $N \rightarrow \infty$ is taken with N_S fixed. Moreover, heuristic arguments [159] suggest that this result holds for more general systems with nonlinear interactions. However nonlinear

models are difficult to treat analytically and careful numerical experiments are necessary to test this hypothesis in such models. In this work our aim is to focus on the analytically solvable harmonic oscillator model, for which exact results can be obtained.

4.3.1 Exact Result for finite N

The integrals appearing in the denominator and numerator of Eq. (4.20) have been computed in Appendix B.1. The trick is to use an integral representation of the delta function in order to transform the integrals over the phase space variables into Gaussian integrals. Once the phase space integrals are performed, it is seen that the integration left over from the representation of the delta function can also be performed exactly. Below we cite the results and refer the reader to Appendix B.1 for the technical details.

Combining Eq. (B.15) for the denominator and Eq. (B.31) for the numerator of Eq. (4.20) we obtain for Eq. (4.20):

$$\langle e^{-\bar{\beta}W} \rangle_{\text{mc}} = e^{-\bar{\beta}\Delta\mathcal{J}} e^{\bar{\beta}\mathcal{G}-\bar{\beta}^{-1}\mathcal{D}} \frac{N!}{(\mathcal{A}\mathcal{D})^{N/2}} I_N(\sqrt{4\mathcal{A}\mathcal{D}}). \quad (4.22)$$

where

$$\mathcal{A} \equiv E + \frac{f(\lambda_0)^2}{2M\bar{\Omega}^2} - \mathcal{J}(\lambda_0), \quad (4.23)$$

$$\mathcal{D} \equiv \frac{\bar{\beta}^2}{M\bar{\Omega}^2} I_f, \quad (4.24)$$

$$I_f = \int_0^\tau dt \int_0^t ds \dot{f}(\lambda_t) K(t-s) \dot{f}(\lambda_s). \quad (4.25)$$

Eq. (4.22) is the exact expression for a system of one harmonic oscillator dragged

up a slope in the presence of gravity and coupled to an environment modelled by N harmonic oscillators in a microcanonical ensemble at energy E .

The effect of the environment is implicit in Eq. (4.22). The microcanonical temperature and \mathcal{A} both depend on the total energy E . Also I_f depends on $K(t)$, which is the homogenous solution to the Langevin equation. Finally \mathcal{D} and \mathcal{A} contain factors of $\bar{\Omega}$ which is the renormalized frequency.

4.3.2 The thermodynamic limit, $N \rightarrow \infty$

In this limit we define energy per particle

$$\mathcal{E} \equiv \frac{E}{N+1} = \frac{E}{N} + O(N^{-1}), \quad (4.26)$$

$$\mathcal{A} = N \left(\mathcal{E} + \frac{1}{N} \left(\frac{f(\lambda_0)^2}{2M\bar{\Omega}^2} - \mathcal{J}(\lambda_0) \right) \right) = N\mathcal{E} + O(1). \quad (4.27)$$

Eq. (4.22) becomes:

$$\langle e^{-\beta W} \rangle_{\text{mc}} = e^{-\beta \Delta \mathcal{J}} e^{\beta g - \beta^{-1} \mathcal{D}} \frac{N!}{(\mathcal{E} \mathcal{D} N)^{N/2}} I_N(\sqrt{4N\mathcal{E}\mathcal{D}}). \quad (4.28)$$

The asymptotic behaviour of the Bessel function $I_N(x)$ is usually given for cases where x goes to zero or infinity while N is fixed. In Eq. (4.28) $x \sim \sqrt{N}$ as $N \rightarrow \infty$.

Luckily there is a formula for the limit we are looking for:¹

$$\lim_{N \rightarrow \infty} I_N(\sqrt{Nx}) = \frac{1}{N!} \left(\frac{Nx}{4} \right)^{N/2} e^{x/4}. \quad (4.29)$$

¹To see this, first note that the Bessel function can be written in terms of the generalized hypergeometric functions as $I_N(y) = {}_0F_1(N+1; y^2/4)(y/2)^N/N!$. Looking at the series expansion of the hypergeometric function it is easy to see that ${}_0F_1(N, Ny^2/4) \rightarrow {}_0F_0(y^2/4)$ as $N \rightarrow \infty$. Finally one notes that ${}_0F_0(y) = e^y$ to arrive at the desired formula. We thank Dr. Yury A. Brychkov for the proof of Eq. (4.29) as outlined here.

Using this formula with $x = 4\mathcal{D}\mathcal{E}$ in Eq. (4.28) we obtain:

$$\lim_{N \rightarrow \infty} \langle e^{-\bar{\beta}W} \rangle_{\text{mc}} = e^{-\bar{\beta}(\Delta\mathcal{J}-\mathcal{G})-\bar{\beta}^{-1}\mathcal{D}+\mathcal{E}\mathcal{D}} = e^{-\bar{\beta}\Delta F+(\mathcal{E}-\bar{\beta}^{-1})\mathcal{D}}, \quad (4.30)$$

which, like Eq. (4.22), is valid for arbitrary $\bar{\beta} > 0$.

Since the quantity \mathcal{D} depends on the protocol used to vary the parameter λ (see Eqs. (4.24), (4.25)), the right side of Eq. (4.22) generally cannot be expressed in terms of a difference between two state functions. However, consider the particular choice

$$\bar{\beta} = \beta_{mc} \equiv \mathcal{E}^{-1}, \quad (4.31)$$

corresponding to the inverse temperature given by the equipartition theorem for a collection of one-dimensional harmonic oscillators. For this choice the protocol dependent term vanishes, and – as advertised (Eq. (4.21)) – we recover the nonequilibrium work relation.

For more general models there is still going to be a well-defined relationship between energy per particle and temperature, but it will no longer be linear as in Eq. (4.31). In such models we expect Eq. (4.30) will be protocol independent only for the particular choice of $\bar{\beta} = \beta_{mc}$ which satisfies the corresponding relationship between energy per particle and temperature.

This concludes the derivation of work fluctuation theorem for the classical Brownian motion model in the microcanonical ensemble.

4.4 Crooks's Fluctuation Theorem

4.4.1 Probability Distribution of Work

The moment generating function of work is defined as:

$$G_W(s) = \langle e^{-\imath s W} \rangle_{mc}. \quad (4.32)$$

It can be obtained from Eq. (4.22) by analytic continuation via $\bar{\beta} \rightarrow \imath s$. The probability distribution of work is the Fourier transform of the moment generating function

$$P_E(W) = \frac{1}{2\pi} \int_{\mathcal{C}} ds e^{\imath s W} G_W(s), \quad (4.33)$$

where $P_E(W)$ has been defined earlier in the Introduction.

Assuming $I_f > 0$ and after some manipulations we are lead to the following formula:

$$P_E(W) = \frac{N! 2^N}{2\pi \sqrt{N 2 \sigma_W^2}} \int_{\mathcal{C}} ds \frac{e^{\imath s \frac{(W - \langle W \rangle)}{\sqrt{N 2 \sigma_W^2}}} J_N(s)}{s^N}, \quad (4.34)$$

where $\langle W \rangle \equiv \Delta F + \frac{I_f}{M \bar{\Omega}^2}$ is the expectation value of work and $\sigma_W^2 \equiv 2 \mathcal{A} I_f / N M \bar{\Omega}^2$ is related to the variance of work in the canonical ensemble, as we will see later. The integral can be done analytically to give:

$$P_E(W) = \frac{N!}{\Gamma(N + 1/2) N^{1/2}} \frac{1}{\sqrt{2\pi \sigma_W^2}} \left(1 - \frac{(W - \langle W \rangle)^2 / 2 \sigma_W^2}{N} \right)^{N-1/2} \times \Theta \left(\sqrt{2N} \sigma_W - |W - \langle W \rangle| \right). \quad (4.35)$$

This is the exact expression for the probability distribution of work done on a single harmonic oscillator coupled to an environment of N harmonic oscillators.

The step function in Eq. (4.35) shows that the maximum deviation from the average value of work scales as the square root of N . The fact that the work is bounded is a consequence of the fact that microcanonical ensemble describes a distribution with finite support over the phase space. By applying the method of Lagrange multipliers on the expression of work (4.13), with the constraint of fixed energy, the extreme values of work can be verified independently. This analysis also yields analytical expressions for the phase space trajectory of each particle for the realizations corresponding to extreme values of work.

The special case of $I_f = 0$ is very easy to handle. Using Eq. (B.32) with $\bar{\beta} \rightarrow is$ in Eq. (4.33) we see that the resulting integral is the representation of the delta function. Hence $P_E(W) = \delta(W - \Delta F)$ for $I_f = 0$.

Next we take the limit of infinite environment. The first factor of Eq. (4.35) can easily be seen to converge to one as $N \rightarrow \infty$. For the third factor we use the formula:

$$e^x = \lim_{N \rightarrow \infty} \left[1 + \frac{x}{N} \right]^N. \quad (4.36)$$

Thus for the infinite environment limit we recover the Gaussian form:

$$\lim_{N \rightarrow \infty} P_E(W) = \frac{e^{-\frac{(W - \langle W \rangle)^2}{2\sigma_W^2}}}{\sqrt{2\pi\sigma_W^2}}, \quad (4.37)$$

$$\lim_{N \rightarrow \infty} \sigma_W^2 = 2\mathcal{E} \frac{I_f}{M\Omega^2} = \frac{2}{\beta_{mc}} (\langle W \rangle - \Delta F). \quad (4.38)$$

Eqs. (4.37) and (4.38) ensure that the nonequilibrium work and Crooks's fluctuation theorems are satisfied whenever β is identified with β_{mc} in Eqs. (4.1, 4.2). (Note that (4.38) is the same condition as (3.48)). The probability distribution (4.37) is

identical to the probability distribution of work for the case where the initial conditions of the system plus environment are sampled from a canonical ensemble, with the temperature of the canonical ensemble related to the total energy of the microcanonical ensemble according to $\beta = \beta_{mc}$. This can be easily checked, since all the integrations are Gaussian for the system plus environment canonical initial conditions (as opposed to the presence of the delta function in the microcanonical initial conditions). Sections 3.5 and 3.6 contain some calculations and results relevant to this point.

4.4.2 Microcanonical Crooks Relation

Below we will show the validity of Eq. (4.3) for our specific model and for finite environments. First we note that the initial density of states Σ_i is given by the denominator of Eq. (4.20), and a similar expression applies to the final density of states Σ_f (only with λ_0 replaced by λ_τ). From Eq. (B.15) we have:

$$\frac{\Sigma_f(E + W)}{\Sigma_i(E)} = \left(\frac{\tilde{\mathcal{A}}}{\mathcal{A}} \right)^N, \quad (4.39)$$

where

$$\tilde{\mathcal{A}} \equiv (E + W) + \frac{f(\lambda_\tau)^2}{2M\bar{\Omega}^2} - \frac{1}{2}M\Omega^2\lambda_\tau^2. \quad (4.40)$$

The expressions for $\Sigma_f(E + W)$ and $\tilde{\mathcal{A}}$ for the reverse process have been obtained from Eqs. (B.15) and (4.23) by letting $\lambda_0 \rightarrow \lambda_\tau$ and $E \rightarrow E + W$. The probability

distribution of work in the forward and reverse processes are given by:

$$P_E(W) \propto \frac{1}{\sigma_W^{2N}} \left(\frac{2N\sigma_W^2 - (W - \langle W \rangle)^2}{2N} \right)^{N-\frac{1}{2}} \Theta \left(\sqrt{2N}\sigma_W - |W - \langle W \rangle| \right), \quad (4.41)$$

$$\tilde{P}_{E+W}(-W) \propto \frac{1}{\tilde{\sigma}_W^{2N}} \left(\frac{2N\tilde{\sigma}_W^2 - (-W - \langle \tilde{W} \rangle)^2}{2N} \right)^{N-\frac{1}{2}} \Theta \left(\sqrt{2N}\tilde{\sigma}_W - |-W - \langle \tilde{W} \rangle| \right), \quad (4.42)$$

where $\tilde{\sigma}_W^2 = 2I_f\tilde{\mathcal{A}}/NM\bar{\Omega}^2$ and the following quantities for the time reversed process have been defined in analogy with the forward process:

$$\langle \tilde{W} \rangle \equiv -\Delta F + \frac{I_f}{M\bar{\Omega}^2} = \langle W \rangle - 2\Delta F = \frac{I_f}{M\bar{\Omega}^2} - \Delta F, \quad (4.43)$$

$$\Delta \tilde{F} \equiv -\Delta F = \frac{f(\lambda_\tau)^2 - f(\lambda_0)^2}{2M\bar{\Omega}^2} - \frac{M\bar{\Omega}^2(\lambda_\tau^2 - \lambda_0^2)}{2}, \quad (4.44)$$

$$\tilde{\sigma}_W^2 = \frac{2I_f}{M\bar{\Omega}^2 N} \left(E + W + \frac{f(\lambda_\tau)^2}{2M\bar{\Omega}^2} - \frac{1}{2}M\bar{\Omega}^2\lambda_\tau^2 \right) = \frac{\tilde{\mathcal{A}}}{\mathcal{A}}\sigma_W^2. \quad (4.45)$$

Here we have used the fact that I_f is the same for the forward and reverse process by the virtue of the symmetry of its defining double integral. Based on these formulas we can write the left-hand side of Eq. (4.3) purely in terms of λ and I_f , whereas the right-hand side is simply given by Eq. (4.39). Ignoring the step functions for the moment Eq. (4.3) can be written as

$$\frac{P_E(W)}{\tilde{P}_{E+W}(-W)} = \left(\frac{\tilde{\sigma}_W^2}{\sigma_W^2} \right)^N \left(\frac{2N\sigma_W^2 - (W - \langle W \rangle)^2}{2N\tilde{\sigma}_W^2 - (-W - \langle \tilde{W} \rangle)^2} \right) \quad (4.46)$$

$$= \left(\frac{\tilde{\mathcal{A}}}{\mathcal{A}} \right)^N \left(\frac{2N\sigma_W^2 - (W - \langle W \rangle)^2}{2N\tilde{\sigma}_W^2 - (-W - \langle \tilde{W} \rangle)^2} \right) = \left(\frac{\tilde{\mathcal{A}}}{\mathcal{A}} \right)^N, \quad (4.47)$$

This implies, again disregarding the step function for the moment,

$$2N\sigma_W^2 - (W - \langle W \rangle)^2 = 2N\tilde{\sigma}_W^2 - (-W - \langle \tilde{W} \rangle)^2, \quad (4.48)$$

$$2W \left(\langle W \rangle + \langle \tilde{W} \rangle \right) + \left(\langle \tilde{W} \rangle^2 - \langle W \rangle^2 \right) = 2N \left(\tilde{\sigma}_W^2 - \sigma_W^2 \right), \quad (4.49)$$

This equality can be verified by calculating the following relations.

$$\langle W \rangle + \langle \tilde{W} \rangle = \frac{2I_f}{M\bar{\Omega}^2}, \quad (4.50)$$

$$\langle \tilde{W} \rangle^2 - \langle W \rangle^2 = -\frac{4I_f}{M\bar{\Omega}^2} \Delta F, \quad (4.51)$$

$$\tilde{\sigma}_W^2 - \sigma_W^2 = \frac{2I_f}{M\bar{\Omega}^2 N} (W - \Delta F). \quad (4.52)$$

Now we return to the question of whether the step functions appearing in $P_E(W)$ and $P_{E+W}(-W)$ are identical, so that they cancel when forming the ratio Eq. (4.3). To this end consider the conditions for the probabilities $P_E(W)$ and $\tilde{P}_{E+W}(-W)$ to vanish:

$$2N\sigma_W^2 = (W - \langle W \rangle)^2, \quad (4.53)$$

$$2N\tilde{\sigma}_W^2 = \left(W + \langle \tilde{W} \rangle \right)^2. \quad (4.54)$$

To see that both conditions are identical observe that the difference of both equations gives Eq. (4.49) which has been shown to hold. Thus we have demonstrated the validity of the microcanonical Crooks relation in our particular model and for finite environments.

4.5 Ensemble Equivalence

In most textbooks the term *ensemble equivalence* is used to describe the following property of extensive systems: macroscopic physical quantities assume the

same value in any equilibrium ensemble, i.e. microcanonical, canonical or grand canonical. In this section we will deviate from this definition in three ways. The system plus environment model considered in this chapter is not extensive. Second, the thermodynamic limit is taken with the system size fixed (in the particular case treated here the system consists of a single oscillator). Thus the quantities we consider do not have to be macroscopic. Third, we will consider multi-time averages taken over nonequilibrium processes.

4.5.1 Initial Phase Space Distribution

In this section we show that as $N \rightarrow \infty$ the phase space probability density, $f_S(\mathbf{Z})$, of the system oscillator approaches that of a canonical distribution if the probability distribution for the system plus environment closed system is given by the microcanonical distribution.

The derivation is similar to the previous sections.

$$f_S(\mathbf{Z}) = \frac{\int d\mathbf{z} \delta(H_C(\lambda; \mathbf{Z}, \mathbf{z}) - E)}{\int d\mathbf{Z} d\mathbf{z} \delta(H_C(\lambda; \mathbf{Z}, \mathbf{z}))}. \quad (4.55)$$

For the numerator we again substitute the integral representation of the delta function to obtain:

$$\int_{\mathcal{C}} dz e^{-izE} \int d\mathbf{z} e^{i(H_S(\mathbf{Z}) + H_I(\mathbf{Z}, \mathbf{z}) + H_E(\mathbf{z}))} = \int_{\mathcal{C}} dz \frac{e^{-iz(E - H^*(\lambda; \mathbf{Z}))}}{z^N}. \quad (4.56)$$

Here we used Eq. (B.9). This integral can be obtained using the Cauchy theorem. The integrand has a pole of order N at the origin and the integration contour \mathcal{C} is passing below this pole in the complex plane. For $E > H^*(\lambda; \mathbf{Z})$ the contour

can be closed from above to enclose the pole, and there is a nonzero outcome. For $E < H^*(\lambda; \mathbf{Z})$ the contour is closed from below where the function is analytic. Hence the outcome of the integral is zero. The final expression for the normalized probability density of system degrees of freedom is given by:

$$f_s(\mathbf{Z}) = \frac{N\bar{\Omega}}{2\pi} \frac{(E - H^*(0; \mathbf{Z}))^{N-1}}{\mathcal{A}^N} \Theta(E - H^*(0; \mathbf{Z})). \quad (4.57)$$

where Θ denotes the Heaviside step function. The existence of the step function is a manifestation of the fact that the energy of the system oscillator cannot exceed that of the system plus environment.

Next consider the $N \rightarrow \infty$ limit.

$$\lim_{N \rightarrow \infty} f_s(\mathbf{Z}) = \frac{\bar{\Omega}}{2\pi\mathcal{E}} \lim_{N \rightarrow \infty} \left(1 - \frac{H^*(\mathbf{Z})/\mathcal{E}}{N+1}\right)^{N-1} = \frac{\bar{\Omega}}{2\pi\mathcal{E}} e^{-\mathcal{E}^{-1}H^*(\mathbf{Z})}, \quad (4.58)$$

where we used (4.36) in the last equality. The limit in Eq. (4.58) needs to be interpreted as follows: For any finite N the probability density (4.57) agrees with the canonical distribution (4.58) for small energies. However at large enough energies relative differences become significant. These differences would also show up at high order moments of position and momenta. The limit in Eq. (4.58) means that given an energy interval or equivalently a maximum order for the moments of interest, one can choose a large enough N such that the microcanonical result will agree with the asymptotic result to the desired degree.

