

## ABSTRACT

Title of dissertation: NON-MARKOVIAN DYNAMICS  
OF OPEN QUANTUM SYSTEMS

Chris H. Fleming, Doctor of Philosophy, 2011

Dissertation directed by: Professor Bei-Lok Hu  
Department of Physics

An *open quantum system* is a quantum system that interacts with some environment whose degrees of freedom have been coarse grained away. This model describes non-equilibrium processes more general than scattering-matrix formulations. Furthermore, the microscopically-derived environment provides a model of noise, dissipation and decoherence far more general than *Markovian* (white noise) models. The latter are fully characterized by Lindblad equations and can be motivated phenomenologically. Non-Markovian processes consistently account for back-reaction with the environment and can incorporate effects such as finite temperature and spatial correlations.

We consider linear systems with bilinear coupling to the environment, or *quantum Brownian motion*, and nonlinear systems with weak coupling to the environment. For linear systems we provide exact solutions with analytical results for a variety of spectral densities. Furthermore, we point out an important mathematical subtlety which led to incorrect master-equation coefficients in earlier derivations, given nonlocal dissipation. For nonlinear systems we provide perturbative solutions

by translating the formalism of canonical perturbation theory into the context of master equations. It is shown that unavoidable degeneracy causes an unfortunate reduction in accuracy between perturbative master equations and their solutions. We also extend the famous theorem of Lindblad, Gorini, Kossakowski and Sudarshan on completely positivity to non-Markovian master equations.

Our application is primarily to model atoms interacting via a common electromagnetic field. The electromagnetic field contains correlations in both space and time, which are related to its relativistic (photon-mediated) nature. As such, atoms residing in the same field experience different environmental effects depending upon their relative position and orientation. Our more accurate solutions were necessary to assess sudden death of entanglement at zero temperature. In contrast to previous claims, we found that all initial states of two-level atoms undergo finite-time disentanglement. We were also able to access regimes which cannot be described by Lindblad equations and other simpler methods, such as near resonance.

Finally we revisit the infamous Abraham-Lorentz force, wherein a single particle in motion experiences backreaction from the electromagnetic field. This leads to a number of well-known problems including pre-acceleration and runaway solutions. We found a more a more-suitable open-system treatment of the nonrelativistic particle to be perfectly causal and dissipative without any extraneous requirements for finite size of the particle, weak coupling to the field, etc..

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OF OPEN QUANTUM SYSTEMS

by

Chris H. Fleming

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Advisory Committee:  
Professor Bei-Lok Hu, Chair/Advisor  
Professor Jeffery Bub  
Professor Alex Dragt  
Professor Philip R. Johnson  
Professor Eite Tiesinga

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## Preface

This thesis represents the majority, but not entirety, of my research under Prof. Bei-Lok Hu. The reader will find no mention of my original research on the topic of dissipative quantum cosmology. In building proper foundations for my understanding of dissipative quantum mechanics, I became sidetracked into more practical pursuits. I soon found the interface of quantum and classical mechanics, non-equilibrium field theory, statistical mechanics and thermodynamics to be sufficiently distracting and challenging without the inclusion of gravity.

Chapter 3 comprises the culmination of my first venture into dissipative quantum mechanics, working out the general master equations and solutions to quantum Brownian motion (QBM). It was a taxing effort, as for the first few years we labored under a set of false assumptions which had propagated throughout the entire literature on the subject. According to my advisor, this work alone was more than sufficient to form a thesis.

Chapter 2, which is presented first for pedagogical reasons, is a result of the direction I took as to consider nonlinear systems. This work gave me a much broader perspective of dissipative quantum mechanics, now of the Feynman-Vernon, Langevin and Fokker-Plank variety among others. With a solid and well-rounded foundation in several perspectives, I was able to steadily work out a number of more-pointed theoretical results in Chapter 4 and applications in Chapter 6.