Eq. (4.58) describes a Boltzmann state with the Hamiltonian of mean force replacing the system Hamiltonian. Note that the same probability distribution is obtained, albeit for any N , if the system plus environment is sampled from a canonical distribution. In fact this is how the Hamiltonian of mean force is usually

motivated. Eq. (4.58) states that for a large environment the phase space density of the system degrees of freedom is the same if the system plus environment is sampled from a canonical or microcanonical distribution.

4.5.2 Multi-time Correlations

The most general multi-time correlation function during the nonequilibrium process can be obtained from the generating functional

$$Z_{ens}[j(\cdot)] = \langle e^{\int_0^\tau dt j(t)X(t)} \rangle_{ens}, \quad (4.59)$$

where $X(t)$ is the solution to the equations of motion with some initial conditions and the averaging is done over the desired ensemble. Here we will compare the generating functionals for the canonical and microcanonical ensembles. Any multi-time correlation can be obtained from the generating functional by applying differential operators to it, for example:

$$\frac{\delta}{\delta j(t_1)} Z[j(\cdot)] \Big|_{j=0} = \langle X(t_1) \rangle, \quad (4.60)$$

$$M \frac{\partial}{\partial t_1} \frac{\delta}{\delta j(t_1)} Z[j(\cdot)] \Big|_{j=0} = \langle P(t_1) \rangle, \quad (4.61)$$

$$M \frac{\partial}{\partial t_1} \frac{\delta}{\delta j(t_1)} \cdots M \frac{\partial}{\partial t_k} \frac{\delta}{\delta j(t_k)} \frac{\delta}{\delta j(t_{k+1})} \cdots \frac{\delta}{\delta j(t_l)} Z[j(\cdot)] \Big|_{j=0} = \langle P(t_1) \cdots P(t_k) X(t_{k+1}) \cdots X(t_l) \rangle. \quad (4.62)$$

Note that even the average appearing in nonequilibrium work relation Eq. (4.20) can be obtained from this generating functional via

$$\langle e^{-\beta W} \rangle = e^{-\beta \Delta \mathcal{J}} \langle e^{\beta \int_0^\tau dt f(t)X(t)} \rangle = e^{-\beta \Delta \mathcal{J}} Z[\beta \dot{f}(\cdot)]. \quad (4.63)$$

The results presented in this section thus include that of Sec. 4.3.1 as a sub-case.

The calculation of the generating functional in both canonical and microcanonical ensembles is straightforward but tedious. For the canonical ensemble the calculation involves only Gaussian integrals and the use of properties of the solutions of the Langevin equation. The derivation for the microcanonical ensemble mimic closely the treatment presented in Appendix B.1. Here we only provide the final results.

$$\begin{aligned} Z_{can}[j(\cdot)] &= e^{\int_0^\tau dt j(t) \left(\frac{f(\lambda_0)}{M\Omega^2} K(t) + \int_0^t ds G(t-s) f(\lambda_s) \right) + \int_0^\tau dt \int_0^t dt' j(t) \left(\frac{K(|t-t'|)}{\beta M\Omega^2} \right) j(t')} \\ &= \exp \left(\int_0^\tau dt j(t) \langle X(t) \rangle + \int_0^\tau dt \int_0^t dt' j(t) \sigma_{xx}(t, t') j(t') \right), \end{aligned} \quad (4.64)$$

$$Z_{mc}[j(\cdot)] = \exp \left(\int_0^\tau dt j(t) \langle X(t) \rangle \right) \frac{N!}{(\mathcal{A}\bar{\mathcal{D}}[j(\cdot)])^{N/2}} I_N \left(\sqrt{4\mathcal{A}\bar{\mathcal{D}}[j(\cdot)]} \right). \quad (4.65)$$

where $\langle X(t) \rangle$ stands for the average position at time t and $\sigma_{xx}(t, t') \equiv \langle X(t)X(t') \rangle - \langle X(t) \rangle \langle X(t') \rangle$ stands for the two time fluctuations of the position. We also defined $\bar{\mathcal{D}}[j(\cdot)] \equiv \int_0^\tau dt \int_0^t dt' j(t) \frac{K(t-t')}{M\Omega^2} j(t')$ analogous to \mathcal{D} whereby $j(t)$ replaces $\beta \dot{f}(\lambda_t)$.

The equivalence of Z_{can} and Z_{mc} in the $N \rightarrow \infty$ limit for fixed $j(\cdot)$ follows directly from the asymptotic formula of the Bessel function given by Eq. (4.29).

$$\lim_{N \rightarrow \infty} Z_{mc}[j(\cdot)] = Z_{can}[j(\cdot)] \quad (4.66)$$

Similar to the discussion at the end of the previous section the meaning of this limit calls for some elaboration. As mentioned before the generating functional can be used to obtain correlation functions. For large but fixed N and given force protocol and temperature, the low order correlation functions for microcanonical and canonical ensembles will be very close. However one can always go to high enough orders

where relative differences will become significant. The limit in Eq. (4.66) means that given a certain order we can always choose a large enough N such that the microcanonical correlation functions up to that order agree with the corresponding canonical correlation functions to the desired degree.

4.5.3 Equilibration

In Chapter 2 we have proven equilibration for the quantum Brownian motion model at late times for an uncorrelated initial state whereby the bath is in its isolated thermal state. As argued in Sec. A.1 in the $\hbar \rightarrow 0$ limit this implies equilibration in the classical Brownian motion model. In this section we show that even a bath initially sampled from a microcanonical ensemble gives rise to equilibration in the classical Brownian motion model. Recall that we use the word bath for an infinite environment. Thus the thermodynamic or $N \rightarrow \infty$ limit is already taken in this setup, which is implicit in the assumptions (2.29).

As argued in Chapter 2 for the uncorrelated initial conditions the noise properties of the environment determine the late-time state of the system completely. Thus we could simply compare the probability distribution of noise for the two ensembles. We can do slightly better by directly comparing the resulting moment-generating functionals at late times. For this purpose let us redefine the generating functional (4.65) to allow for the late-time limit:

$$Z_{ens}[j(\cdot)] = \langle e^{\int_{t_0}^{t_0+\tau} dt j(t) X(t)} \rangle_{ens}, \quad (4.67)$$

Using the methods of this section (and almost identical calculations to that of Ap-

pendix B.1) one can show that:

$$\lim_{t_0 \rightarrow \infty} \lim_{N \rightarrow \infty} Z_{mc}[j(\cdot)] = Z_{can}[j(\cdot)], \quad (4.68)$$

which is valid for the proper choice of the microcanonical temperature (4.31).

4.6 Discussion

In this chapter we treated the exactly solvable model of a harmonic oscillator driven out of equilibrium by an external force and bilinearly coupled to an environment of N harmonic oscillators. An exact expression for the probability distribution of work, i.e. Eq. (4.35), is obtained for any value of N , assuming that the combined system and environment is initially sampled microcanonically. Using this expression the microcanonical Crook's relation (4.3) is verified. In the limit of an infinite environment, nonequilibrium work theorem (4.1) and Crooks's fluctuation theorem (4.2) are shown to hold. Finally in Sec. 4.5.2 the equivalence of all multi-time correlations of the system oscillator in the canonical and microcanonical ensembles in the infinite environment limit is obtained.

Our results support the hypothesis that for macroscopically large environments the sampling of the initial conditions from a canonical or microcanonical distribution is equivalent as far as system observables are concerned.

In the model used in this chapter the system oscillator is singled out not just by the virtue of the time-dependent force being only applied to it but also by the fact that all the environmental modes are coupled to it but not to each other. This may seem like a limitation of the model. However, the most general system of

coupled harmonic oscillators, i.e. allowing for the environmental oscillators to couple among themselves, can be represented by the model used in this chapter by first decomposing the environment into its eigenmodes, which in turn leads to a trivial change in the environment frequencies ω_n and coupling constants c_n [162]. Since we allow for arbitrary ω_n and c_n in our derivation, our model is able to represent any set of coupled harmonic oscillators. Also refer to Sec. 1.4.3 for further arguments about the generality of this model.

The fact that a microcanonical bath causes relaxation of the system to the same equilibrium state as that of a canonical bath (with the proper choice of temperature) when coupled to the system is yet another form of ensemble equivalence. Although all the forms of *ensemble equivalence* we discussed in this chapter are quite intuitive and by no means unexpected, they do not imply each other. At least there is no *a priori* reason why all these forms should be satisfied. In our studies of the Brownian motion model we have not come across any meaningful setup in which there is no *ensemble equivalence* in the thermodynamic limit.

Chapter 5: Macroscopic Quantum Phenomena

5.1 Introduction

Macroscopic quantum phenomena (MQP) manifest in a number of systems. Superconductivity is probably the oldest example; Bose-Einstein condensate (BEC) [163] and electro- and opto-mechanical devices [164, 165] are amongst the recent excitements. It is a relatively new research venue, with exciting ongoing experiments and bright prospects, yet with surprisingly little theoretical activity. From the traditional point of view that macroscopic objects are classical and quantum describes the microscopic realm, MQP appears like a transgression. This of course is what makes it interesting intellectually. This simplistic and hitherto rarely challenged view needs to be scrutinized anew, perhaps eventually with much of the conventional wisdoms repealed. In a series of papers we attempted to explore systematically into some key foundational issues of MQP, with the hope of finding a viable theoretical framework for this new endeavour. The three major themes discussed are the large N expansion, the correlation hierarchy and quantum entanglement for systems of “large” sizes, with many components or degrees of freedom.

5.1.1 Quantum / classical, micro / macro

There are many ways to deal with the issue of quantum-classical correspondence [122]. In the most common and traditional view the classical limit corresponds to $\hbar \rightarrow 0$, or, invoking the Bohr correspondence principle, the principal quantum number of a system $n \rightarrow \infty$, or regarding the coherent state as the “most classical” quantum state, or the Wigner function as the “closest to classical” distribution. Less precise criteria also abound, such as the loose concept that a system at high temperature behaves classically, or viewing the thermodynamic / hydrodynamic limits (of a quantum system) as classical (For a description of the various criteria, see, e.g., [121]). There are holes in almost all of the above common beliefs. A more sophisticated viewpoint invokes decoherence, the process whereby a quantum system loses its coherence (measured by its quantum phase information) through interaction with its environment [119]. In this chapter we examine the common belief that quantum pertains to the small (mass, scale) while classical to the large (size, multiplicity). This common belief now requires a much closer scrutiny in the face of new challenges from macroscopic quantum phenomena (MQP), namely, quantum features may show up even at macroscopic scales. A common example is superconductivity where the Cooper pairs can extend to very large scales compared to interatomic distances and Bose-Einstein condensate (BEC) where a large number N of atoms occupy the same quantum state, the N -body ground state. Other examples include nanoelectromechanical devices [166] where the center of mass of a macroscopic classical object, the cantilever, obeys a quantum mechanical equation

of motion. Experiments to demonstrate the quantum features such as the existence of interference between two macroscopic objects have been carried out, e.g., for C^{60} molecules passing through two slits [167] or proposed mirror superposition experiments [168, 169].

A most direct account of the difference between the microscopic and the macroscopic behaviours of a quantum system is by examining N , the number of physically relevant (e.g. for atomic systems, forgetting about the tighter-bound substructures) quantum particles or components in a macroscopic object. One may ask: At what number of N will it be suitable to describe the object as mesoscopic with qualitatively distinct features from microscopic and macroscopic? For classical systems significant advances in the recent decade have been made in providing a molecular dynamics basis to the foundations of thermodynamics [7], relating the macroscopic thermodynamic behaviour of a gas to the chaotic dynamics of its molecular constituents. One could even calculate the range in the number of molecules where a microscopic system begins to acquire macroscopic behaviour and hence identify the approximate boundaries of mesoscopia [170]. For quantum systems one needs to deal with additional concerns of quantum coherence and entanglement which are critically important issues in quantum information processing [171]. A fundamental issue in quantum information processing is how the performance of a quantum information processor alters as one scales the system up. This dependence on N is known as the “scaling” problem [172].

There are many important and interesting issues of MQP. Of special interest to us is how quantum expresses itself in the macroscopic domain since usually

macro conjures classicality. In this inquiry even the simplest yet far from naive questions need to be reconsidered properly. For example, why is it that an ostensibly macroscopic object such as a cantilever should follow a quantum equation of motion. The “center of mass ” is that the quantum mechanical behaviour of a macroscopic object placed in interaction with an environment, behavior such as quantum decoherence, fluctuations, dissipation and entanglement, can be captured by its COM behavior. This is implicitly assumed in many descriptions of MQP but rarely justified. The conditions upon which this can be justified are explored in [173] with the derivation of a master equation for N coupled harmonic oscillators (NHO) in a finite temperature harmonic oscillator bath (A mathematically more vigorous and complete treatment of NHO system is given in [174]). We continue to explore the conditions where one could infer macroscopic quantum behavior, specifically in terms of the existence and degree of quantum entanglement in this coupled NHO model. One aspect is in terms of entanglement at finite temperature [175–178] and large distance [69, 179], the other in terms of entanglement between different levels of structure (micro to meso to macro) [180] and the crucial role in a judicious choice of the appropriate collective variables [66, 181]. This is discussed in Section 5.2. In Section 5.4 we use the results of a recent paper on complex quantum network [70] to illustrate the somewhat counterintuitive finding that entanglement does not necessarily increase with connectivity but varies with the strength of coupling and the type of connectivity. (See also [182].)

5.1.2 Pathways toward understanding macroscopic quantum phenomena

In what follows we present two pathways as explored in two recent essays [64, 65]. The first concerns what macroscopic means. Does it mean large size or number of constituents? What about the degree of complexity of its constituents? What if the constituents are non-interacting versus interacting? Weakly interacting versus strongly interacting? The second pathway explores how quantum correlations and fluctuations impact on MQP using the n-particle-irreducible (nPI) representation.

5.1.2.1 Pathway 1: From the large N perspective

In this paper [64] we used different theories in a variety of contexts to examine the conditions or criteria whereby a macroscopic quantum system may take on classical attributes, and, more interestingly, that it keeps some of its quantum features.

The theories we considered there are, the $O(N)$ quantum mechanical model, semiclassical stochastic gravity and gauge / string theories; the contexts include that of a “quantum roll” in inflationary cosmology, entropy generation in quantum Vlasov equation for plasmas, the leading order and next-to-leading order large N behaviour, and hydrodynamic / thermodynamic limits. The criteria for classicality in our consideration include the use of uncertainty relations, the correlation between classical canonical variables, randomization of quantum phase, environment-induced

decoherence, decoherent history of hydrodynamic variables, etc.

All this exercise is to ask only one simple question: Is it really so surprising that quantum features can appear in macroscopic objects? By examining different representative systems where detailed theoretical analysis has been carried out, we found that there is no a priori good reason why quantum phenomena in macroscopic objects cannot exist.

5.1.2.2 Pathway 2: From the Correlation, Coupling and Criticality Perspectives

In this sequel paper [65] we explored how macroscopic quantum phenomena can be measured or understood from the behavior of quantum correlations which exist in a quantum system of many particles or components and how the interaction strengths change with energy or scale, under ordinary situations and when the system is near its critical point. We used the nPI (master) effective action related to the Boltzmann-BBGKY / Schwinger-Dyson hierarchy of equations as a tool for systematizing the contributions of higher order correlation functions to the dynamics of lower order correlation functions.

Together with the large N expansion discussed in our first paper [64] we explored the conditions whereby an H-theorem is obtained for the $O(N)$ model, which can be viewed as a signifier of the emergence of macroscopic behavior in the system. We compared the nonequilibrium dynamics of N atoms in an optical lattice under the large \mathcal{N} (field components), $2PI$ and second-order perturbative expansions, il-

illustrating the role of N and \mathcal{N} in quantum correlations, coherence and coupling strength. We also explored the behavior of an interacting quantum system near its critical point, the effects of quantum and thermal fluctuations and the conditions under which the system manifests infrared dimensional reduction. Finally we discussed how the effective field theory concept bears on macroscopic quantum phenomena: the running of the coupling parameters with energy or scale imparts a dynamical-dependent and an interaction-sensitive definition of “macroscopia”.

5.2 Levels of structure and the special role of collective variables

“Macroscopic” conveys a sense of being “large”, but what exactly does “largeness” mean? Do all the basic constituents of a large object contribute equally towards its quantum feature? (This point is highlighted in footnote 2 of [\[173\]](#).) In some cases we may actually know what the basic constituents are and how they are organized. A C^{60} molecule is made of carbon atoms, each atom is made of nuclei and electrons, each nucleus contains a certain number of protons and neutrons, each of them in turn is made up of quarks and gluons. Are we to simply count the number of quarks /gluons or protons /neutrons when we say an object is macroscopic? Obviously the tight binding of them to form a nucleus enters into our consideration when we treat the nucleus as a unit which maintains its own more or less distinct identity, features and dynamics. Thus when one talks about the mesoscopic or macroscopic behavior of an object one needs to specify which level of structure is of special interest, and how important each level contributes to these

characteristics.

The coupling strength between constituents at each level of structure (e.g., inter-atomic) compared to that structure’s coupling with the adjacent and remaining levels (which can be treated as an environment to this specific level of structure in an effective theory description, and its influence on it represented as some kind of noise [183, 184]) will determine the relative weight of each level of structure’s partaking of the macroscopic object’s overall quantum behavior. Often the best description of the behavior and dynamics of a particular level of structure is given by an effective theory for the judiciously chosen “collective variables”.

5.2.1 Choose the right collective variables before considering their quantum behavior

Same consideration should enter when one looks for the “quantumness” of an object, be it of meso or macro scale. One can quantize any linear system of whatever size, even macroscopic objects, such as sound waves from their vibrations. Giving it a name which ends with an “on” such as phonon and crowning it into a quantum variable is almost frivolous compared to the task of identifying the correct level of structure and finding the underlying constituents – the atoms in a lattice in this example, and their interactions. Constructing the relevant collective variables which best capture the salient physics of interest should come before one considers their quantum features. Thus, viewed in this perspective in terms of collective variables, we see that quantum features need not be restricted to microscopic objects. In

fact “micro” is ultimately also a relative concept as new “elementary” particles are discovered which make up the once regarded “micro” objects.

We illustrate this idea first with a discussion of the relevance of the center of mass variable in capturing the quantum features of a macroscopic object, then in the following sections, with a description of quantum entanglement between two macroscopic objects.

5.2.2 The quantum and macroscopic significance of center of mass variable

We can ask the question: what are the conditions upon which the mechanical and statistical mechanical properties of a macroscopic object can be described adequately in terms mainly of its center of mass (CoM) variable kinematics and dynamics, as captured by a master equation (for the reduced density matrix, with the environmental variables integrated out). The CoM axiom is an implicit assumption made in many MQP investigations, namely, that the quantum mechanical behavior of a macroscopic object, like the nanoelectromechanical oscillator [164, 166], a mirror [168], or a C^{60} molecule [167], placed in interaction with an environment can be captured adequately by its CoM behavior. This assertion is intuitively reasonable, as one might expect it to be true from normal-mode decompositions familiar in classical mechanics, but when particles (modeled by NHO) interact with each other (such as in a quantum bound state problem) in addition to interacting with their common environment, all expressed in terms of the reduced density matrix, it is not

such a clear-cut result.

With the aim of assessing the validity of the CoM axiom Chou, Hu and Yu [173] considered a system modeled by N harmonic oscillators interacting with an environment consisting of n harmonic oscillators and derived an exact non-Markovian master equation for such a system in a bath with arbitrary spectral density and temperature. The authors outlined a procedure to find a canonical transformation to transform from the individual coordinates (x_i, p_i) to the collective coordinates $(\tilde{X}_i, \tilde{P}_i), i = 1, \dots, N$ where \tilde{X}_1, \tilde{P}_1 are the center of mass coordinate and momentum respectively. In fact they considered a more general type of coupling between the system and the environment in the form $f(x_i)q_j$ (instead of the ordinarily assumed $x_i q_j$) and examined if the CoM coordinate dynamics separates from the reduced variable dynamics. They noted that if the function $f(x)$ has the property $\sum_{i=1}^N f(x_i) = \tilde{f}(\tilde{X}_1) + g(\tilde{X}_2, \dots, \tilde{X}_N)$, for example $f(x) = x$ or $f(x) = x^2$, one can split the coupling between the system and environment into couplings containing the CoM coordinate and the relative coordinates. Tracing out the environmental degrees of freedom q_i , one can easily get the influence action which characterizes the effect of the environment on the system.

However, as the authors of [173] emphasized, the coarse graining made by tracing out the environmental variables q_i does not necessarily lead to the separation of the CoM and the relative variables in the effective action. When they are mixed up and can no longer be written as the sum of these two contributions, the form of the master equation will be radically altered as it would contain both the relative variable and the center of mass variable dynamics.

With these findings they conclude that for the N harmonic oscillators quantum Brownian motion model, the coupling between the system and the environment need be bi-linear, in the form $x_i q_j$, for this axiom to hold. They also proved that the potential $V_{ij}(x_i - x_j)$ is independent of the center of mass coordinate. In that case, one can say that the quantum evolution of a macroscopic object in a general environment is completely described by the dynamics of the center of mass canonical variables $(\tilde{X}_1, \tilde{P}_1)$ obeying a master equation of the Hu-Paz-Zhang (HPZ) [28] type.

What is the relevance of this finding to MQP? Within the limitations of the N harmonic oscillator model it conveys at least two points: 1) For certain types of coupling the center of mass variable of an object composed of a large number of constituents does play a role in capturing the collective behavior of this object 2) Otherwise, more generally, the environment-induced quantum statistical properties of the system such as decoherence and entanglement could be more complicated. (For a similar conclusion considering the cross level (of structure) coarse graining, see [180].)

We next investigate the quantum entanglement between two macroscopic objects, comparing the entanglement between the micro-variables of their constituents in two types of couplings: one-to-one and one-to-all. The very different natures between these two types serve to illustrate the relevance of how the microscopic constituents organize into a macro object and how entanglement between collective variables reveals the quantum features of a macroscopic entity.

5.2.3 Two different interaction patterns

We first apply the methods developed in [173] to the study of the entanglement between the CoM coordinates of two macroscopic objects. Each macroscopic object is modeled by N identical coupled oscillators. However, unlike [173], we do not include an environment in our discussion because our focus is on the entanglement between the two objects induced by various types of direct interactions between their microscopic constituents. We denote the coordinates and the momenta of the microscopic constituents of the two macroscopic objects A and B by $\{x_{Ai}, p_{Ai}\}$ and $\{x_{Bi}, p_{Bi}\}$ respectively. The interactions between the microscopic constituents of one macroscopic object are assumed to be functions of the difference of variables only and we restrict ourselves to bilinear couplings between the microscopic constituents of the two macroscopic objects. The total Hamiltonian is then given by:

$$H_A = \sum_{i=1}^N \left(\frac{p_{Ai}^2}{2M} + \frac{1}{2} M \Omega^2 x_{Ai}^2 \right) + \sum_{iN_E j}^N V_{ij} (x_{Ai} - x_{Aj}), \quad (5.1)$$

$$H_B = \sum_{i=1}^N \left(\frac{p_{Bi}^2}{2M} + \frac{1}{2} M \Omega^2 x_{Bi}^2 \right) + \sum_{iN_E j}^N V_{ij} (x_{Bi} - x_{Bj}), \quad (5.2)$$

$$H_I = \sum_{i,j}^N \mathcal{G}_{ij} x_{Ai} x_{Bj}. \quad (5.3)$$

The canonical transformation described in the Appendix A of [173] can be applied to each object separately to yield a new set of phase space variables $\{\tilde{X}_{Ai}, \tilde{P}_{Ai}\}$ and $\{\tilde{X}_{Bi}, \tilde{P}_{Bi}\}$ and the associated masses \tilde{M}_i . Here $\tilde{X}_{A1} = \frac{1}{N} \sum_{n=1}^N x_{An}$ and $\tilde{X}_{B1} = \frac{1}{N} \sum_{n=1}^N x_{Bn}$ are the CoM coordinates. The Hamiltonians of the macroscopic objects

can be written in terms of these variables as:

$$H_A = \sum_{i=1}^N \left(\frac{\tilde{P}_{Ai}^2}{2\tilde{M}_i} + \frac{1}{2}\tilde{M}_i\Omega^2\tilde{X}_{Ai}^2 \right) + \tilde{V}(\tilde{X}_{A2}, \dots, \tilde{X}_{AN}) = H_{A,CoM} + H_{A,REL}, \quad (5.4)$$

$$H_B = \sum_{i=1}^N \left(\frac{\tilde{P}_{Bi}^2}{2\tilde{M}_i} + \frac{1}{2}\tilde{M}_i\Omega^2\tilde{X}_{Bi}^2 \right) + \tilde{V}(\tilde{X}_{B2}, \dots, \tilde{X}_{BN}) = H_{B,CoM} + H_{B,REL} \quad (5.5)$$

It has been proven in [173] that the potential \tilde{V} is not a function of the CoM variable.

This is a consequence of the form assumed for the potential energy. For a general bilinear coupling characterized by \mathcal{G}_{ij} the interaction Hamiltonian H_I can take on a complicated form, possibly mixing the CoM variables with the relative variables.

In what follows we will focus on two particular choices of \mathcal{G}_{ij} , inspired by Martins [181]. The use of the new set of canonical variables which include the CoM will help interpret the behaviour of macroscopic entanglement.

5.2.3.1 One-to-one interaction pattern

The one-to-one interaction pattern is defined by $\mathcal{G}_{ij} = \lambda\delta_{ij}$ (see Fig. 1(a)). In other words one constituent particle modeled by an oscillator from object A couples to one oscillator from object B, assuming that all pairwise couplings have the same strength. Using the canonical transformation of [173] it can be shown that the interaction Hamiltonian takes the form:

$$H_I = \sum_i^N \frac{\lambda}{M} \tilde{M}_i \tilde{X}_{Ai} \tilde{X}_{Bi}. \quad (5.6)$$

Note that pairwise interactions among the original variables translate into pairwise interactions among the transformed variables. A very important difference is that whereas the pairwise interactions in the original variables were all of equal strength,

the strength of the interactions scale with the mass of the variables after the transformation. As a result the relative strength of interactions between variable pairs are the same for all the variables, including the CoM. To see this explicitly let us consider the case with $\tilde{V} = 0$ for simplicity, namely the microscopic constituents of each macroscopic object do not interact with each other. Then we rescale the coordinates by $\tilde{X}_{Ai} \rightarrow \bar{X}_{Ai} \sqrt{M/\tilde{M}_i}$ and $\tilde{X}_{Bi} \rightarrow \bar{X}_{Bi} \sqrt{M/\tilde{M}_i}$, after which the Hamiltonian takes the form:

$$H = \sum_{i=1}^N \left(\frac{\bar{P}_{Ai}^2}{2M} + \frac{1}{2} M \Omega^2 \bar{X}_{Ai}^2 + \frac{\bar{P}_{Bi}^2}{2M} + \frac{1}{2} M \Omega^2 \bar{X}_{Bi}^2 + \lambda \bar{X}_{Ai} \bar{X}_{Bi} \right). \quad (5.7)$$

In this form it is easy to see that the effective strength of interactions in the CoM variable is the same as the effective strength of interactions in all the other variables. Hence the pairwise interaction pattern will induce the same amount of entanglement between pairs of transformed variables, without distinguishing the CoM variable. Entanglement between non-CoM variables would be directly effected if the interactions among the oscillators within the same object, i.e. V_{ij} , are not set to zero. If we only focus on the effect of the pairwise interactions, it is fair to say that such interactions couple the pairwise transformed variables with equal effective strength independent of the size N of the macroscopic objects. As a consequence we expect the behavior of entanglement between the corresponding variables of the objects to be independent of the size of the macroscopic objects, even for the CoM coordinate. For instance, at a given temperature the amount of entanglement between the two corresponding variables of the objects will not depend on N . Also the critical temperature above which entanglement ceases to exist is independent of N .

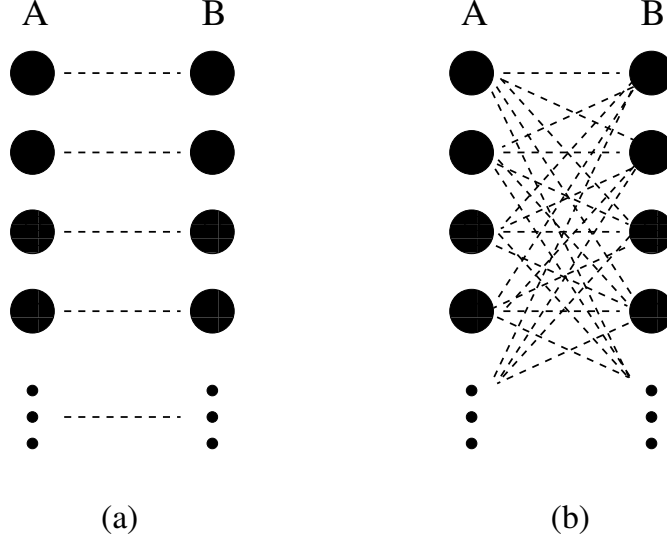


Figure 5.1: Schematic representation of the two types of couplings studied in this chapter:

(a) Pairwise or one-to-one interaction pattern (b) One-to-all interaction pattern

5.2.3.2 One-to-all interaction pattern

The one-to-all interaction pattern is characterized by $\mathcal{G}_{ij} = \lambda$ (see Fig. 1(b)). Then it is easy to see that the interaction Hamiltonian in the transformed variables takes the form:

$$H_I = N^2 \lambda \tilde{X}_{A1} \tilde{X}_{B1}. \quad (5.8)$$

Note that one-to-all interaction pattern corresponds to a coupling only between the CoM variables of the macroscopic objects, the relative variable Hamiltonian is unaffected. Thus one-to-all pattern differs from the pairwise pattern in that it distinguishes the CoM variable. Moreover if we perform the same rescaling of the previous section to determine the effective strength of this coupling we get:

$$H_{CoM} = \frac{\bar{P}_{A1}^2}{2M} + \frac{1}{2} M \Omega^2 \bar{X}_{A1}^2 + \frac{\bar{P}_{B1}^2}{2M} + \frac{1}{2} M \Omega^2 \bar{X}_{B1}^2 + N \lambda \bar{X}_{A1} \bar{X}_{B1}. \quad (5.9)$$

We see that the effective strength of the coupling increases with increasing N for the one-to-all pattern. Thus, in this case we expect the entanglement between the CoM variables to increase with increasing size of the macroscopic objects and survive at higher temperatures.

The one-to-all interaction pattern is crucial for the scaling of the entanglement of CoM variables with N . Hence it is important to investigate if this type of interaction pattern can occur in realistic situations and if so how common it is. It is clear that this pattern can not occur in arbitrarily large objects since this would imply forces which don't decay with distance. However, it might be applicable to a certain extend to many-body systems with long-range interactions or to nucleons in a large nucleus.

5.3 Conditions for CoM variable to decouple and its role in MQP

In this section we derive the necessary and sufficient conditions for the CoM variables of two macroscopic objects to decouple from the rest of the degrees of freedom. The macroscopic objects are modeled by N coupled oscillators and interact via bilinear couplings. Eqs.(5.4, 5.5) show that H_A and H_B obey the CoM axiom independently. Below, we demonstrate under which conditions $H_I = H_{I,CoM} + H_{I,REL}$ where $H_{I,CoM}$ is a function of the CoM coordinates of both macroscopic objects only and $H_{I,REL}$ does not depend on the CoM coordinates of either object.

Specifically, we will derive the conditions under which

$$\begin{aligned}
H_I &= H_{I,CoM}(\tilde{X}_{A1}, \tilde{X}_{B1}) + H_{I,REL}(\tilde{X}_{A2}, \dots, \tilde{X}_{AN}, \tilde{X}_{B2}, \dots, \tilde{X}_{BN}) \\
&= \tilde{G}_{11} \tilde{X}_{A1} \tilde{X}_{B1} + \sum_{i,j \in N_E} \tilde{G}_{ij} \tilde{X}_{Ai} \tilde{X}_{Bj}
\end{aligned} \tag{5.10}$$

To this end we follow the strategy adopted in Appendix C of Ref. [173] and determine the functional form of H_I by calculating its partial derivatives.

$$\frac{\partial H_I}{\partial \tilde{X}_{A1}} = \sum_{i,j} \mathcal{G}_{ij} \left(\frac{\partial x_{Ai}}{\partial \tilde{X}_{A1}} \right) x_{Bj} = \sum_j x_{Bj} \left(\sum_i \mathcal{G}_{ij} \right) \tag{5.11}$$

where in the second equality we used $\partial x_{Ai} / \partial \tilde{X}_{A1} = 1$ for all i , which can be shown by explicitly constructing a coordinate transformation whereby one coordinate is the CoM coordinate. For an example, consider the construction described in the Appendix B of [173] or the explicit construction described in Section 5.3.1, albeit with a different normalization for the CoM coordinate. Since we want H_I to have the form given by Eq.(5.10) we require $N \sum_i \mathcal{G}_{ij} = \tilde{G}_{11}$, which is independent of j . Repeating this derivation by replacing subscript A with B we obtain the second condition that $N \sum_j \mathcal{G}_{ij} = \tilde{G}_{11}$, which is independent of i . To summarize, the necessary and sufficient conditions for the CoM variables of both macroscopic objects to decouple from the relative coordinates is:

$$\tilde{G}_{11} = N \sum_i \mathcal{G}_{ij} = N \sum_j \mathcal{G}_{ij}. \tag{5.12}$$

If we do the same rescaling as in Eq. (5.7) we obtain:

$$H_{I,CoM} = \bar{G}_{11} \bar{X}_{A1} \bar{X}_{B2} \tag{5.13}$$

$$\bar{G}_{11} = \sum_{i=1}^N g_{ij} = \sum_{j=1}^N g_{ij} \tag{5.14}$$

As a quick check it can be easily verified that both the “pairwise” and “one-to-all” couplings satisfy this condition with $\bar{G}_{11} = \lambda$ and $\bar{G}_{11} = N\lambda$ respectively. These results agree with Eq. (5.7) and Eq. (5.9).

Up to this point we have only discussed two different patterns of couplings in detail, the pairwise and one-to-all. By analyzing Eq. (5.14) we can see how the effective coupling strength of the CoM variables will behave for different patterns. For instance, if the summations converge as $N \rightarrow \infty$ we conclude that in the thermodynamic limit the effective coupling strength of the CoM variables is an intrinsic quantity, independent of the size of the system. The only interaction pattern for which the effective coupling strength is extensive is the one-to-all pattern. Any other pattern for which the summation in Eq. (5.14) is divergent corresponds to an effective coupling that increases with the system size. If $|g_{ij}| > |g_{ik}|$ for $k > j > i$, this corresponds to a sub-linear growth. For example:

$$g_{ij} = \frac{\lambda}{(i-j)_{\text{mod}N} + c_1}, \quad \text{where } c_1 \text{ is a constant,} \quad (5.15)$$

$$\bar{G}_{11} = \lambda \sum_{i=1}^N \frac{1}{(i-j)_{\text{mod}N} + c_1} = \lambda \sum_{i=0}^{N-1} \frac{1}{i + c_1} \xrightarrow{\lim N \rightarrow \infty} \log(N) + c_2, \quad (5.16)$$

where c_2 is a constant that depends on c_1 and we used the convention $(-i)_{\text{mod}N} = N - i$ for $i < N$. The reason we used $(i-j)_{\text{mod}N}$ in the interaction term is in order to satisfy the condition Eq. (5.12). We see that in the thermodynamical limit the effective interaction strength scales as $\log(N)$.

5.3.1 Explicit Canonical Transformation for $N = 2^\nu$

In this section we describe a change of coordinates from the original set $\{x_{Ai}, x_{Bi}\}$ into a new set $\{X_{Ai}, X_{Bi}\}$. This new canonical transformation is more symmetric than the one used in the previous section and in [173], and allows for a general analysis for $N = 2^\nu$, where ν is an arbitrary integer. Since we are mainly interested in the behavior of the CoM coordinate (and how it differs from the rest of the coordinates), we require that the new set includes two coordinates X_{A1} and X_{B1} , which correspond to the CoM of objects A and B .¹

This set of new coordinates allow us to generalize our previous analysis to include randomness in the couplings between the microscopic constituents of objects A and B which is shown in the next subsection. Our results show that the CoM variable is singled out by i.i.d. (independent and identically distributed) random couplings between the micro-variables. Moreover, the properties of the canonical transformation provide insights into the reason why the CoM variable is special.

To distinguish this set of coordinates from those defined in Section 5.2.3 we drop the tilde. The transformation described below is only valid for $N = 2^\nu$. However, this is enough for our purpose of addressing MQP for large N .

Let \mathbf{A}_N be the matrix associated with the linear transformation from the

¹Note however that these are not the standard CoM coordinates but are rescaled by a factor of \sqrt{N} , i.e. $X_{A1} = (x_{A1} + \dots + x_{AN})/\sqrt{N}$. See Figure 5.3.1 for the definition of the rest of the coordinates. This rescaling is purely conventional and does not effect the physical conclusions drawn about the CoM.

original coordinates $\mathbf{x} = \{x_i\}$ to the new coordinates $\mathbf{X} = \{X_i\}$. In the rest of the chapter we denote vectors and matrices with bold characters, whereas individual entries will be indicated by regular characters with subscripts. Then

$$\mathbf{X} = \mathbf{A}_N \cdot \mathbf{x}. \quad (5.17)$$

Explicit form of \mathbf{A}_N for $N = 1, 2, 4, 8$ is given in Figure 5.3.1 and the procedure for obtaining $N = 2^\nu$ for arbitrary integer n is described. For brevity of notation the subscript N will be dropped in the rest of this section. A nice property of the transformation matrix is that $\mathbf{A}^{-1} = \mathbf{A} = \mathbf{A}^T$. Note that \mathbf{A} has the first column (row) of identical entries, which corresponds to the CoM coordinate. The relative coordinates defined in this section are different from those used in Section 5.2.3 as defined in [173]. The coordinates of this section are in a way more symmetric; for instance all the associated masses are equal to the original mass M . Demanding this symmetry together with the condition that there are two coordinates proportional to the CoM of each object forces a specific form on the matrix \mathbf{A} : all the columns (rows) corresponding to the relative coordinates have half of the entries with positive and the other half with negative sign and all equal magnitudes. This property, which singles out the CoM coordinate kinematically, will play an important role in the proceeding analysis.