Having had some experience with particles in fields, I naturally reconsidered the infamous Abraham-Lorentz force, a pathological physical law which one can find

described in any introductory textbook on electrodynamics. The Abraham-Lorentz force, which has been derived in a variety of ways, dictates that if you accelerate an electrically charged point particle, then it will radiate electromagnetic energy until this acceleration ceases. However, on a much slower timescale the particle will slowly build up acceleration and ultimately fly off to the nether regions of the universe. Immediately I knew that the unphysical aspects of this problem could not be *mathematically* correct, at least for the nonrelativistic particle. Linear coupling to a thermal reservoir (which is what one has in the non-relativistic regime) should not lead to runaway solutions. With the theoretical foundation that I had built for myself, it took me only minutes to find a well-behaved solution to the problem of the dissipation (or backreaction) that a nonrelativistic charged particle experiences in the electromagnetic field. However, it then took me many months to explore all of the nuances which distinguish this result from others. This most recent result we present in our final Chapter 7.

## Dedication

*To Amanda,*

*my wife and friend,*

*whose loving support has never wavered,*

*given sufficient time averaging.*

## Acknowledgments

First and foremost I must acknowledge my family, without whose support this work would not be possible. Especial dedication goes out to my patient wife Amanda, who supported me throughout graduate school, to my providing father Howard who supported me throughout undergraduate school, and to my generous in-laws David and Rhonda Smith, who supported my wife while she was supporting me.

I received a far more than adequate undergraduate education at the University of South Alabama, under the tutelage of my physics advisor Prof. Justin Sanders and my mathematics advisor Prof. Vasilij Prokhorov, among many other excellent professors. Prof. Sanders was unable to sway me into the direction of experimental physics, but our published work did exempt me from graduate laboratory. I am exceptionally grateful for this exemption, as are all of the people not harmed by giving me access to dangerous laboratory equipment. I am equally grateful to Prof. Prokhorov, who gave me a very useful background in approximation theory, which sounds harmless enough, and more specifically in the subject of Padé approximants, which I have heard another scientist describe as a “black art”. Prof. Prokhorov will likely be happy that I rely upon rational approximants in various parts of this manuscript. When asked about such peculiarities, I inform others that I was trained in mathematics by Russians.

I have also received an exceptional graduate education at the University of Maryland. I am grateful for the oversight of my advisor Prof. Bei-Lok Hu, whose

broad interests in fundamental physics allowed me to find my own research path, yet without venturing too far into the realm of more nebulous topics.

I am grateful to Dr. Albert Roura, whom I worked very closely with on quantum Brownian motion (QBM) and subsequent papers. It was, in fact, Albert who first noticed that the propagators of initial conditions and sources did not have to be the same for integro-differential equations. With this small discovery, a modest paper detailing some solutions to QBM became a voluminous Annals of Physics manuscript necessary to correct the previous literature. Chapter 3 of this thesis could easily make for another such paper.

I am grateful to my coauthor and fellow rogue Nick Cummings who has frequently helped me translate physics into my native language of mathematics. It was Nick who carefully determined that some of my solutions to the second-order master equation violate positivity for vanishing temperature. This finding allowed me to track down an important and yet previously undiscovered problem which plagues all perturbative master equations, and which has subsequently been described by a fellow physicist as an “horrible” discovery and that “you found this, you must fix it”. Unfortunately I have not found the time to do that.

I am grateful to Prof. Charis Anastopoulos for his collaboration on the study of the rotating-wave approximation and multipartite entanglement dynamics. Our discussion of non-Markovian processes and non-equilibrium renormalization schemes were particularly useful.

Finally I must express gratitude to my most recent collaborator Prof. Philip Johnson, whom I have been working with on the Abraham-Lorentz force up to the

moment of this writing. When I first discovered stable solutions to electromagnetic backreaction of charged particles, I had only the most superficial understanding of the relevant literature. But this was an ignorance of fortune, as a better understanding would have likely directed me towards the standard result. It was Phil's prodding and discussion that eventually led me to truly understand the problem.