Using the transformation (5.17) we can write the Hamiltonian for the new set of canonical variables. In this section we will set $V_{ij} = 0$ since we are interested in the effect of interactions between the constituents of the two macroscopic objects. The Hamiltonians H_A and H_B preserve their original form under the transformation

$$\mathbf{A}_1 = (1)$$

$$\mathbf{A}_2 = \frac{1}{\sqrt{2}} \begin{pmatrix} \boxed{1} & \boxed{1} \\ \boxed{1} & \boxed{-1} \end{pmatrix}$$

$$\mathbf{A}_4 = \frac{1}{\sqrt{4}} \begin{pmatrix} \boxed{1} & \boxed{1} & \boxed{1} & \boxed{1} \\ \boxed{1} & \boxed{-1} & \boxed{1} & \boxed{-1} \\ \boxed{1} & \boxed{1} & \boxed{-1} & \boxed{-1} \\ \boxed{1} & \boxed{-1} & \boxed{-1} & \boxed{1} \end{pmatrix}$$

$$\mathbf{A}_8 = \frac{1}{\sqrt{8}} \begin{pmatrix} \boxed{1} & \boxed{1} & \boxed{1} & \boxed{1} & \boxed{1} & \boxed{1} & \boxed{1} & \boxed{1} \\ \boxed{1} & \boxed{-1} & \boxed{1} & \boxed{-1} & \boxed{1} & \boxed{-1} & \boxed{1} & \boxed{-1} \\ \boxed{1} & \boxed{1} & \boxed{-1} & \boxed{-1} & \boxed{1} & \boxed{1} & \boxed{-1} & \boxed{-1} \\ \boxed{1} & \boxed{-1} & \boxed{-1} & \boxed{1} & \boxed{1} & \boxed{-1} & \boxed{-1} & \boxed{1} \\ \boxed{1} & \boxed{1} & \boxed{1} & \boxed{1} & \boxed{-1} & \boxed{-1} & \boxed{-1} & \boxed{-1} \\ \boxed{1} & \boxed{-1} & \boxed{1} & \boxed{-1} & \boxed{-1} & \boxed{1} & \boxed{-1} & \boxed{1} \\ \boxed{1} & \boxed{1} & \boxed{-1} & \boxed{-1} & \boxed{-1} & \boxed{-1} & \boxed{1} & \boxed{1} \\ \boxed{1} & \boxed{-1} & \boxed{-1} & \boxed{1} & \boxed{-1} & \boxed{1} & \boxed{1} & \boxed{-1} \end{pmatrix}$$

Figure 5.2: \mathbf{A}_N is the transformation matrix from the original coordinates to the new set of coordinates which include the CoM. A procedure to explicitly construct \mathbf{A}_N is illustrated above for $N = 2^\nu$. If \mathbf{A}_{2^ν} is known, $\mathbf{A}_{2^{\nu+1}}$ can be constructed by copying the block matrix \mathbf{A}_{2^ν} to the off-diagonal blocks and by putting the negative of it to the lower diagonal. Note that any transformation matrix for a smaller N can be obtained by restricting to the upper left corner of the larger matrix.

since $\mathbf{A}^T \mathbf{A} = \mathbf{1}$. The interaction Hamiltonian becomes:

$$H_I = \sum_{ij} X_{Ai} G_{ij} X_{Bj}, \quad \mathbf{G} = \mathbf{A} \cdot \mathbf{g} \cdot \mathbf{A}. \quad (5.18)$$

As a quick check of this formalism we calculate \mathbf{G} for the pairwise and one-to-all interaction patterns studied in Sections 5.2.3.1 and 5.2.3.2. For pairwise interactions $g_{ab} = \lambda \delta_{ab}$ and we get $G_{ij} = \lambda \sum_{ab} A_{ia} \delta_{ab} A_{bj} = \lambda (\mathbf{A}^2)_{ij} = \lambda \mathbf{1}_{ij} = \lambda \delta_{ij}$. Note that each coordinate is coupled with equal strength in this type of coupling (there was no need to renormalise the coordinates since the associated masses are already equal in this set of coordinates). For one-to-all interactions we have $\mathcal{G}_{ab} = \lambda$ which translates to $G_{ij} = \lambda \sum_{ab} A_{ia} A_{bj} = \lambda (\sum_a A_{ia}) (\sum_b A_{bj}) = N \lambda \delta_{i1} \delta_{1j}$. Note that only the CoM coordinates are coupled in this type of interaction and the coupling strength scales as N , which agrees with previous analysis.

5.3.2 Independent and Identically Distributed Gaussian Couplings

In previous sections we treated coupling patterns that are deterministic. It is reasonable to ask whether the conclusions we reach about the significance of the CoM coordinate and its decoupling from the relative coordinates are stable under perturbations. To investigate this issue, we reconsider the one-to-one and one-to-all patterns and this time allow for Gaussian variations around the non-zero coupling strengths. Our analysis shows that the conclusions of previous sections regarding the significance of the CoM variable and the decoupling of it from the relative coordinates are not altered by the addition of fluctuations.

Note that in what follows we do not allow for fluctuations in the vanishing

coupling strengths, for example the non-pairwise coupling strengths in the one-to-one pattern (see Eq. (5.24)). We motivate this choice by noting that the vanishing couplings can be the result of a constraint based on symmetry or geometry and thus immune to fluctuations. On the other hand, to assume that the values of non-zero coupling constants are fixed without fluctuations would be more difficult to justify, hence the need to study fluctuations.

5.3.2.1 One-to-all interaction pattern

With the canonical transformation of the previous section we can address the case where the coupling constants g_{ij} are sampled from identical independent Gaussian distributions characterised by the mean and variances:

$$\langle \mathcal{G}_{ab} \rangle = \bar{\mathcal{G}} \quad (5.19)$$

$$\langle \mathcal{G}_{ab} \mathcal{G}_{cd} \rangle - \langle \mathcal{G}_{ab} \rangle \langle \mathcal{G}_{cd} \rangle = \delta_{ac} \delta_{bd} \sigma_{\mathcal{G}}^2 \quad (5.20)$$

We now ask the question, how do the coupling constants G_{ij} behave? We can use the transformation (5.17) and the properties of the matrix \mathbf{A} to calculate the statistical

properties as:

$$\begin{aligned}
\langle G_{ij} \rangle &= \sum_{ab} A_{ia} \langle \mathcal{G}_{ab} \rangle A_{bj} \\
&= \bar{\mathcal{G}} \left(\sum_a A_{ia} \right) \left(\sum_b A_{bj} \right) = N \bar{\mathcal{G}} \delta_{i1} \delta_{j1} \quad (5.21) \\
\langle G_{ij} G_{kl} \rangle &= \left\langle \sum_{ab} A_{ia} \mathcal{G}_{ab} A_{bj} \sum_{cd} A_{kc} \mathcal{G}_{cd} A_{dl} \right\rangle \\
&= \sum_{abcd} A_{ia} A_{bj} A_{kc} A_{dl} (\bar{\mathcal{G}}^2 + \delta_{ac} \delta_{bd} \sigma_{\mathcal{G}}^2) \\
&= N^2 \bar{\mathcal{G}}^2 \delta_{i1} \delta_{j1} \delta_{k1} \delta_{l1} + \sigma_{\mathcal{G}}^2 (\mathbf{A}^2)_{ik} (\mathbf{A}^2)_{jl}
\end{aligned}$$

$$\langle G_{ij} G_{kl} \rangle - \langle G_{ij} \rangle \langle G_{kl} \rangle = \sigma_{\mathcal{G}}^2 \delta_{ik} \delta_{jl} \quad (5.22)$$

Thus if the couplings between the constituents of the macroscopic objects are statistically independent Gaussian random variables, the corresponding couplings between the new variables are also independent Gaussian random variables, which follows from the fact that the new and old variables are related by a linear transformation. The main difference is that only the CoM-to-CoM coupling has a non vanishing expectation value which is equal to the expectation value of the couplings of the original coordinates multiplied by N . As expected, the average behavior is that of the deterministic rule around which we are perturbing. On the other hand, the coupling constants of all of the new coordinates, i.e. both CoM and relative coordinates, have the same variance which is the same as the variance of the couplings of the original coordinates. Note that the fluctuations of the CoM coupling become negligible in the thermodynamic limit, but not those of the relative coordinates. Thus the conclusions of previous sections about the CoM hold with respect to the perturbations considered here in the thermodynamic limit.

5.3.2.2 One-to-one interaction pattern

Here we repeat the analysis of the previous section for the one-to-one coupling pattern. The coupling constants g_{ij} are sampled from identical independent Gaussian distributions characterised by the mean and variances:²

$$\langle \mathcal{G}_{ab} \rangle = \bar{\mathcal{G}} \delta_{ab} \quad (5.23)$$

$$\langle \mathcal{G}_{ab} \mathcal{G}_{cd} \rangle - \langle \mathcal{G}_{ab} \rangle \langle \mathcal{G}_{cd} \rangle = \delta_{ab} \delta_{ac} \delta_{bd} \sigma_{\mathcal{G}}^2 \quad (5.24)$$

We now ask the question, how do the coupling constants G_{ij} behave? After some algebra we get:

$$\begin{aligned} \langle G_{ij} \rangle &= \sum_{ab} A_{ia} \langle \mathcal{G}_{ab} \rangle A_{bj} = \bar{\mathcal{G}} \left(\sum_a A_{ia} A_{aj} \right) = \bar{\mathcal{G}} \delta_{ij} \quad (5.25) \\ \langle G_{ij} G_{kl} \rangle &= \left\langle \sum_{ab} A_{ia} \mathcal{G}_{ab} A_{bj} \sum_{cd} A_{kc} \mathcal{G}_{cd} A_{dl} \right\rangle \\ &= \sum_{abcd} A_{ia} A_{bj} A_{kc} A_{dl} \left(\bar{\mathcal{G}}^2 \delta_{ab} \delta_{cd} + \delta_{ab} \delta_{ac} \delta_{bd} \sigma_{\mathcal{G}}^2 \right) \\ &= \bar{\mathcal{G}}^2 \delta_{ij} \delta_{kl} + \sigma_{\mathcal{G}}^2 \sum_a (\mathbf{A}_{ia} \mathbf{A}_{ja} \mathbf{A}_{ka} \mathbf{A}_{la}) \\ |\langle G_{ij} G_{kl} \rangle - \langle G_{ij} \rangle \langle G_{kl} \rangle| &\leq \sigma_{\mathcal{G}}^2 / N. \quad (5.26) \end{aligned}$$

In the last step we used the fact that the matrix elements of \mathbf{A} are $\pm 1/\sqrt{N}$ to conclude that $|\sum_a (\mathbf{A}_{ia} \mathbf{A}_{ja} \mathbf{A}_{ka} \mathbf{A}_{la})| \leq 1/N$. We see that, unlike the one-to-all case, the CoM coupling behaves the same as those of the relative coordinates and for CoM as well as relative coordinates the fluctuations become negligible in the thermodynamic limit.

²Note the difference with Eq. (5.20) in the variance term. As mentioned before, we allow for fluctuations of couplings with non-vanishing means only.

5.4 Macroscopic quantum phenomena from the entanglement perspective

Entanglement is considered as a uniquely quantum feature [25], and quantum is habitually viewed as a zero or low temperature phenomenon, pertaining only to small systems. Both of these conditions are now being reconsidered, the “small system” restriction facing new challenges from macroscopic quantum phenomena (MQP) we are considering here. The “low temperature” restriction is lifted by theoretical observations [68, 185] and experimental proposals [186] that entanglement can survive at high temperatures, some even speculate that it is witnessed in biological systems [187].

5.4.1 Quantum entanglement at high temperatures and long distances?

Theoretical analysis of this issue for such systems has been carried out for coupled oscillator chain (1D) [175] or lattices (2D or 3D), where bounds and phase diagrams showing entangled and separable states were obtained by Anders [176]. For a nice exposé of the general issues on this topic we refer readers to the Discussion and Conclusion Section of [177].

Thermal entanglement can be studied by generalizing the (zero-temperature) quantum field mimicking a harmonic lattice to a thermal (finite temperature) field. In terms of model description quantum entanglement between two inertial harmonic

oscillators interacting via a zero-temperature quantum field was studied earlier by Lin and Hu [188] who showed that in addition to the temporal evolution of their entanglement there is also a dependence on their spatial separation. This generalization is done recently in [178] wherein both the temporal and spatial dependence of quantum entanglement studied before are shown to be sensitive to temperature variation. These authors also considered thermal entanglement in a harmonic lattice but with strong coupling, extending the comprehensive study of [177].

Another aspect is how much quantum entanglement can survive at large distance. *Long-range entanglement* in a coupled oscillator chain was claimed by Wolf et al [69]. Their setup of two harmonic oscillators interacting with a one dimensional harmonic lattice in a Gibbs state and their choice of parameters (continuum limit) map snugly to the thermal field model mentioned above. There are advantages in approaching the thermal entanglement issue for continuum systems from a quantum field theory perspective. Besides the technical ease to perform integrals over finite sums, the special properties of lower-dimensional systems (such as the Coleman-Mermin-Wegner theorem and the Berezinskii-Kosterlitz-Thouless phase transition) are well known, in some cases aided by elegant conformal field theory properties. This calculation is presently carried out in [179] where existence of zeros in the spectral density is found to be the cause of long-range entanglement. How general is this tie has yet to be decided.

5.4.2 Quantum networks: more connected not always more entangled

Finally we mention the results from a recent paper to illustrate a point on the relation between connectivity in a quantum network and entanglement. The following are excerpted from Cardillo *et al.* [70].

A network is defined as a set of N nodes and E edges accounting for their pairwise interactions. The network is usually characterized by its adjacency matrix, A , with elements $A_{ij} = 1$ if an edge connects nodes i and j while $A_{ij} = 0$ otherwise. We restrict attention to the undirected network where $A_{ij} = A_{ji}$. The Laplacian is related to the adjacency matrix by $L_{ij} = k_i \delta_{ij} - A_{ij}$, where $k_i = \sum_j A_{ij}$ is the connectivity of node i , *i.e.*, the number of nodes connected to i .

We can represent the nodes of the network by identical quantum oscillators interacting in accordance to the network topology encoded in L . The Hamiltonian of the harmonic quantum network is given by:

$$H_{\text{network}} = \frac{1}{2} \left(\mathbf{p}^T \mathbf{p} + \mathbf{x}^T (\mathbb{I} + 2cL) \mathbf{x} \right), \quad (5.27)$$

here \mathbb{I} is the $N \times N$ identity matrix, c is the coupling strength between connected oscillators while $\mathbf{p}^T = (p_1, p_2, \dots, p_N)$ and $\mathbf{x}^T = (x_1, x_2, \dots, x_N)$ are the operators corresponding to the momenta and positions of nodes respectively, satisfying the usual commutation relations: $[\mathbf{x}, \mathbf{p}^T] = i\hbar \mathbb{I}$.

The properties of the ground state of Hamiltonian (5.27) can be studied to quantify the amount of information each element of a network shares with the rest of the system via quantum fluctuations. Even at zero temperature the nodes are not

at rest due to Heisenberg uncertainty principle. Their spatial fluctuations depend on the pattern of physical interactions, *i.e.*, the network structure. To show this, the authors of Cardillo *et al.* [70] consider the partition of the network into a node, say i , and its complement i^c , *i.e.* the rest of the network. The mutual information shared by the two parties is given by:

$$\mathcal{I}(i|i^c) = S_i + S_{i^c} - S_{\text{tot}}. \quad (5.28)$$

Here S_i and S_{i^c} are marginal entropies and S_{tot} is the total entropy of the network. It is natural to choose the Von Neumann entropy to quantify the quantum information of the system, yielding $S_{\text{tot}} = 0$ for the ground state (as it is a *pure state*). Since the total network is in its ground (and pure) state we have $S_i = S_{i^c} = \mathcal{I}(i|i^c)/2$. Therefore, the information that a node shares with the network is intrinsically due to quantum correlations. Equivalently, the mutual information is, itself, a measure of the entanglement (quantified by S_i) between a single node and the rest of the system.

The authors then quantify the entanglement entropies of nodes embedded in different network topologies. They consider two homogeneous network substrates: (i) Random Regular Graphs (RRG), in which all the nodes have the same number of contacts ($k_i = \langle k \rangle, \forall i$), and (ii) Erdős-Rényi (ER) networks [189], for which the probability of finding a node with k neighbors, $P(k)$, follows a Poisson distribution so that most of the nodes have a degree k close to the average $\langle k \rangle$. They also analyze two networks having a scale-free (SF) pattern for the probability distribution, $P(k) \sim k^{-3}$, constructed by means of a configurational random model

(SF-CONF) [190] and the Barabási-Albert model (SF-BA) [191]. Their results are presented in plots of the average entanglement entropy of a node with connectivity k , i.e $\langle S_k \rangle$, vs k for the three network models: ER, SF-CONF and SF-BA.

Interesting features can be gleaned from the figures in [70]: Fig.2 plots $\langle S_k \rangle$ for fixed average connectivity $\langle k \rangle$ and 4 different values of coupling strength c . It shows that the average entanglement of a node with given connectivity k increases with increasing coupling strength. As a check the case $c = 0$ corresponds to non-interacting oscillators which in their ground state are not entangled. It is expected that as the interactions get stronger the ground state becomes more and more entangled. Fig.3 plots $\langle S_k \rangle$ vs k for fixed c and different values of $\langle k \rangle$. It can be seen that for fixed k the entanglement $\langle S_k \rangle$ increases for decreasing $\langle k \rangle$ for all graphs.

Here we offer some tentative explanations on such qualitative behaviors. We can understand this using the idea of monogamy of entanglement, which says that a system which is fully entangled to another system cannot be entangled to a third system. Keeping k fixed while decreasing $\langle k \rangle$ amounts to reducing the connections the neighbors of the node of interest has. Thus its neighbors have less neighbors to get entangled with. As a result they can be entangled more with the node of interest.

Another observation we can make from both Fig.2 and Fig.3, which is less intuitive, is the fact that $\langle S_k \rangle$ flattens out for ER for large $\langle k \rangle$ and first rises and then falls for SF. This indicates that for ER the nodes with large connectivity have all the same amount of entanglement with the rest of the network. On the other hand for SF there is an optimal number for the connectivity such that those nodes

with the optimal number of connections have the highest amount of entanglement with the rest of the network.

How can we make sense of this? Naively one expects the entanglement to increase with increasing number of connections, because more connections means more correlations. However entanglement is not just correlations. There may be a competition between correlations and monogamy of entanglement (or some argument using properties of quantum mutual information) that causes the rise and fall of entanglement in SF and the saturation in ER.

5.5 Discussion

In this chapter, we studied the relevance of interaction patterns between the microscopic constituents of two macroscopic objects, modeled by N harmonic oscillators, to the adequacy of the CoM variable as a quantifier of the quantum behavior of macroscopic objects. We focused on two types of coupling patterns : one-to-one (or pairwise) and one-to-all. In the former case we observed that although a transformation to a set of decoupled coordinates is possible, no linear transformation singles out a coordinate. This is in contrast to the latter case, where only the CoM coordinates of the objects are coupled and the coupling strength scales with N . Using the explicit transformation [5.3.1](#) we constructed for $N = 2^n$, we analyzed the effect of Gaussian fluctuations in the couplings among the microscopic constituents on the couplings among transformed coordinates. We showed that in the large N limit the fluctuations of the CoM coupling becomes negligible for both patterns, meaning that our conclusions are stable against this type of perturbations.

Chapter 6: Conclusion

In this dissertation we presented our contributions in three areas: nonequilibrium fluctuation theorems, equilibrium states in the strong coupling limit and macroscopic quantum phenomena. In this chapter we summarize our findings, discuss their implications to the issues raised in the Motivation [1.1](#) and point at future directions.

6.1 Equilibrium states in the strong coupling regime

An important component of nonequilibrium statistical mechanics is to describe the relaxation to equilibrium. In closed systems, like a gas in an isolated container, equilibration can occur due to the large number of constituents and the complexity of the underlying dynamics, as exemplified by the Boltzmann equation. On the other hand, integrable systems like QBM or small systems need to be coupled to a bath in order to equilibrate. Assuming the existence of thermal states in nature, we asked the following questions: to what state do systems (not necessarily weakly) coupled to large thermal environments relax, and how does this state differ from the thermal state for non-vanishing coupling strength? These questions are relevant for practical purposes, since in most experimental setups equilibrium systems are

prepared by being placed in thermal contact with a heat bath.

To address this question we investigated the late-time steady states of open quantum systems coupled to a thermal reservoir in the strong coupling regime in Chapter 2. Beyond the weak coupling limit the asymptotic state shows deviations from the canonical density matrix. We used a variety of nonequilibrium formalisms valid for non-Markovian processes to show that starting from a product state of the closed system = system + environment, with the environment in its thermal state, the open system which includes the back-action of the coarse grained environment will evolve towards an equilibrium state at late times. This state can be expressed as the state obtained by tracing out the environment in the closed system thermal state (at the temperature of the initial state of the environment):

$$\rho_C(0) = \rho_S(0) \otimes \frac{e^{-\beta \mathbf{H}_E}}{Z_E(\beta)}, \quad (6.1)$$

$$\lim_{t \rightarrow \infty} \rho_S(t) = \text{Tr}_E \left[\frac{e^{-\beta \mathbf{H}_C}}{Z_C(\beta)} \right] \equiv \rho_{\text{eq}}. \quad (6.2)$$

Above, S, E and C refer to system, environment and closed system = system + environment. \mathbf{H}_C includes an interaction term besides the system and environment Hamiltonians. $Z(\beta)$ denotes the partition function. We expect this result to be relevant to small systems to the degree they are likely to be strongly coupled to their environments.

In the vanishing coupling limit the relaxation rates vanish and relaxation takes an infinite amount of time. Thus, it is important to go beyond this limit in order to gain access to the time scales of equilibration. Note that the relaxation rates we are referring to here are those due to environmental influences. Other mechanisms

may bring about local equilibration in large systems at different rates which are independent of system size. Yet the time scales to reach equilibrium with an external environment are determined by the system-environment coupling strength.

For the exactly solvable quantum Brownian motion model we were able to go beyond the limitations of the density matrix formalism and show in a rigorous way that all multi-time correlations of the open system evolve towards those of the closed system thermal state. Multi-time correlations are especially relevant in the non-Markovian regime, since they cannot be generated by the dynamics of the single-time correlations using the quantum regression theorem. Showing equilibration at the level of density matrices only may not be enough for all applications. For instance, let us imagine a situation in which the equilibrium system of interest is coupled to yet another, much smaller system to which it serves as an environment. The system will induce non-Markovian dynamics on the smaller system since it is a finite environment, with the system's multi-time correlations determining the nature of the noise, such as multiplicative for nonlinear systems and colored as opposed to white. The state of the small system, given by its density matrix, and as a consequence all single-time expectation values will depend on noise statistics. This demonstrates that multi-time correlations play an important role in an equilibrium system's capacity to serve as an environment to other, much smaller, systems.

In this dissertation we have not focused on the nature of the difference between the equilibrium state defined in Eq. 6.2 and the thermal state. A quantification in terms of the Hamiltonian of mean force for the special case of an Ohmic environment is given by Hilt *et al.* [93]. Characterizing this difference for general environments

would be a useful next step.

For general systems beyond the linear quantum Brownian motion model, which cannot be exactly solved, we were able to provide a general proof that all single-time correlations of the open system evolve to those of the closed system thermal state, to second-order in coupling strength. For the special case of a zero-temperature reservoir, we were able to explicitly construct the reduced closed system thermal state in terms of the environmental correlations. Due to unavoidable degeneracy the second-order master equation cannot determine the second-order corrections to the diagonal elements of the density matrix. A fourth-order master equation is required to obtain the full second-order solution, so a useful extension of our work would be to attempt to derive the equilibrium state from the fourth-order master equation in order to prove full equilibration of the density matrix to second-order.

A complete proof, which would be non-perturbative for non-linear systems, would have to be very different than the second-order proof presented here. Our nonlinear proof, though very general in its application to different systems and environments, is not robust enough for non-perturbative multi-time correlations. It is not immediately clear how such a proof could be attempted, whereas the elegance of the final result makes the possibility of its existence seem reasonable.

An analogous proof for classical systems should be attempted by coarse graining the symplectomorphic (Hamiltonian) time evolution of the system and environment in much the same way that quantum master equations result from coarse graining the unitary time evolution of the system and environment. Unfortunately the literature on such an analog is not well developed (e.g., it would involve higher-

order Fokker-Planck equations which might only perturbatively preserve probability), and this would be more mathematically challenging than the quantum proof. Note that the $\hbar \rightarrow 0$ limit of the quantum results obtained in this chapter yield the corresponding classical results, as has been argued in Appendix A.1.

Finally, equilibration via coupling to a microcanonical environment, as opposed to a thermal state, has been demonstrated for the classical Brownian motion model in Chapter 4. It would be interesting to extend this result to the QBM model.

6.2 Quantum Fluctuation Theorems

The main conceptual obstacle in deriving fluctuation theorems for quantum systems is how to make sense of quantities like work in a quantum setting. Approaches in the literature to address this difficulty may be grouped into two, those who try to make sense of trajectories in quantum mechanics [98,103,107,192] (mostly applied to systems interacting with an environment or being monitored via measurements) and those proposing definitions of work that do not rely on the trajectory concept at all [63] (mostly applied to closed quantum systems). Our approach falls into the former category.

In Chapter 3 we used the decoherent histories conceptual framework to explain how the notion of trajectories in a quantum system can be made viable and subsequently used these trajectories to define work for open quantum systems. *Decoherent histories* formalism enables one to consider trajectories in quantum mechanics without the need to make measurements (or the existence of an observer), much like in

classical mechanics. In this formalism trajectories emerge in quantum mechanics as a result of coarse graining. Having justified the use of trajectories, we provided a first-principles analysis of the nonequilibrium work relation and Crooks’s fluctuation theorem for a quantum system interacting with a general environment based on the QBM model.

In the high-temperature limit we recover the classical fluctuation theorems as applied to the system + environment as a closed system. Moreover this result is proven for a bath composed of uncountably many oscillators, whereas standard proofs usually presuppose systems with a finite number of degrees of freedom. A phenomenological Langevin equation has been used before to derive fluctuation theorems for a damped harmonic oscillator [130] which is equivalent to the classical limit of our result. Yet phenomenological approaches have their limitations. For instance, in [130] the free energy difference is mistakenly interpreted as that of the free oscillator since there is not enough information to track down its origin. Moreover the system is initially assumed to be in its isolated thermal state with the noise being uncorrelated from initial conditions. We show that this scheme works only due to some coincidences particular to the linear oscillator model, whereas in the true Langevin equation there are strong correlations between system initial conditions and the noise, and the free energy difference is that of the combined system. This kind of ambiguity and disconnectedness, often found in phenomenological models in the literature, heightens the importance and advantage of using a first-principles approach based on microphysics models, as is adopted here.

At low temperatures we speculated on the existence of a regime where fluctu-

ation theorems formulated according to the classical prescription may not hold, but came short of working this out quantitatively. An analysis of the statistics of noise reveals that the discrepancy between quantum and classical gets larger at lower temperatures. At the same time noise gets weaker and only coarsely grained, or *fuzzy*, trajectories decohere. This is because the minimum level of coarse graining that would allow a consistent trajectory description is determined by the strength of the noise in the *decoherent histories* approach and thus depends on temperature. The *fuzziness* (or lack of precision) of the trajectories in turn reduces the accuracy at which averages can be calculated. Thus, there are two competing effects regarding possible violations of fluctuation theorems. At low temperatures deviations due to quantum effects become significant, but they are masked to some degree by the decreased precision at which trajectories can be meaningfully resolved. Here we emphasize a fundamental difference between classical and quantum systems. In quantum mechanics the ability to resolve trajectories is not only determined by the precision of the measurement device but also by the temperature.

In the future we want to answer the following question: Is there a temperature range for which experiments done to the precision prescribed by the decoherent histories interpretation result in deviations from the fluctuation theorems beyond that caused by the limited precision. If so what are the form of the corrections? Such a deviation would be due to quantum mechanics exclusively, and this is what we mean by violations of the fluctuation theorems at low temperatures due to quantum mechanical effects. To the best of our knowledge, this statement of the problem in terms of the interplay between precision and temperature is original to us.

The use of open quantum system concepts and especially the influence functional method adopted here enable us to define and quantify heat flow in terms of the dissipative dynamics of the open system which results from a self-consistent treatment of the back-action from its environment. We want to take advantage of this approach to address questions about energy exchange between the system and the bath, and apply it to fluctuation theorems related to entropy production.

6.3 Microcanonical Fluctuation Theorems

The sampling of initial conditions from the canonical ensemble in the derivation of fluctuation theorems for isolated systems is somewhat counter-intuitive since isolated systems are typically associated with the microcanonical ensemble. On the other hand, for a system interacting with a large environment it has been argued [96, 159, 160] that the validity of these theorems might be insensitive to the ensemble used and that the canonical ensemble should be viewed primarily as a computational convenience. This intuition is based on the idea of *ensemble equivalence*. In Chapter 4 we confirmed this intuition for the Brownian motion model via exact calculations, yet there are many reasons why this intuition should be doubted.

First, ensemble equivalence is usually discussed in homogenous systems (like a gas in a container) and not a system + environment setting. Also equivalence of ensembles is established for certain thermodynamic quantities like energy, pressure, etc. In the context of fluctuation theorems we are interested in fluctuations of work, which is a functional of the system trajectory over a time interval. Moreover, very

rare large fluctuations are known to dominate the average in the nonequilibrium work theorem. *Ensemble equivalence* results do not make any claim about such small probability events or about fluctuations in general; in fact they are aimed at the thermodynamic limit where fluctuations are small and usually neglected.

As a result, our demonstration of the validity of fluctuation theorems using the microcanonical ensemble in the infinite environment limit, albeit for a particular model, is valuable as a proof of principle. In the process we derived exact expressions for the work distribution for finite environments, from which the rate of convergence of various quantities can be obtained. We also showed the equivalence of all multi-time correlation functions of the system in the infinite environment limit for canonical and microcanonical ensembles. Finally we showed that when coupled to a harmonic oscillator both ensembles cause the system oscillator to relax to the same equilibrium state. Thus we have demonstrated *ensemble equivalence* in a number of ways that are usually not considered.

Treatment of a nonlinear model would be a natural extension to this work. The $O(N)$ x^4 theory we studied in some previous work [64, 65], not presented in this dissertation, appears to be a good candidate. This theory has a sensible large N limit and a controlled (non-perturbative) $1/N$ expansion we can work with.

6.4 Macroscopic Quantum Phenomena

In Chapter 5 we presented several pathways toward understanding MQP, identified some key issues we need to address or be concerned with, and provided some

examples to illustrate possibly counter-intuitive behavior. For quantum entanglement, specifically, using its extent and behavior to measure the quantumness of a system, we pointed out the necessity to recognize the levels of structure and the usefulness of collective variables in describing a macroscopic composite object when we try to identify its quantum features. One needs to be aware of the qualitative differences between the entanglement amongst the micro-constituents and that between collective variables which reveal MQP.

For concrete calculations we studied a model where two macro objects are made up of a large number of coupled oscillators. To understand how the coupling pattern amongst the constituents of the two macro objects enters into the picture, we considered two types of coupling, each constituent particle is coupled to only one other particle (1-to-1) versus coupled to all particles (1-to-all). In the 1-1 case with pairwise interactions of equal strength, the entanglement is independent of the number of constituent particles N in the macroscopic object. In the 1-to-all case the relative coordinates are decoupled and the center of mass (CoM) coupling scales with N . Here we expect the entanglement between the CoM variables to increase with increasing size of the macroscopic objects and survive at higher temperatures. We provided a general proof of the conditions whereby the CoM variables decouple from the rest. For the case the macro objects contain $N = 2^n$ constituents, we provided an explicit canonical transformation, with high symmetry properties, into a new set of coordinates containing the CoM. Using this transformation we showed that the decoupling of the CoM coordinates is stable under Gaussian fluctuations of the coupling strengths.

Finally we discussed the possibility of entanglement at finite temperature and at long-ranges, and used quantum coupled oscillator networks to illustrate the varying degrees of entanglement with different types of connectivity. We hope this sampling of ideas, approaches and illustrative examples will serve as a guide to thinking about the quantum nature of macroscopic objects, and, perhaps along the way, lead to a deeper understanding of quantum physics itself.

Chapter A: Appendix for Chapter 2

In this appendix we provide some technical details that were left out of the presentation in Chapter 2.

A.1 The triviality of classical, Gaussian noise

While Ref. [81] gives many cases in quantum mechanics in which the effect of system-environment coupling on the equilibrium state may be overlooked, here we would like to motivate the fact that this point is often overlooked in the classical regime as well, perhaps due to the ubiquitous employment of Gaussian noise. Let us consider the Hamiltonian of a system coupled linearly, via the system operator \mathbf{L} , to an environment of harmonic oscillators, indexed by k , which mock our Gaussian noise [26, 193].

$$\begin{aligned}\mathbf{H}_C &= \mathbf{H}_S + \sum_k \left[\frac{\mathbf{p}_k^2}{2m_k} + \frac{m_k \omega_k^2}{2} \mathbf{x}_k^2 \right] + \mathbf{L} \sum_k g_k \mathbf{x}_k + \mathbf{H}_R, \\ &= \mathbf{H}_S + \sum_k \left[\frac{\mathbf{p}_k^2}{2m_k} + \frac{m_k \omega_k^2}{2} \left(\mathbf{x}_k - \frac{g_k \mathbf{L}}{m_k \omega_k^2} \right)^2 \right],\end{aligned}\tag{A.1}$$

where the linear interaction is included in the square of the environment potential as a means of “renormalization”. Otherwise, the influence of the environment effectively introduces a negative \mathbf{L}^2 term proportional to the cutoff into the system

Hamiltonian when considering the open-system dynamics.

Tracing over the environmental degrees of freedom is equivalent to integrating over the environmental dimensions in phase space,

$$\mathrm{Tr}_E[\cdots] = \prod_k \int dx_k \int dp_k \cdots, \quad (\text{A.2})$$

where classically-speaking, x_k and p_k are independent, commuting variables. Therefore, in the classical and Gaussian model, relations (2.3) and (2.4) are equivalent as tracing over the environmental degrees of freedom constitutes a trivial Gaussian integral in phase space. The classical result can also be reached as the $\hbar \rightarrow 0$ limit of the quantum result. This limit is most straight-forward when applied to the Wigner function, since the $\hbar \rightarrow 0$ limit of the thermal state Wigner function is well-defined and gives the classical Boltzmann distribution function. In Eq. 1.15 this is shown explicitly for the simple harmonic oscillator but is true in general.

For classical open systems it is well known that if the system + environment is in a thermal state of the full Hamiltonian, which includes the system-environment coupling, then the reduced distribution of the system is in general not the thermal distribution of the system Hamiltonian alone. The term *potential of mean force* is used in (classical) chemical-physics literature for the quantity that replaces the potential energy in the Hamiltonian in the familiar Boltzmann distribution [161]. In the literature on quantum open systems the term *Hamiltonian of mean force* is used more often nowadays, which in addition includes the kinetic energy term as the name suggests. We will use this name for the classical case as well. The linear reservoir is a special case where the Hamiltonian of mean force coincides with the

system Hamiltonian. The Hamiltonian of mean force is defined by¹:

$$\mathbf{H}^*(\mathbf{X}, \mathbf{P}) \equiv -\frac{1}{\beta} \log \frac{\prod_k \int dx_k \int dp_k e^{-\beta \mathbf{H}_C(\mathbf{X}, \mathbf{P}; x, p)}}{\prod_{k'} \int dx_{k'} \int dp_{k'} e^{-\beta \mathbf{H}_E(x, p)}}. \quad (\text{A.3})$$

To the best of our knowledge, the asymptotic time evolution of a general classical open system, with a nonlinear environment initially in its thermal state, is not known. We conjecture that the reduced system is asymptotically described by $e^{-\beta \mathbf{H}^*}$ as described in the previous paragraph, and as would follow from (2.4). In this work we provide a proof of the analogous statement for quantum systems to second-order in interaction strength. Obviously, our second-order proof extends to classical systems which can arise in the limit $\hbar \rightarrow 0$. For linear systems we have an exact proof, and unlike its classical counterpart, the quantum linear case is highly nontrivial.

A.2 Theorems on matrix derivatives

Notation and Remarks: A letter in bold like \mathbf{A} indicates a matrix. Referring to an element of the matrix we use subscripts: \mathbf{A}_{ab} . The inverse of the matrix is indicated by \mathbf{A}^{-1} . An element of the inverse matrix is written as $(\mathbf{A}^{-1})_{ab}$ to avoid confusion with $1/\mathbf{A}_{ab}$. Transpose of the matrix is denoted by \mathbf{A}^T . Tr without a subscript indicates ordinary matrix trace. Tr_C indicates quantum mechanical trace

¹In most treatments \mathbf{H}_R is absent. In that case even for linear reservoir \mathbf{H}^* differs from \mathbf{H}_S by a frequency “renormalization”. For instance see Eq. (4.10) of Chapter 4. However, the point is that this simple frequency shift is much more trivial compared to the quantum mechanical case, where the Wigner function is altered in a highly nontrivial way due to the coupling to the environment.

over the closed system Hilbert space. A systematic study of matrix derivatives including some of the theorems below is given by [194].

Before proceeding to the derivations we clarify a mathematical subtlety. The theorems derived in this appendix will mostly be applied to symmetric matrices for which $\mathbf{A}_{ab} = \mathbf{A}_{ba}$. When taking the derivative of such a matrix with respect to one of its elements one can adopt two different conventions. If the derivative is taken under the constraint that only symmetric variations of the matrix is allowed the result is:

$$\frac{\partial \mathbf{A}_{ab}}{\partial \mathbf{A}_{cd}} = \delta_{ac}\delta_{bd} + \delta_{ad}\delta_{bc}(1 - \delta_{ab}). \quad (\text{A.4})$$

On the other hand if independent variations of all matrix elements are allowed the second term in the above equation is absent. In the following theorems we adopt the second convention.