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## List of Abbreviations

$\alpha$	correlation kernel
$\beta$	inverse temperature
$\gamma$	damping kernel
$\kappa$	FDR kernel
$\mu$	dissipation kernel
$\nu$	damping kernel
$T$	temperature
CP	Completely Positive
DOF	Degrees of Freedom
EOM	Equation of Motion
ESD	Entanglement Sudden Death
FDI	Fluctuation-Dissipation Inequality
FDR	Fluctuation-Dissipation Relation
FO	Ford and O'Connell
HPZ	Hu, Paz and Zhang
HUP	Heisenberg Uncertainty Principle
IR	Infrared
KMS	Kubo, Martin and Schwinger
ODE	Ordinary Differential Equation
PDE	Partial Differential Equation
QBM	Quantum Brownian Motion
QRT	Quantum Regression Theorem
RDM	Reduced Density Matrix
UV	Ultraviolet

# Chapter 1

## Introduction

An *open quantum system* refers to a quantum system that interacts with some environment whose degrees of freedom have been coarse grained away (i.e. traced or averaged over). Open quantum systems provide a microscopic theory suitable for the investigation of the properties and dynamics of non-equilibrium quantum systems in the Langevin perspective. The open system perceives the coarse-grained environment as a source of fluctuations and dissipation, both of which have a time correlation or memory kernel. Mathematically the environmental influence can be described by a hierarchy of  $N$ -time correlation functions, or influence kernels, which may be perturbatively truncated if necessary. This is in contrast to the Boltzmann perspective, which considers large (closed) systems as a hierarchy of  $N$ -particle distribution functions or  $N$ -point correlation functions, which may then be perturbatively truncated and coarse grained (see, e.g., Ref. [31]).

Non-equilibrium descriptions of quantum mechanics offer more physical content than statistical mechanics, wherein the system remains in equilibrium through interaction with a reservoir. Non-equilibrium dynamics include dissipation, decoherence, disentanglement, the displacement from equilibrium and the relaxation towards equilibrium. Key among non-equilibrium descriptions of quantum mechanics are those driven by *Markovian* (white noise) processes. In the Langevin description,

the Markovian regime corresponds to the limit in which all relevant environment correlation timescales are much shorter than all relevant system timescales. Such dynamics are fully characterized by Lindblad equations (as opposed to Liouville-von Neumann-Schrödinger equations for closed systems) and can be motivated phenomenologically. Non-Markovian dynamics are not so easily characterized, but are required to fully incorporate the effects of finite temperature and spatial correlations present in the environment. A microscopic derivation of the open system consistently accounts for backreaction with the environment and will always produce a non-Markovian theory which is quantum-mechanically valid (i.e. completely positive).

Quantum open systems play an important role in addressing the fundamental issues such as the quantum-to-classical transition through the environment-induced decoherence mechanism [74, 164]. For practical purposes they have been effectively applied to exciting phenomena in many new directions of micro and mesoscopic physics in the last two decades, made possible by innovative experiments aided by technological advances in high-precision instrumentation. These include the areas of superconductivity such as quantum dissipative tunneling in SQUIDs [28, 104, 151], atomic and quantum optical systems using ultrafast lasers with atoms in cavities and optical lattices [138, 113, 153], as well as nanoelectromechanical devices [119, 102] which have great potential in physical, chemical and bioscience applications. For an accurate description of the system's properties and evolution in these processes, the effects of its interaction with the environment are essential.

Throughout the paper we use units with  $c = \hbar = k_B = 1$ , except when we

explicitly restore these constants for discussion.