Theorem 1. *Consider a system in a thermal state at inverse temperature β described by a Hamiltonian with parametric dependence on a set of variables $\{\lambda_n\}$. Then the expectation value of the derivative of the Hamiltonian with respect to these parameters can be calculated from the partition function by:*

$$\left\langle \frac{\partial \mathbf{H}}{\partial \lambda_n} \right\rangle_{\text{C}} \equiv \text{Tr}_{\text{C}} \left[\frac{\partial \mathbf{H}}{\partial \lambda_n} \frac{e^{-\beta \mathbf{H}}}{Z} \right] = -\frac{1}{\beta} \frac{\partial}{\partial \lambda_n} \ln(Z). \quad (\text{A.5})$$

Proof. In this proof we will make use of the following operator identity valid for an arbitrary operator \mathcal{O} :

$$\frac{\partial}{\partial \lambda_n} e^{\mathcal{O}} = \int_0^1 du e^{u\mathcal{O}} \frac{\partial \mathcal{O}}{\partial \lambda_n} e^{(1-u)\mathcal{O}}. \quad (\text{A.6})$$

Using this formula we can write the right hand side (RHS) of Eq. (A.5) as:

$$-\frac{1}{\beta} \frac{\partial}{\partial \lambda_n} \ln(Z) = -\frac{1}{\beta Z} \text{Tr}_C \left[\frac{\partial}{\partial \lambda_n} e^{-\beta \mathbf{H}} \right] = \frac{1}{\beta Z} \text{Tr}_C \left[-\int_0^1 du e^{-u\beta \mathbf{H}} \frac{\partial \beta \mathbf{H}}{\partial \lambda_n} e^{-(1-u)\beta \mathbf{H}} \right]. \quad (\text{A.7})$$

We use the cyclic property of trace to get:

$$-\frac{1}{\beta} \frac{\partial}{\partial \lambda_n} \ln(Z) = \frac{1}{Z} \text{Tr}_C \left[\int_0^1 du \frac{\partial \mathbf{H}}{\partial \lambda_n} e^{-\beta \mathbf{H}} \right] = \frac{1}{Z} \text{Tr}_C \left[\frac{\partial \mathbf{H}}{\partial \lambda_n} e^{-\beta \mathbf{H}} \right] = \left\langle \frac{\partial \mathbf{H}}{\partial \lambda_n} \right\rangle_C. \quad (\text{A.8})$$

□

Theorem 2. *For a matrix \mathbf{A}*

$$\text{Tr} \log \mathbf{A} = \log \det \mathbf{A}. \quad (\text{A.9})$$

Proof. Trace operation is basis-independent. In the basis in which \mathbf{A} is diagonal $\log \mathbf{A}$ is also a diagonal matrix with entries $\log a_n$ where a_n are the eigenvalues of \mathbf{A} . Taking the trace gives:

$$\text{Tr} \log \mathbf{A} = \sum_n \log a_n = \log \left(\prod_n a_n \right). \quad (\text{A.10})$$

The last expression is recognized to be $\log \det \mathbf{A}$ since the product of eigenvalues equals the determinant. □

Theorem 3. *For an arbitrary number of matrices \mathbf{A}_k indexed by k , the following is true:*

$$\text{Tr} \log \left(\prod_k \mathbf{A}_k \right) = \sum_k \text{Tr} \log(\mathbf{A}_k). \quad (\text{A.11})$$

Proof. To show this equality we make use of Theorem 2, the well known fact that the determinant of the product of matrices equals the product of the determinants

and properties of ordinary logarithms:

$$\begin{aligned}\mathrm{Tr} \log \left(\prod_k \mathbf{A}_k \right) &= \log \det \left(\prod_k \mathbf{A}_k \right) = \log \left(\prod_k \det \mathbf{A}_k \right) \\ &= \sum_k \log \det \mathbf{A}_k = \sum_k \mathrm{Tr} \log(\mathbf{A}_k) .\end{aligned}\tag{A.12}$$

□

A corollary of this theorem is the fact that $\mathrm{Tr} \log$ is invariant under any permutation of its arguments.

Theorem 4. *Consider a matrix \mathbf{A} and a parameter λ . Then:*

$$\frac{\partial}{\partial \lambda} \mathrm{Tr} \log \mathbf{A} = \mathrm{Tr} \left[\mathbf{A}^{-1} \frac{\partial \mathbf{A}}{\partial \lambda} \right].\tag{A.13}$$

In particular:

$$\frac{\partial}{\partial \mathbf{A}} \mathrm{Tr} \log \mathbf{A} = (\mathbf{A}^{-1})^T.\tag{A.14}$$

where $\frac{\partial}{\partial \mathbf{A}}$ is defined as the matrix obtained by differentiating with respect to the entries of matrix \mathbf{A} .

Proof. Let $\mathbf{A} \equiv 1 + \mathbf{B}$ and use

$$\log(1 + \mathbf{B}) = \mathbf{B} - \mathbf{B}^2/2 + \mathbf{B}^3/3 + \dots,\tag{A.15}$$

to write the left hand side (LHS) of Eq. (A.13) as:

$$\begin{aligned}\frac{\partial}{\partial \lambda} \mathrm{Tr} \left[\mathbf{B} - \frac{\mathbf{B}^2}{2} + \frac{\mathbf{B}^3}{3} + \dots \right] \\ = \mathrm{Tr} \left[\frac{\partial \mathbf{B}}{\partial \lambda} - \frac{1}{2} \left(\frac{\partial \mathbf{B}}{\partial \lambda} \mathbf{B} + \mathbf{B} \frac{\partial \mathbf{B}}{\partial \lambda} \right) + \frac{1}{3} \left(\frac{\partial \mathbf{B}}{\partial \lambda} \mathbf{B}^2 + \mathbf{B} \frac{\partial \mathbf{B}}{\partial \lambda} \mathbf{B} + \mathbf{B}^2 \frac{\partial \mathbf{B}}{\partial \lambda} \right) + \dots \right].\end{aligned}\tag{A.16}$$

Using the cyclic property of trace we obtain:

$$\text{Tr} \left[\frac{\partial \mathbf{B}}{\partial \lambda} (1 - \mathbf{B} + \mathbf{B}^2 - \mathbf{B}^3 \dots) \right]. \quad (\text{A.17})$$

Note that $\partial \mathbf{B} / \partial \lambda = \partial \mathbf{A} / \partial \lambda$ and

$$1 - \mathbf{B} + \mathbf{B}^2 - \mathbf{B}^3 + \dots = (1 + \mathbf{B})^{-1} = \mathbf{A}^{-1}, \quad (\text{A.18})$$

which proves Eq. (A.13). To prove Eq. (A.14) let $\lambda \equiv \mathbf{A}_{ab}$.

$$\frac{\partial}{\partial \mathbf{A}_{ab}} \text{Tr} \log \mathbf{A} = \text{Tr} \left[\mathbf{A}^{-1} \frac{\partial \mathbf{A}}{\partial \mathbf{A}_{ab}} \right] = \sum_{cd} (\mathbf{A}^{-1})_{cd} \frac{\partial \mathbf{A}_{dc}}{\partial \mathbf{A}_{ab}} = (\mathbf{A}^{-1})_{ba}. \quad (\text{A.19})$$

□

Theorem 5. *Let \mathbf{A} be an invertible matrix and λ a parameter. Then:*

$$\frac{\partial \mathbf{A}^{-1}}{\partial \lambda} = -\mathbf{A}^{-1} \frac{\partial \mathbf{A}}{\partial \lambda} \mathbf{A}^{-1}. \quad (\text{A.20})$$

In particular:

$$\frac{\partial (\mathbf{A}^{-1})_{ab}}{\partial \mathbf{A}_{mn}} = -(\mathbf{A}^{-1})_{am} (\mathbf{A}^{-1})_{nb}. \quad (\text{A.21})$$

Proof. We write $\mathbf{A}^{-1} = \mathbf{A}^{-1} \mathbf{A} \mathbf{A}^{-1}$, and differentiate both sides with respect to λ .

Looking at an element of this matrix equation we have:

$$\begin{aligned} \frac{\partial (\mathbf{A}^{-1})_{ab}}{\partial \lambda} &= \sum_{cd} \left(\frac{\partial (\mathbf{A}^{-1})_{ac}}{\partial \lambda} \mathbf{A}_{cd} (\mathbf{A}^{-1})_{db} + (\mathbf{A}^{-1})_{ac} \frac{\partial \mathbf{A}_{cd}}{\partial \lambda} (\mathbf{A}^{-1})_{db} \right. \\ &\quad \left. + (\mathbf{A}^{-1})_{ac} \mathbf{A}_{cd} \frac{\partial (\mathbf{A}^{-1})_{db}}{\partial \lambda} \right), \end{aligned} \quad (\text{A.22})$$

$$= \frac{\partial (\mathbf{A}^{-1})_{ab}}{\partial \lambda} + \left(\mathbf{A}^{-1} \frac{\partial \mathbf{A}}{\partial \lambda} \mathbf{A}^{-1} \right)_{ab} + \frac{\partial (\mathbf{A}^{-1})_{ab}}{\partial \lambda}. \quad (\text{A.23})$$

This proves Eq. (A.20). For the proof of Eq. (A.21) we set $\lambda = \mathbf{A}_{mn}$. □

A corollary of this theorem is the following identity valid for independent matrices \mathbf{A}_k :

$$\frac{\partial}{\partial \mathbf{A}_1} \text{Tr} [\mathbf{A}_2 \mathbf{A}_1^{-1} \mathbf{A}_3] = - (\mathbf{A}_1^{-1} \mathbf{A}_3 \mathbf{A}_2 \mathbf{A}_1^{-1})^T. \quad (\text{A.24})$$

A.3 N-QBM Partition Function

In this section we calculate the partition function of the N-QBM model. Our treatment mimics and generalizes that of Weiss [30], which treats one system oscillator only and does not allow for interactions among reservoir oscillators and non-diagonal mass matrix.² The partition function has an imaginary-time path integral representation given by:

$$Z_C = \oint \mathcal{D}\mathbf{x} \mathcal{D}\mathbf{X} \exp (-S^{(\text{E})}[\mathbf{x}, \mathbf{X}]/\hbar), \quad (\text{A.25})$$

$$S^{(\text{E})} = \int_0^{\hbar\beta} d\tau L^{(\text{E})}(\tau), \quad (\text{A.26})$$

$$L^{(\text{E})}(\tau) = \frac{1}{2} \left(\dot{\mathbf{X}}^T \mathbf{M} \dot{\mathbf{X}} + \mathbf{X}^T \mathbf{C} \mathbf{X} \right) + \frac{1}{2} \left(\dot{\mathbf{x}}^T \mathbf{m} \dot{\mathbf{x}} + [\mathbf{x} - \mathbf{c}^{-1} \mathbf{g} \mathbf{X}]^T \mathbf{c} [\mathbf{x} - \mathbf{c}^{-1} \mathbf{g} \mathbf{X}] \right), \quad (\text{A.27})$$

where $S^{(\text{E})}$ is the Euclidean action, τ the imaginary time and the path integral is over all periodic trajectories in the interval $[0, \hbar\beta]$. This path integral is Gaussian and can be evaluated exactly. It is convenient to represent the integration paths via

²Since a set of non-interacting oscillators can represent the most general Gaussian thermal reservoir, considering a non-diagonal mass matrix may appear superfluous. However we need the non-diagonal elements to generate the correlation function of two different reservoir momenta by partial differentiation of the partition function.

their Fourier series, which takes care of the condition on periodicity.

$$\mathbf{x}(\tau) = \sum_{r=-\infty}^{\infty} \mathbf{x}_r e^{i\nu_r \tau}, \quad (\text{A.28})$$

$$\mathbf{X}(\tau) = \sum_{r=-\infty}^{\infty} \mathbf{X}_r e^{i\nu_r \tau}, \quad (\text{A.29})$$

where $\mathbf{x}_{-r} = \mathbf{x}_r^\dagger$, $\mathbf{X}_{-r} = \mathbf{X}_r^\dagger$ (dagger stands for Hermitian conjugation) since $\mathbf{x}(\tau)$ and $\mathbf{X}(\tau)$ are real and $\nu_r \equiv 2\pi r/\hbar\beta$ are the bosonic Matsubara frequencies. Written in terms of the Fourier coefficients the Euclidean action becomes:

$$S^{(\text{E})} = \frac{\hbar\beta}{2} \sum_{r=-\infty}^{\infty} (\mathbf{X}_r^\dagger (\nu_r^2 \mathbf{M} + \mathbf{C}) \mathbf{X}_r) + \frac{\hbar\beta}{2} \sum_{r=-\infty}^{\infty} \left(\mathbf{x}_r^\dagger \nu_r^2 \mathbf{m} \mathbf{x}_r + [\mathbf{x}_r - \mathbf{c}^{-1} \mathbf{g} \mathbf{X}_r]^\dagger \mathbf{c} [\mathbf{x}_r - \mathbf{c}^{-1} \mathbf{g} \mathbf{X}_r] \right). \quad (\text{A.30})$$

Next we decompose $\mathbf{x}_r = \bar{\mathbf{x}}_r + \mathbf{y}_r$ where

$$\bar{\mathbf{x}}_r = (\nu_r^2 \mathbf{m} + \mathbf{c})^{-1} \mathbf{g} \mathbf{X}_r, \quad (\text{A.31})$$

is chosen such that $S^{(\text{E})}$ does not have a term linear in \mathbf{y}_r . The action can be written as:

$$S^{(\text{E})} = S_{\text{reservoir}}^{(\text{E})}[\mathbf{y}] + S_{\text{system}}^{(\text{E})}[\mathbf{X}], \quad (\text{A.32})$$

$$= \frac{\hbar\beta}{2} \sum_{r=-\infty}^{\infty} (\mathbf{y}_r^\dagger (\nu_r^2 \mathbf{m} + \mathbf{c}) \mathbf{y}_r) + \frac{\hbar\beta}{2} \sum_{r=-\infty}^{\infty} (\mathbf{X}_r^\dagger (\nu_r^2 \mathbf{M} + \mathbf{C} + 2\nu_r \hat{\gamma}(\nu_r)) \mathbf{X}_r), \quad (\text{A.33})$$

where the damping kernel is given by

$$\hat{\gamma}(z) = \frac{1}{2} \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \boldsymbol{\omega}^{-1} \frac{z}{\boldsymbol{\omega}^2 + z^2} \boldsymbol{\omega}^{-1} \mathbf{m}^{-\frac{1}{2}} \mathbf{g}, \quad (\text{A.34})$$

which is the Laplace transform of Eq. (1.34). The partition function of the closed system is given by:

$$Z_C = \mathcal{N} \int \prod_{r=-\infty}^{\infty} d\mathbf{X}_r \exp \left(-S_{\text{system}}^{(\text{E})}[\mathbf{X}]/\hbar \right) \int \prod_{r=-\infty}^{\infty} d\mathbf{y}_r \exp \left(-S_{\text{reservoir}}^{(\text{E})}[\mathbf{y}]/\hbar \right). \quad (\text{A.35})$$

The normalization factor \mathcal{N} is yet unspecified because it is not easy to determine the measure of the path integral. \mathcal{N} will be determined indirectly at the final stage of this calculation by considering the limiting case of no system-environment coupling.

The integrals in Eq. (A.35) are all Gaussian. Ignoring the normalization for now the integration gives:

$$Z_C \propto \prod_{r=-\infty}^{\infty} \frac{1}{\sqrt{\det[\nu_r^2 \mathbf{m} + \mathbf{c}]}} \frac{1}{\sqrt{\det[\nu_r^2 \mathbf{M} + \mathbf{C} + 2\nu_r \hat{\gamma}(\nu_r)]}}, \quad (\text{A.36})$$

$$\propto \frac{1}{\sqrt{\det[\mathbf{c}]}} \frac{1}{\sqrt{\det[\mathbf{C}]}} \prod_{r=1}^{\infty} \frac{1}{\det[\nu_r^2 \mathbf{m} + \mathbf{c}]} \frac{1}{\det[\nu_r^2 \mathbf{M} + \mathbf{C} + 2\nu_r \hat{\gamma}(\nu_r)]}. \quad (\text{A.37})$$

In the second line we used the fact that the elements of the product corresponding to positive and negative values of r are identical to restrict the product to positive r and pulled out the $r = 0$ entry. To determine the normalization let us recall the partition function for a simple harmonic oscillator:

$$Z_{\text{1HO}} = \frac{1}{2 \sinh(\beta \hbar \omega / 2)} = \frac{1}{\beta \hbar \omega} \prod_{r=1}^{\infty} \frac{\nu_r^2}{\omega^2 + \nu_r^2}. \quad (\text{A.38})$$

This naturally generalizes to N harmonic oscillators by:

$$Z_{\text{NHO}} = \frac{1}{\det[2 \sinh(\beta \hbar \boldsymbol{\omega} / 2)]} = \frac{1}{\beta \hbar \det[\boldsymbol{\omega}]} \prod_{r=1}^{\infty} \frac{\nu_r^2}{\det[\boldsymbol{\omega}^2 + \nu_r^2]}. \quad (\text{A.39})$$

In the limit of no coupling we demand that the partition function be a product of two partition functions of this form. This condition fixes the normalization and the

final answer is:

$$Z_C = Z_E \times \det\left(\frac{1}{\beta\hbar\Omega}\right) \prod_{r=1}^{\infty} \det\left(\frac{\nu_r^2}{\Omega^2 + \nu_r^2 + 2\mathbf{M}^{-\frac{1}{2}}\nu_r\hat{\gamma}(\nu_r)\mathbf{M}^{-\frac{1}{2}}}\right), \quad (\text{A.40})$$

where $Z_E = \text{Tr}[\exp(-\beta\mathbf{H}_E)]$ is the partition function of reservoir oscillators without coupling to the system. Using the definition(1.38) the partition function can also be written as:

$$Z_C = Z_E \times \det\left(\frac{1}{\hbar\beta\Omega}\right) \prod_{r=1}^{\infty} \det\left(\mathbf{M}\nu_r^2\hat{\mathbf{G}}(\nu_r)\right). \quad (\text{A.41})$$

A.4 Derivation of Eqs. (2.19-2.24)

In this appendix we derive some of the results presented in Sec. 2.2.2. Angular bracket with the subscript C denotes expectation values in the closed system thermal state. Expectation values in the uncorrelated state are denoted by attaching the subscript E to the bracket. Note that the damping kernel depends on the environmental variables and the coupling constants alone. There is no dependence on system variables. Using Eq. (2.10) we calculate the single-time system position-position correlation as:

$$\langle(\mathbf{X}\mathbf{X}^T)_{AB}\rangle_C = \frac{1}{\beta} \frac{\partial}{\partial \mathbf{C}_{AB}} \text{Tr} \log \mathbf{C} + \frac{2}{\beta} \frac{\partial}{\partial \mathbf{C}_{AB}} \sum_{r=1}^{\infty} \text{Tr} \log \left[\mathbf{M}^{-1} \hat{\mathbf{G}}(\nu_r)^{-1} \right], \quad (\text{A.42})$$

$$= \frac{1}{\beta} (\mathbf{C}^{-1})_{AB} + \frac{2}{\beta} \sum_{r=1}^{\infty} \text{Tr} \left[\left(\mathbf{M}^{-1} \hat{\mathbf{G}}(\nu_r)^{-1} \right)^{-1} \mathbf{M}^{-1} \frac{\partial \hat{\mathbf{G}}(\nu_r)^{-1}}{\partial \mathbf{C}_{AB}} \right], \quad (\text{A.43})$$

$$= \frac{1}{\beta} \hat{\mathbf{G}}(\nu_0)_{AB} + \frac{2}{\beta} \sum_{r=1}^{\infty} \hat{\mathbf{G}}(\nu_r)_{AB}, \quad (\text{A.44})$$

$$\langle \mathbf{X}\mathbf{X}^T \rangle_C = \frac{1}{\beta} \hat{\mathbf{G}}(\nu_0) + \frac{2}{\beta} \sum_{r=1}^{\infty} \hat{\mathbf{G}}(\nu_r), \quad (\text{A.45})$$

where we used the fact that \mathbf{C} and $\hat{\mathbf{G}}(\nu_r)$ are symmetric matrices and $\hat{\mathbf{G}}(\nu_0) = \hat{\mathbf{G}}(0) = \mathbf{C}^{-1}$. The system momentum-momentum correlations can be calculated in a similar way using Eq. (2.12).

$$\langle (\mathbf{P}\mathbf{P}^T)_{AB} \rangle_C = \frac{1}{\beta} \frac{\partial}{\partial (\mathbf{M}^{-1})_{AB}} \text{Tr} \log(\mathbf{M}^{-1}) \quad (\text{A.46})$$

$$+ \frac{2}{\beta} \frac{\partial}{\partial (\mathbf{M}^{-1})_{AB}} \sum_{r=1}^{\infty} \left(\text{Tr} \log [\mathbf{M}^{-1}] + \text{Tr} \log [\hat{\mathbf{G}}(\nu_r)^{-1}] \right),$$

$$= \frac{\mathbf{M}_{AB}}{\beta} + \frac{2}{\beta} \sum_{r=1}^{\infty} \left(\mathbf{M}_{AB} + \text{Tr} \left[\hat{\mathbf{G}}(\nu_r) \frac{\partial \hat{\mathbf{G}}(\nu_r)^{-1}}{\partial (\mathbf{M}^{-1})_{AB}} \right] \right), \quad (\text{A.47})$$

$$\langle \mathbf{P}\mathbf{P}^T \rangle_C = \frac{\mathbf{M}}{\beta} + \frac{2}{\beta} \sum_{r=1}^{\infty} \left(\mathbf{M} - \mathbf{M} \nu_r^2 \hat{\mathbf{G}}(\nu_r) \mathbf{M} \right). \quad (\text{A.48})$$

We used Theorem 3 in the first line. In the second line we used Theorem 4 for all terms and Theorem 5 for the last term with $\mathbf{A}_1, \mathbf{A}_2, \mathbf{A}_3 \rightarrow \mathbf{M}^{-1}, \nu_r^2 \hat{\mathbf{G}}(\nu_r), \mathbf{1}$.

For the system-environment position correlations note that only the damping kernel depends on the interaction matrix:

$$\frac{1}{\beta} \frac{\partial \log Z_C}{\partial (\mathbf{g}^T)_{Aa}} = -\frac{1}{\beta} \sum_{r=1}^{\infty} \frac{\partial}{\partial (\mathbf{g}^T)_{Aa}} \text{Tr} \log [\hat{\mathbf{G}}(\nu_r)^{-1}] - \frac{2}{\beta} \sum_{r=1}^{\infty} \sum_{BC} \hat{\mathbf{G}}(\nu_r)_{BC} \nu_r \frac{\partial \hat{\gamma}(\nu_r)_{CB}}{\partial (\mathbf{g}^T)_{Aa}}. \quad (\text{A.49})$$

The partial derivative of the damping kernel can be calculated explicitly. For this differentiation it is useful to rewrite $\hat{\gamma}(\nu_r)$ as:

$$2\nu_r \hat{\gamma}(\nu_r) = \nu_r^2 \mathbf{g}^T \mathbf{c}^{-1} (\mathbf{m}^{-1} + \nu_r^2 \mathbf{c}^{-1})^{-1} \mathbf{c}^{-1} \mathbf{g} \quad (\text{A.50})$$

For brevity of notation we define $\mathbf{a}(\nu_r)$ such that $\hat{\gamma}(\nu_r) = \frac{1}{2}\mathbf{g}^T \nu_r \mathbf{a}(\nu_r) \mathbf{g}$.

$$2\nu_r \frac{\partial \hat{\gamma}(\nu_r)_{\text{CB}}}{\partial (\mathbf{g}^T)_{\text{Aa}}} = \frac{\partial}{\partial \mathbf{g}_{\text{aA}}} \sum_{\text{ef}} (\mathbf{g}^T)_{\text{Ce}} (\nu_r^2 \mathbf{a}(\nu_r))_{\text{ef}} \mathbf{g}_{\text{fB}}, \quad (\text{A.51})$$

$$= \sum_{\text{ef}} \left\{ \delta_{\text{ea}} \delta_{\text{CA}} (\nu_r^2 \mathbf{a}(\nu_r))_{\text{ef}} \mathbf{g}_{\text{fB}} + (\mathbf{g}^T)_{\text{Ce}} (\nu_r^2 \mathbf{a}(\nu_r))_{\text{ef}} \delta_{\text{fa}} \delta_{\text{BA}} \right\}, \quad (\text{A.52})$$

$$= (\nu_r^2 \mathbf{a}(\nu_r) \mathbf{g})_{\text{aB}} \delta_{\text{CA}} + (\mathbf{g}^T \nu_r^2 \mathbf{a}(\nu_r))_{\text{Ca}} \delta_{\text{BA}}. \quad (\text{A.53})$$

Plugging this result in Eq. (A.49) we get:

$$\frac{1}{\beta} \frac{\partial \log Z_C}{\partial \mathbf{g}^T} = -\frac{2}{\beta} \sum_{r=1}^{\infty} \nu_r \hat{\mathbf{G}}(\nu_r) \hat{\gamma}(\nu_r) \mathbf{g}^{-1}. \quad (\text{A.54})$$

Using this result in Eq. (2.13) we get Eq. (2.21).

To derive Eq. (2.23) we start from Eq. (2.18):

$$-\frac{2}{\beta} \frac{\partial \log Z_C}{\partial \mathbf{m}^{-1}} = \langle p \mathbf{p}^T \rangle_{\text{E}} + \frac{2}{\beta} \sum_{r=1}^{\infty} \frac{\partial}{\partial \mathbf{m}^{-1}} \text{Tr} \log \left[\mathbf{M}^{-1} \hat{\mathbf{G}}^{-1}(\nu_r) \right]. \quad (\text{A.55})$$

Using Theorem 4 we get:

$$\frac{\partial}{\partial (\mathbf{m}^{-1})_{\text{ab}}} \text{Tr} \log \left[\mathbf{M}^{-1} \hat{\mathbf{G}}^{-1}(\nu_r) \right] = \text{Tr} \left[\hat{\mathbf{G}}(\nu_r) \frac{\partial \hat{\mathbf{G}}^{-1}(\nu_r)}{\partial (\mathbf{m}^{-1})_{\text{ab}}} \right], \quad (\text{A.56})$$

where

$$\frac{\partial \hat{\mathbf{G}}^{-1}(\nu_r)}{\partial (\mathbf{m}^{-1})_{\text{ab}}} = 2\nu_r \frac{\partial \hat{\gamma}(\nu_r)}{\partial (\mathbf{m}^{-1})_{\text{ab}}}. \quad (\text{A.57})$$

Next use Theorem 5 and plug the result back into Eq. (A.56):

$$2\nu_r \frac{\partial \hat{\gamma}(\nu_r)}{\partial (\mathbf{m}^{-1})_{ab}} = -\nu_r^2 \mathbf{g}^T \mathbf{a}(\nu_r)^{-1} \frac{\partial \mathbf{a}(\nu_r)}{\partial (\mathbf{m}^{-1})_{ab}} \mathbf{a}(\nu_r)^{-1} \mathbf{g}, \quad (\text{A.58})$$

$$= -\nu_r^2 \mathbf{g}^T \mathbf{c}^{-1} (\mathbf{m}^{-1} + \nu_r^2 \mathbf{c}^{-1})^{-1} \frac{\partial (\mathbf{m}^{-1} + \nu_r^2 \mathbf{c}^{-1})}{\partial (\mathbf{m}^{-1})_{ab}} (\mathbf{m}^{-1} + \nu_r^2 \mathbf{c}^{-1})^{-1} \mathbf{c}^{-1} \mathbf{g}, \quad (\text{A.59})$$

$$\begin{aligned} \text{Tr} \left[\hat{\mathbf{G}}(\nu_r) \frac{\partial \hat{\mathbf{G}}^{-1}(\nu_r)}{\partial (\mathbf{m}^{-1})_{ab}} \right] \\ = \sum_{\text{ABcd}} \hat{\mathbf{G}}(\nu_r)_{\text{AB}} \left(-\nu_r^2 (\mathbf{g}^T \mathbf{a}(\nu_r) \mathbf{c})_{\text{Bc}} \frac{\partial (\mathbf{m}^{-1} + \nu_r^2 \mathbf{c}^{-1})_{\text{cd}}}{\partial (\mathbf{m}^{-1})_{ab}} (\mathbf{c} \mathbf{a}(\nu_r) \mathbf{g})_{\text{dA}} \right). \end{aligned} \quad (\text{A.60})$$

Observe that:

$$\frac{\partial (\mathbf{m}^{-1} + \nu_r^2 \mathbf{c}^{-1})_{\text{cd}}}{\partial (\mathbf{m}^{-1})_{ab}} = \delta_{\text{ca}} \delta_{\text{db}}. \quad (\text{A.61})$$

It follows that

$$\begin{aligned} \text{Tr} \left[\hat{\mathbf{G}}(\nu_r) \frac{\partial \hat{\mathbf{G}}^{-1}(\nu_r)}{\partial (\mathbf{m}^{-1})_{ab}} \right] &= - \sum_{\text{AB}} \nu_r^2 \hat{\mathbf{G}}(\nu_r)_{\text{AB}} (\mathbf{g}^T \mathbf{a}(\nu_r) \mathbf{c})_{\text{Ba}} (\mathbf{c} \mathbf{a}(\nu_r) \mathbf{g})_{\text{bA}}, \\ &= - \left(\mathbf{c} \mathbf{a}(\nu_r) \mathbf{g} \nu_r^2 \hat{\mathbf{G}}(\nu_r) \mathbf{g}^T \mathbf{a}(\nu_r) \mathbf{c} \right)_{\text{ba}}. \end{aligned} \quad (\text{A.62})$$

In the last step we used the fact that both $\hat{\mathbf{G}}(\nu_r)$ and $\mathbf{a}(\nu_r)$ are symmetric matrices.

Eq. (A.56) becomes:

$$\frac{\partial}{\partial (\mathbf{m}^{-1})} \text{Tr} \log [\mathbf{M}^{-1} \hat{\mathbf{G}}^{-1}(\nu_r)] = -\mathbf{c} \mathbf{a}(\nu_r) \mathbf{g} \nu_r^2 \hat{\mathbf{G}}(\nu_r) \mathbf{g}^T \mathbf{a}(\nu_r) \mathbf{c}. \quad (\text{A.63})$$

We plug this into Eq. (A.55) and note that $\mathbf{c} \mathbf{a}(\nu_r) = \mathbf{m} \mathbf{f}(\nu_r)$ to get Eq. (2.23).

The derivation of Eq. (2.24) is almost identical to that of Eq. (2.23) but with more terms. We do not show the details of that derivation here.

A.5 Proof of conclusions of Sec. 2.2.4

Using the fact that all position-momentum correlations vanish we get:

$$\langle \boldsymbol{\xi}(s) \mathbf{X}_0^T \rangle_C = \mathbf{g}^T \dot{\mathbf{f}}(s) \mathbf{m} \langle \mathbf{x}_0 \mathbf{X}_0^T \rangle_C, \quad (\text{A.64})$$

$$\langle \boldsymbol{\xi}(s) \mathbf{P}_0^T \rangle_C = \mathbf{g}^T \mathbf{f}(s) \langle \mathbf{p}_0 \mathbf{P}_0^T \rangle_C, \quad (\text{A.65})$$

where the expectation values on the RHS are given by Eqs. (2.21, 2.22).

$$\begin{aligned} \langle \boldsymbol{\xi}(s) \mathbf{X}_0^T \rangle_C &= \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\boldsymbol{\omega} s)}{\boldsymbol{\omega}^2} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} \langle \mathbf{X}_0 \mathbf{X}_0^T \rangle_C \\ &\quad + \frac{2}{\beta} \sum_{r=1}^{\infty} \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\boldsymbol{\omega} s) \nu_r^2}{\boldsymbol{\omega}^2 (\boldsymbol{\omega}^2 + \nu_r^2)} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} \hat{\mathbf{G}}(\nu_r). \end{aligned} \quad (\text{A.66})$$

The first term on the right-hand side can be seen to decay by the fact that

$$\mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\boldsymbol{\omega} s)}{\boldsymbol{\omega}^2} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} = 2\gamma(s). \quad (\text{A.67})$$

The second term can be seen to decay by noting the inequality

$$\mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\boldsymbol{\omega} s) \nu_r^2}{\boldsymbol{\omega}^2 (\boldsymbol{\omega}^2 + \nu_r^2)} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} \leq \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\boldsymbol{\omega} s)}{\boldsymbol{\omega}^2} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} \leq 2\gamma(s),$$

in the sense of positive-definite matrix kernels, since both $\boldsymbol{\omega}^2$ and $(\boldsymbol{\omega}^2 + \nu_r^2)$ are positive matrices and cosine is a positive-definite kernel. The summation over r in Eq. (A.66) is finite as can be seen from Eq. (2.19). As a result $\langle \boldsymbol{\xi}(s) \mathbf{X}_0^T \rangle_C$ is a function that decays over time like $\gamma(s)$. When we take the convolution of this with another decaying function $\hat{\mathbf{G}}(t - s)$ and let $t \rightarrow \infty$ the overlap goes to zero. This way we argue that second line of Eq. (2.28) vanishes. A similar calculation

establishes the same goes for the third line.

$$\langle \boldsymbol{\xi}(s) \mathbf{P}_0^T \rangle_C = \mathbf{g}^T \mathbf{f}(s) \mathbf{c} \langle \mathbf{x}_0 \mathbf{X}_0^T \rangle_C \mathbf{M} + \mathbf{g}^T \mathbf{f}(s) \mathbf{g} \langle \mathbf{X}_0 \mathbf{X}_0^T \rangle_C \mathbf{M}, \quad (\text{A.68})$$

$$= \frac{1}{\beta} \sum_{r=1}^{\infty} \mathbf{g}^T \mathbf{f}(s) \mathbf{c} \mathbf{m}^{-\frac{1}{2}} \frac{\nu_r^2}{\omega^2(\omega^2 + \nu_r^2)} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} \hat{\mathbf{G}}(\nu_r) \mathbf{M}, \quad (\text{A.69})$$

$$= \frac{1}{\beta} \sum_{r=1}^{\infty} \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\sin(\omega s) \nu_r^2}{\omega(\omega^2 + \nu_r^2)} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} \hat{\mathbf{G}}(\nu_r) \mathbf{M}, \quad (\text{A.70})$$

$$= -\frac{1}{\beta} \sum_{r=1}^{\infty} \frac{d}{ds} \left[\mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\omega s) \nu_r^2}{\omega^2(\omega^2 + \nu_r^2)} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} \right] \hat{\mathbf{G}}(\nu_r) \mathbf{M}. \quad (\text{A.71})$$

The term inside square brackets decays as $\gamma(s)$ as can be seen from Eq. (A.68) and the argument following it. The summation over r is finite as before. Hence $\langle \boldsymbol{\xi}(s) \mathbf{P}_0^T \rangle_C$ decays over time like $\dot{\gamma}(s)$. The convolution of this with another decaying function $\mathbf{G}(t - \tau)$ gives zero in the limit $t \rightarrow \infty$.

The second and third lines of Eq. (2.28) are zero for the uncorrelated initial state as well. This follows trivially from: $\langle \boldsymbol{\xi}(s) \mathbf{X}_0^T \rangle_E = \langle \boldsymbol{\xi}(s) \mathbf{P}_0^T \rangle_E = 0$.

Finally we need to show that the fourth line of Eq. (2.28) is the same for both cases. This requires showing that the late-time limit of the noise kernel is the same. We know that the noise kernel is stationary for the uncorrelated initial state. Let us focus on the noise kernel of the closed system thermal state.

$$\langle \boldsymbol{\xi}(s) \boldsymbol{\xi}(s')^T \rangle_C = \mathbf{g}^T \left(\dot{\mathbf{f}}(s) \mathbf{m} \langle \mathbf{x}_0 \mathbf{x}_0^T \rangle_C \mathbf{m} \dot{\mathbf{f}}(s') + \mathbf{f}(s) \langle \mathbf{p}_0 \mathbf{p}_0^T \rangle_C \mathbf{f}(s') \right) \mathbf{g}. \quad (\text{A.72})$$

We use Eqs. (2.23, 2.24) on the RHS. The derivation is straightforward but tedious. The theorems in App. A.2 are utilized repeatedly.

The uncorrelated noise kernel is obtained if only the first terms in Eqs. (2.23, 2.24) are kept and the rest ignored. Hence we need to show that all the other terms vanish in the late-time limit. The strategy is the same as before: we show that

these terms are bounded by a function proportional to the damping kernel or its derivatives. We work out the details for two terms explicitly.

First consider the term in the noise kernel Eq. (A.72) due to the second term in Eq. (2.24).

$$\begin{aligned}
& \mathbf{g}^T \dot{\mathbf{f}}(s) \mathbf{m} \mathbf{c}^{-1} \mathbf{g} \langle \mathbf{X}_0 \mathbf{X}_0^T \rangle_C \mathbf{g}^T \mathbf{c}^{-1} \mathbf{m} \dot{\mathbf{f}}(s') \mathbf{g} \\
&= \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \cos(\omega s) \mathbf{m}^{-\frac{1}{2}} \mathbf{m} \mathbf{m}^{-\frac{1}{2}} \omega^{-2} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} \\
&\quad \times \langle \mathbf{X}_0 \mathbf{X}_0^T \rangle_C \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \omega^{-2} \mathbf{m}^{-\frac{1}{2}} \mathbf{m} \mathbf{m}^{-\frac{1}{2}} \cos(\omega s') \mathbf{m}^{-\frac{1}{2}} \mathbf{g}, \tag{A.73}
\end{aligned}$$

$$\begin{aligned}
&= \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\omega s)}{\omega^2} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} \langle \mathbf{X}_0 \mathbf{X}_0^T \rangle_C \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\omega s')}{\omega^2} \mathbf{m}^{-\frac{1}{2}} \mathbf{g}, \\
&= 4\gamma(s) \langle \mathbf{X}_0 \mathbf{X}_0^T \rangle_C \gamma(s'). \tag{A.74}
\end{aligned}$$

Unlike previous cases we were able to express this term exactly in terms of the damping kernel. It is a decaying function in both s and s' variables. The convolution of $\gamma(s)$ with $\hat{\mathbf{G}}(t-s)$ in Eq. (2.28) goes to zero if we let $t \rightarrow \infty$. Similarly the overlap of $\gamma(s')$ with $\hat{\mathbf{G}}(t'-s')$ vanishes in the limit $t' \rightarrow \infty$.

Secondly, consider the term in the noise kernel Eq. (A.72) due to the third term in Eq. (2.24).

$$\begin{aligned}
& -\frac{2}{\beta} \sum_{r=1}^{\infty} \mathbf{g}^T \dot{\mathbf{f}}(s) \mathbf{m}^{-\frac{1}{2}} \frac{1}{\omega^2(\omega^2 + \nu_r^2)} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} \nu_r^2 \hat{\mathbf{G}}(\nu_r) \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{1}{\omega^2(\omega^2 + \nu_r^2)} \omega^2 \mathbf{m}^{-\frac{1}{2}} \mathbf{m} \dot{\mathbf{f}}(s') \mathbf{g} \\
&= -\frac{2}{\beta} \sum_{r=1}^{\infty} \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\omega s)}{\omega^2(\omega^2 + \nu_r^2)} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} \nu_r^2 \hat{\mathbf{G}}(\nu_r) \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\omega s')}{\omega^2 + \nu_r^2} \mathbf{m}^{-\frac{1}{2}} \mathbf{g}, \\
&= \frac{2}{\beta} \sum_{r=1}^{\infty} \left[\mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\omega s) \nu_r^2}{\omega^2(\omega^2 + \nu_r^2)} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} \right] \frac{\hat{\mathbf{G}}(\nu_r)}{\nu_r^2} \frac{d^2}{ds'^2} \left[\mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\omega s') \nu_r^2}{\omega^2(\omega^2 + \nu_r^2)} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} \right]. \tag{A.75}
\end{aligned}$$

As before we conclude that the terms in square brackets decay like the damping kernel. The summation over r is finite as can be seen from Eq. (2.19) and noting

that $\nu_r > 1$ for all positive r .

Close inspection of all the other terms in Eq. (A.72) reveals that they have roughly the same form as those we worked out the details explicitly. All these terms vanish in the late-time limit.

This proves the equivalence of the late-time limit of the uncorrelated initial state to that of the late-time limit of the closed system thermal state. Since the closed system thermal state is stationary our proof is complete.

Chapter B: Appendix for Chapter 4

In this appendix we provide some technical details that are left out of the presentation in Chapter 4.

B.1 Derivation of the Main Result Eq.(4.20)

In this section we will compute the integrals appearing in Eq. (4.20). But first we review the integral representation of the delta function to be used in the derivation.