## 1.1 Outline of Thesis

In Chapter 2 we report our perturbative solutions to the general problem of quantum open systems [63], with no requirement of linearity for the system, environment, or interaction, but wherein we consider the system-environment interaction perturbatively. We primarily focus upon time-constant Hamiltonians, though our formalism is general and some details will be specified for periodic and piecewise time-dependent Hamiltonians. Essentially, our solutions are a translation of canonical perturbation theory from the closed-system Schrödinger equation to the open-system Liouville equation. Some subtleties arise as the Liouville equation is non-Hermitian for open systems, unavoidably degenerate for all systems, and rarely known exactly. In particular, unavoidable degeneracy leads to an unfortunate loss of accuracy between perturbative master equations, which may be specified to particular order, and their resulting solutions, which can then only be accurate to a lesser perturbative order.

Markovian processes result in a master equation of a form described by the semi-group theorem of Lindblad [106] and Gorini, Kossakowski and Sudarshan [76]. This form of master equation ensures completely-positive evolution, which is a necessary ingredient for generally valid quantum theories. But for non-Markovian processes (e.g. finite temperature), the resultant master equation is not directly characterized by the Lindblad-GKS theorem. Yet if a master equation is microscopically

derived, it must necessarily result in completely-positive evolution. Another important result of our work in [63] was in determining a perturbative formulation of the Lindblad-GKS theorem for master equations which describe non-Markovian dynamics and do not take the Lindblad form.

In Chapter 3 we report our exact solutions to the problem of *quantum Brownian motion* [64, 66], which considers quantum Brownian oscillators bilinearly coupled (position-position) to an environment of quantum oscillators, so that the interacting system and environment is linear and thus tractable. This naturally provides a quantum mechanically valid model of a damped oscillator. Unlike in classical mechanics, one cannot simply write down a Newtonian Langevin equation in the quantum regime. Such a naive equation may not preserve positivity or even the two-time commutation relations of the system operators. By introducing a compact and particularly well-suited formulation, we give a rather quick and direct derivation of the master equation and its solutions for general spectral-density functions and finite temperatures. The flexibility of our approach allows cases with an arbitrary number of Brownian oscillators and external forces. Previous master equations were essentially limited to a single oscillator. In previous calculations there was also some confusion between the implications of the time-local and nonlocal representations, and thus diffusion coefficients (and often force response) have been incorrectly specified in the literature. Furthermore, we provide explicit, exact analytical results for the master equation coefficients and its solutions in a wide variety of cases, including ohmic, sub-ohmic and supra-ohmic environments with a finite cut-off.

In Chapter 4 we present a number of more generally applicable theoretical

results which draw heavily from our work in Chapters 2 and 3. We start in Sec. 4.2 with a more thorough explanation of the accuracy loss inherent to the solutions of all perturbative master equations. In Sec. 4.3 we detail how the general formalism of time-dependent master equations is capable of handling the preparation of properly correlated initial states without abandoning the linear formalism. This simultaneously serves two purposes: to avoid the affine master equation formalism and to better select the most appropriate correlated initial states of the system and environment. In Sec. 4.4 we study the widely used *rotating-wave approximation* (RWA) in its application to open-system dynamics. Using the general formalism of Chapter 2 we are able to precisely determine what information is preserved and discarded in this family of approximations. In Sec. 4.5 we detail the reduced equilibrium states of open systems, given environments initially in equilibrium. In Sec. 4.6 we discuss our newly discovered *fluctuation-dissipation inequality*, which applies to non-equilibrium environment correlations and relates to the Heisenberg uncertainty principle. Finally in Sec. 4.7 we apply the non-Markovian Lindblad-GKS generators to characterize the *decoherence strength* of non-equilibrium environments, in much the same way that temperature and resistance characterize a Markovian environment.

In Chapter 5 we report some newfound results pertaining to the strong-coupling dynamics of open systems (quantum or classical) of continuous variables. This includes the dissipative master equations of Brownian particles for which the system potential energy is taken to be small as compared to system-environment potential. Our strong-coupling master equations are very different from other so-called “strong-coupling” master equations which are perturbing off a limit in which the

entire system energy (including kinetic energy) is taken to be perturbative and thus the dynamics is principally Markovian. Such approximations also require the system mass to be asymptotically large (even as compared to the ratio of noise and induced system frequencies) and thus they do not fully categorize the regime of what one might consider to be strong coupling. Our relations describe highly non-Markovian dynamics at all orders and are radically different for different system potentials, admitting no apparent generic form. This result is quite exciting as it brings forth a new regime for theoretical exploration: the regime of strong noise and dissipation yet non-Markovian, such as strong coupling to a low-temperature quantum environment with large  $1/f$  fluctuations.

In Chapter 6 we apply our formalisms to the consideration of multiple atoms held fixed in a common quantum field. QBM translates quite naturally to local oscillators residing in a scalar field, whereas we apply the perturbative master equation formalism to the problem of two-level atoms residing in the electromagnetic field. In each case the quantum field contains intricate space-time correlations related to its phonon/photon-mediate structure. These spatial correlations play a role in enforcing causal behavior as well as in producing different emission and disentanglement rates for different global states of the combined system. Most interesting is the disentanglement which occurs at zero temperature, wherein the system relaxes perturbatively close to its ground state, which is a pure state and exists on the boundary between separable and entangled states. Our more accurate solutions were necessary to assess sudden death of entanglement at zero temperature, as the asymptotic state is not necessarily separable for non-vanishing interaction with

the environment. In contrast to previous claims, we found that all initial states of two-level atoms undergo finite-time disentanglement.

Finally, in Chapter 7 we revisit the infamous Abraham-Lorentz force, wherein we consider a single (moving) nonrelativistic particle in the electromagnetic field, and we contrast this effect to the damped oscillator of QBM. While in motion, the particle experiences backreaction from the electromagnetic field, i.e. the moving particle generates a dynamic electromagnetic field which then reacts back upon the particle. Though physical, when naively considered this force leads to a number of well-known problems including acausality, in the form of pre-acceleration, and runaway solutions. We derive from first principles the time-local, causal and runaway-free stochastic equations of motion of a nonrelativistic charged particle in the electromagnetic field. Our equations of motion are found to be equivalent to the structureless Langevin equation of Ford & O'Connell (FO), though more directly derived from a microscopic theory. These equations are not more accurate than the FO equations, however they provide a very different microscopic perspective wherein the bare mass of the system remains positive for arbitrarily large cutoff. In our treatment the corresponding radiative force (more accurately, backreaction) is taken to be proportional to  $\dot{\mathbf{p}}$  and not  $\ddot{\mathbf{x}}$ ; these two variables are inequivalent non-perturbatively. The resulting stochastic equations of motion for the particle are then shown to be non-perturbatively dissipative and causal.

We conclude this work in Chapter 8 with a summary of findings and and discussion of future avenues. In the remainder of this introductory chapter, we review material common to Chapters 2-4 and most all open-system formalisms. We

begin with some preliminary information on open systems pertaining to the mathematical structure of non-unitary (dissipative) evolution. We carefully note the distinction between the (instantaneous) dynamical semi-group and (all-time) algebraic semi-group when employing the Lindblad-GKS theorem. We additionally wish to distinguish among the various usages of the term *Markovian* with regard to their distinct properties. We place the most emphasis on distinguishing the Markovian *representation*, which can be rather superficial, and the Markovian *process*, which is much more important.

## 1.2 Quantum Open Systems

Our microscopic theory consists of a system ‘S’ and environment ‘E’ driven by some combined ‘C’ Hamiltonian

$$\mathbf{H}_C \equiv \underbrace{\mathbf{H}_S + \mathbf{H}_E}_{\mathbf{H}_F} + \mathbf{H}_I + \mathbf{H}_R, \quad (1.1)$$

where the system and environment interact ‘I’ via  $\mathbf{H}_I$ . A suitable renormalization  $\mathbf{H}_R$  is included for several reasons (see Sec. 2.5.2.1), key among them is to match the open-system dynamics to the free ‘F’ system dynamics. Otherwise our notion of “system” becomes less physically motivated.