B.1.1 The Delta Function

The delta functions make the integrals in (4.20) difficult to evaluate. To get around this difficulty we invoke the following integral representation of the delta function:

$$\delta(H_C(\mathbf{Z}, \mathbf{z}; \lambda_0) - E) = \frac{1}{2\pi} \int_{-\infty}^{\infty} ds e^{-is(H_C(\mathbf{Z}, \mathbf{z}; \lambda_0) - E)}. \quad (\text{B.1})$$

The logic behind this is to convert the phase space integral into a simple Gaussian integral. After we perform that integral we will be able to do the s integration as well.

Observe that the integral formula for the delta function can be modified by allowing the integration variable s to have a constant imaginary part. We rename it z to emphasize the complex nature:

$$\frac{1}{2\pi} \int_{-\infty-i\epsilon}^{\infty-i\epsilon} dz e^{-iz(H_C(\mathbf{Z}, \mathbf{z}; \lambda_0) - E)} = \frac{1}{2\pi} \int_{-\infty}^{\infty} ds e^{-i(s-i\epsilon)(H_C(\mathbf{Z}, \mathbf{z}; \lambda_0) - E)} \quad (\text{B.2})$$

$$= e^{-\epsilon(H_C(\mathbf{Z}, \mathbf{z}; \lambda_0) - E)} \frac{1}{2\pi} \int_{-\infty}^{\infty} ds e^{-is(H_C(\mathbf{Z}, \mathbf{z}; \lambda_0) - E)} \quad (\text{B.3})$$

$$= e^{-\epsilon(H_C(\mathbf{Z}, \mathbf{z}; \lambda_0) - E)} \delta(H_C(\mathbf{Z}, \mathbf{z}; \lambda_0) - E) \quad (\text{B.4})$$

$$= \delta(H_C(\mathbf{Z}, \mathbf{z}; \lambda_0) - E), \quad (\text{B.5})$$

In the complex plane this contour passes parallel to the real axis, and is shifted down by an amount ϵ . One could reach the same result by noting that the integrand in (B.1) is an analytical function everywhere and thus the integration contour can be shifted down without changing the value of the integral. We will denote this contour by \mathcal{C} and use

$$\delta(H_C(\mathbf{Z}, \mathbf{z}; \lambda_0) - E) = \frac{1}{2\pi} \int_{\mathcal{C}} dz e^{-iz(H_C(\mathbf{Z}, \mathbf{z}; \lambda_0) - E)}. \quad (\text{B.6})$$

B.1.2 Denominator of Eq. (4.20)

The denominator of Eq. (4.20) gives the density of states associated with the initial Hamiltonian. Using Eq. (B.6), we write this density as:

$$\Sigma_i(E) = \frac{1}{2\pi} \int_{\mathcal{C}} dz e^{izE} \int d\mathbf{Z} e^{-izH_S(\mathbf{Z}; \lambda_0)} \int d\mathbf{z} e^{-iz(H_I(\mathbf{Z}, \mathbf{z}) + H_E(\mathbf{z}))} \quad (\text{B.7})$$

We begin by evaluating the last factor appearing above:

$$\int d\mathbf{z} e^{-\imath z(H_I(\mathbf{Z}, \mathbf{z}) + H_E(\mathbf{z}))} = \left(\frac{2\pi}{\imath\omega z}\right)^N \exp\left(\imath z X^2 \sum_n \frac{g_n^2}{2m_n\omega_n^2}\right) \quad (\text{B.8})$$

$$= \left(\frac{2\pi}{\imath\omega z}\right)^N e^{\imath z \gamma(0) X^2} \quad (\text{B.9})$$

where $\omega^N \equiv \omega_1 \cdots \omega_N$. The integrals are convergent due to the negative imaginary part of z as the contour \mathcal{C} is shifted below the real axis.

Using the definition of the renormalized frequency (4.10) we get:

$$\Sigma_i(E) = \frac{1}{2\pi} \left(\frac{2\pi}{\imath\omega}\right)^N \int_{\mathcal{C}} dz \frac{e^{\imath z E}}{z^N} \int d\mathbf{Z} e^{-\imath z H^*(\mathbf{Z}; \lambda_0)} \quad (\text{B.10})$$

$$= \frac{1}{\imath\bar{\Omega}} \left(\frac{2\pi}{\imath\omega}\right)^N \int_{\mathcal{C}} dz \frac{e^{\imath z \mathcal{A}}}{z^{N+1}}, \quad (\text{B.11})$$

where in the last equality we used the definition of \mathcal{A} introduced in Eq. (4.23). The sign of \mathcal{A} will play an important role later in the derivation.

$$\mathcal{A} = E + \mathcal{J}(\lambda) - \frac{f(\lambda)^2}{2M\bar{\Omega}^2} = H_C(\mathbf{Z}, \mathbf{z}; \lambda) + \mathcal{J}(\lambda) - \frac{f(\lambda)^2}{2M\bar{\Omega}^2} \quad (\text{B.12})$$

$$= \frac{P^2}{2M} + \frac{1}{2} M \bar{\Omega}^2 \left(X - \frac{f(\lambda)}{M\bar{\Omega}^2}\right)^2 + \sum_{n=1}^N \left[\frac{p_n^2}{2m_n} + \frac{1}{2} m_n \omega_n^2 \left(x_n - \frac{g_n}{m_n \omega_n^2} X\right)^2 \right] \geq 0. \quad (\text{B.13})$$

$\mathcal{A} = 0$ occurs only for a single point in the phase space. In the rest of this work we take $\mathcal{A} > 0$. The integral in Eq. (B.11) can be evaluated by enclosing the residue at the origin,

$$\int_{\mathcal{C}} dz \frac{e^{\imath z \mathcal{A}}}{z^{N+1}} = \frac{2\pi \imath}{N!} \imath^N \mathcal{A}^N. \quad (\text{B.14})$$

which finally brings us to the expression:

$$\Sigma_i(E) = \frac{1}{N!} \frac{(2\pi)^{N+1}}{\bar{\Omega}\omega^N} \mathcal{A}^N. \quad (\text{B.15})$$

B.1.3 Numerator of Eq. (4.20)

We begin by using Eq. (B.6) to express the numerator as follows:

$$\begin{aligned}
& e^{-\bar{\beta}\Delta\mathcal{J}} \frac{1}{2\pi} \int_{\mathcal{C}} dz e^{\imath z E} \int d\mathbf{Z} e^{-\imath z H_S(\mathbf{Z}; \lambda_0)} \int d\mathbf{z} e^{-\imath z (H_I(\mathbf{Z}, \mathbf{z}) + H_E(\mathbf{z}))} \\
& \quad \times e^{\bar{\beta} \int_0^\tau dt \dot{f}(\lambda_t) [XK(t) + PG(t) + \int_0^t ds G(t-s)(f(\lambda_s) - 2X\gamma(s) + \xi(s))]} \quad (\text{B.16}) \\
& = e^{-\bar{\beta}\Delta\mathcal{J}} e^{\bar{\beta} \int_0^\tau dt \int_0^t ds \dot{f}(\lambda_t) G(t-s) f(\lambda_s)} \frac{1}{2\pi} \int_{\mathcal{C}} dz e^{\imath z E} \\
& \quad \times \int d\mathbf{Z} e^{-\imath z H_S(\mathbf{Z}; \lambda_0) + X[\bar{\beta} \int_0^\tau dt \dot{f}(\lambda_t) K(t) - 2\bar{\beta} \int_0^\tau dt \int_0^t ds \dot{f}(\lambda_t) G(t-s) \gamma(s)] + P\bar{\beta} \int_0^\tau dt \dot{f}(\lambda_t) G(t)} \\
& \quad \times \int d\mathbf{z} e^{-\imath z (H_I(\mathbf{Z}, \mathbf{z}) + H_E(\mathbf{z})) + \bar{\beta} \int_0^\tau dt \int_0^t ds \dot{f}(\lambda_t) G(t-s) \sum_n g_n (x_n \cos(\omega_n s) + \frac{p_n}{m_n \omega_n} \sin(\omega_n s))}. \quad (\text{B.17})
\end{aligned}$$

To simplify the notation we define

$$\phi_n = g_n \bar{\beta} \int_0^\tau dt \int_0^t ds \dot{f}(\lambda_t) G(t-s) \cos(\omega_n s), \quad (\text{B.18})$$

$$\psi_n = g_n \bar{\beta} \int_0^\tau dt \int_0^t ds \dot{f}(\lambda_t) G(t-s) \sin(\omega_n s), \quad (\text{B.19})$$

$$I_K = \int_0^\tau dt \dot{f}(\lambda_t) K(t), \quad (\text{B.20})$$

$$I_G = M\bar{\Omega} \int_0^\tau dt \dot{f}(\lambda_t) G(t). \quad (\text{B.21})$$

After integration by parts the second factor of the first line of Eq. (B.17) can be rewritten as

$$e^{\bar{\beta} \int_0^\tau dt \int_0^t ds \dot{f}(\lambda_t) G(t-s) f(\lambda_s)} = e^{\bar{\beta} \mathcal{G} - \frac{\bar{\beta}}{M\bar{\Omega}^2} I_f - \frac{\bar{\beta}}{M\bar{\Omega}^2} f(\lambda_0) I_K}, \quad (\text{B.22})$$

where I_f has been defined in (4.25). The last Gaussian integral over \mathbf{z} in Eq. (B.17) yields:

$$\left(\frac{2\pi}{\imath \omega z} \right)^N e^{\imath z X^2 \sum_n \frac{g_n^2}{2m_n \omega_n^2} + X \sum_n \frac{g_n \phi_n}{m_n \omega_n^2} - \frac{\imath}{z} \sum_n \frac{\phi_n^2 + \psi_n^2}{2m_n \omega_n^2}}. \quad (\text{B.23})$$

The first term in the exponent above can be added to $-izH_S(\mathbf{Z}; \lambda_0)$ on the second line of Eq. (B.17) to give $-izH^*(\mathbf{Z}; \lambda_0)$. The second term in the exponent of Eq. (B.23) can be shown to be equal to:

$$X \sum_n \frac{g_n \phi_n}{m_n \omega_n^2} = X \bar{\beta} 2 \int_0^\tau dt \int_0^t ds \dot{f}(\lambda_s) G(t-s) \gamma(s). \quad (\text{B.24})$$

This term cancels the corresponding term on the second line of Eq. (B.17).

The third term of the sum in the exponent of Eq. (B.23) is independent of \mathbf{Z} and can be pulled out of the \mathbf{Z} integration. Using the definitions of ϕ_n and ψ_n it can also be written as

$$\mathcal{B} \equiv \sum_n \frac{\phi_n^2 + \psi_n^2}{2m_n \omega_n^2} = \bar{\beta}^2 \int_0^\tau dt \int_0^\tau dt' \dot{f}(t) \dot{f}(t') \int_0^t ds \int_0^{t'} ds' G(t-s) \gamma(s-s') G(t'-s'). \quad (\text{B.25})$$

In Appendix B.2 it is shown that the expression for \mathcal{B} can be simplified further by using the relations (3.25) to obtain:

$$2 \int_0^t ds \int_0^{t'} ds' G(t-s) \gamma(s-s') G(t'-s') = \frac{K(|t-t'|)}{M\bar{\Omega}^2} - \frac{K(t)K(t')}{M\bar{\Omega}^2} - G(t)G(t'), \quad (\text{B.26})$$

$$\mathcal{B} = \frac{\bar{\beta}^2}{2\bar{\Omega}^2} (2I_f - I_K^2 - I_G^2). \quad (\text{B.27})$$

The factor of two in front of I_f is due to the fact that both integration limits are from 0 to τ in Eq. (B.25) whereas the second integral is from 0 to t in Eq. (4.25).

Note that $\mathcal{B} \geq 0$, which can be seen from its definition (B.25). Together with (B.27) this indicates that $I_f \geq 0$. This fact will soon be used in the following derivation.

The \mathbf{Z} integration of Eq. (B.17) yields:

$$\int d\mathbf{Z} e^{-\imath z H^*(\mathbf{Z}, \lambda_0) + X \bar{\beta} I_K} = \frac{2\pi}{\imath \bar{\Omega} z} e^{-\imath \frac{(\bar{\beta} I_K + \imath z f(\lambda_0))^2}{2z M \bar{\Omega}^2} - \imath z \mathcal{J}(\lambda_0) - \frac{\imath}{z} \frac{\bar{\beta}^2 I_G^2}{2M \bar{\Omega}^2}}. \quad (\text{B.28})$$

Gathering all the terms Eq. (B.17) becomes, after a number of cancellations:

$$\frac{(2\pi)^N}{\imath^{N+1} \omega^N \bar{\Omega}} e^{-\bar{\beta} \Delta \mathcal{J}} e^{\bar{\beta} \mathcal{G} - \frac{\bar{\beta}}{M \bar{\Omega}^2} I_f} \int_{\mathcal{C}} dz \frac{e^{\imath z \mathcal{A} - \frac{\imath}{z} \frac{\bar{\beta}^2}{M \bar{\Omega}^2} I_f}}{z^{N+1}}. \quad (\text{B.29})$$

In order to proceed further we have to treat two cases separately: $I_f > 0$ and $I_f = 0$. For the more general case $I_f > 0$ we define \mathcal{D} as in (4.24) and change the integration variable to $z \rightarrow z \sqrt{\mathcal{D}/\mathcal{A}}$, where $\mathcal{D}/\mathcal{A} > 0$ as explained before. Then the integral becomes

$$\left(\frac{\mathcal{A}}{\mathcal{D}}\right)^{N/2} \int_{\mathcal{C}} dz \frac{e^{\imath \sqrt{\mathcal{A} \mathcal{D}} \left(z - \frac{1}{z}\right)}}{z^{N+1}}. \quad (\text{B.30})$$

which is proportional to a Bessel function of second kind: $(2\pi \imath) J_N(\imath 2 \sqrt{\mathcal{A} \mathcal{D}}) = (2\pi \imath) \imath^N I_N(\sqrt{4 \mathcal{A} \mathcal{D}})$. Therefore, the numerator in Eq. (4.20) can finally be written as

$$e^{-\bar{\beta} \Delta \mathcal{J}} e^{\bar{\beta} \mathcal{G} - \bar{\beta}^{-1} \mathcal{D}} \frac{(2\pi)^{N+1}}{\omega^N \bar{\Omega}} \frac{\mathcal{A}^{N/2}}{\mathcal{D}^{N/2}} I_N(\sqrt{4 \mathcal{A} \mathcal{D}}). \quad (\text{B.31})$$

Combining (B.15) and (B.31) we obtain the final result (4.22).

In the $I_f = 0$ case the integral in (B.29) is identical to Eq. (B.11), and cancels that term in Eq. (4.20) to yield:

$$\langle e^{-\bar{\beta} W} \rangle_{mc} = e^{-\bar{\beta} \Delta F}. \quad (\text{B.32})$$

Note that this is true for any choice of $\bar{\beta}$ irrespective of the total energy E . As is shown in Sec. 4.4.1, this is a consequence of the fact that $I_f = 0$ corresponds to a

delta function work distribution at $W = \Delta F$. One example of this case is given in Ref. [61]. For realistic environments we expect $I_f > 0$.¹

Finally we note that the result of $I_f = 0$ case, i.e. (B.32), can be recovered from that of $I_f > 0$ case by taking the limit $I_f \rightarrow 0$ (or equivalently $\mathcal{D} \rightarrow 0$) in (4.22) and using the asymptotic formula $I_N(x) \approx x^N/N!2^N$ as $x \rightarrow 0$. Thus Eq. (4.22) is valid for the most general case $I_f \geq 0$.

B.2 Derivation of Eq. (B.26)

To derive Eq. (B.25) we follow the method described in [130]. First observe that like any even function the damping kernel can be written as: $\gamma(t) = \gamma(t)\theta(t) + \gamma(-t)\theta(-t)$. We substitute this form into Eq. (B.25) and then take Laplace transforms with respect to t and t' denoted by the operators $\mathcal{L}_t(z)$ and

¹ $I_f \geq 0$ is a direct consequence of and can be proven directly using the fact that $\gamma(t)$ is a positive function. Realistic environments will most likely be described by strictly positive dissipation kernels which in turn yield the strict inequality $I_f > 0$ via application of Bohner's theorem [195]. The reason behind this is that for an environment with strictly positive dissipation kernel the average dissipated energy is always positive, whereas for a positive dissipation kernel it is possible that after a while all the dissipated energy, but not more, can flow back into the system. For any finite N and arbitrarily large τ this is certainly the case. But for large environments and realistic τ we expect this special case to be very improbable. $I_f < 0$ can not occur in our model as mentioned before, which is due to the fact that the harmonic oscillator environment is a passive environment.

$\mathcal{L}_{t'}(z')$ respectively.

$$\begin{aligned} \mathcal{L}_{t'}(z')\mathcal{L}_t(z) \left\{ 2 \int_0^t ds \int_0^{t'} ds' G(t-s)\gamma(s-s')\theta(s-s')G(t'-s') \right. \\ \left. + 2 \int_0^t ds \int_0^{t'} ds' G(t-s)\gamma(s'-s)\theta(s'-s)G(t'-s') \right\}. \end{aligned} \quad (\text{B.33})$$

Let us consider the first term. If we treat $\gamma(s-s')\theta(s-s')$ as a function of s only, the Laplace transform with respect to t has the form of a convolution of this function with $G(t-s)$. The result is the product of Laplace transforms of each function. Using the formula for the Laplace transform of time-shifted functions:

$$\mathcal{L}_t(z) \{f(t-a)\theta(t-a)\} = e^{-az}\hat{f}(z), \quad (\text{B.34})$$

we get for the first term of Eq. (B.33):

$$\mathcal{L}_{t'}(z') \left\{ 2 \int_0^{t'} ds' G(t'-s')\hat{\gamma}(z)e^{-zs'}\hat{G}(z) \right\} = 2 \frac{\hat{G}(z)\hat{G}(z')}{z+z'}\hat{\gamma}(z). \quad (\text{B.35})$$

An identical calculation, except for the change of the order of Laplace transforms, gives $2 \frac{\hat{G}(z)\hat{G}(z')}{z+z'}\hat{\gamma}(z')$ for the second term of Eq. (B.33). To write the final answer independent of the damping kernel we use Eq. (3.25) to express $\hat{\gamma}$ in terms \hat{G} and \hat{K} .

$$\frac{\hat{G}(z)\hat{G}(z')}{z+z'} \left(\frac{\hat{K}(z)}{M\hat{G}(z)} + \frac{\hat{K}(z')}{M\hat{G}(z')} - (z+z') \right) = \frac{\hat{K}(z)\hat{G}(z') + \hat{G}(z)\hat{K}(z')}{M(z+z')} - \hat{G}(z)\hat{G}(z'). \quad (\text{B.36})$$

Then write the first term exclusively in terms of \hat{K} again using Eq. (3.25), i.e.

$$\hat{G}(z) = \left(1 - z\hat{K}(z)\right) / M\bar{\Omega}^2.$$

$$\frac{1}{M^2\bar{\Omega}^2} \frac{\hat{K}(z) + \hat{K}(z')}{(z+z')} - \frac{\hat{K}(z)\hat{K}(z')}{M^2\bar{\Omega}^2} - \hat{G}(z)\hat{G}(z'). \quad (\text{B.37})$$

Using $\mathcal{L}_t(z)\mathcal{L}_{t'}(z')\left\{\frac{\hat{K}(z)}{z+z'}\right\} = \mathcal{L}_t(z)\left\{e^{-t'z}\hat{K}(z)\right\} = K(t-t')\theta(t-t')$, it is easily verified that the double inverse Laplace transform of Eq. (B.37) proves Eq. (B.25).

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