The most common choice of interaction and environment consists of linear coupling to a bath of harmonic oscillators, e.g. coupling to an otherwise free quantum field bilinearly with its field operator. This provides a general model of Gaussian noise [57] and is most tractable in the influence-functional (path-integral) [57] and quantum Langevin equation formalisms [67]. For weak coupling to the environment,

it will also be seen that Gaussian noise is a sufficient model for general environments (in particular, see Sec. 2.2.1.2).

We consider the environment to be comprised of an uncountable number of degrees of freedom, as to give rise to irreversible dynamics in the open system. For mathematical simplicity, we assume the initial state of the system and environment to be factorized

$$\rho_C(0) = \rho_S(0) \otimes \rho_E(0), \quad (1.2)$$

or a product of marginal distributions in the phase-space representation. This will give rise to certain pathological transient behaviors which we shall discuss more thoroughly in Sec. 4.3.

For all future times we consider the open system, such as its reduced density matrix

$$\rho(t) = \text{Tr}_E[\rho_C(t)] \quad (1.3)$$

which can generate all single-time correlation functions (but not all multi-time correlation functions in the non-Markovian regime, Sec. 2.4).

We are most interested in initially equilibrated states of the environment

$$\rho_E(0) = \frac{e^{-\beta \mathbf{H}_E}}{\text{Tr}_E[e^{-\beta \mathbf{H}_E}]}, \quad (1.4)$$

in order to consider non-equilibrium thermodynamics. Given the vast number of environment degrees of freedom, the open system should relax to global equilibrium

$$\lim_{t \rightarrow \infty} \rho(t) = \frac{\text{Tr}_E[e^{-\beta \mathbf{H}_C}]}{\text{Tr}_C[e^{-\beta \mathbf{H}_C}]}, \quad (1.5)$$

and this has been proven to second order (see Sec. 4.5). It is often a mistaken assumption that the open system will relax to its free equilibrium state either exactly

or to second order (though this does happen in certain special cases [73]). Some of this mistaken assumption is likely due to intuition drawn from the classical theory where for Gaussian noise the free and global equilibrium state of the open system are equivalent (due to commutivity).

### 1.2.1 Closed and Open-System Evolution

Closed-system evolution is extremely well understood in both classical and quantum mechanics. In quantum mechanics our pure states are unit vectors  $\boldsymbol{\psi}$  in Hilbert space  $\mathcal{H}$  equipped with an inner product  $\langle \psi_1, \psi_2 \rangle = \boldsymbol{\psi}_1^\dagger \boldsymbol{\psi}_2$ . Mixed states are trace unity and positive-definite operators  $\boldsymbol{\rho}$  in  $\mathcal{H} \otimes \mathcal{H}$  and can be expressed as a convex combination of orthogonal pure states or  $\sum_k p_k \boldsymbol{\psi}_k \boldsymbol{\psi}_k^\dagger$ . There are a number of measures to calculate the state overlap of mixed states, one of the better choices being the fidelity and its associated Bures distance and volume [141].

Closed-system evolution may then be described by unitary transformations, or isomorphisms between the initial and final Hilbert spaces. The algebraic generators of these unitary transformations  $\mathbf{G}$  (which form a Lie group) are characterized by anti-Hermitian operators  $-i\boldsymbol{\Theta}$ , where  $\boldsymbol{\Theta}$  is Hermitian.

$$\mathbf{G} : \boldsymbol{\psi} \rightarrow \mathbf{G} \boldsymbol{\psi} = e^{-i\boldsymbol{\Theta}} \boldsymbol{\psi}. \quad (1.6)$$

If these maps are time-translation invariant

$$\mathbf{G}(t) \mathbf{G}^{-1}(\tau) = \mathbf{G}(t-\tau), \quad (1.7)$$

then the algebraic generator can be directly identified with the time-translation

generator.

$$\frac{d}{dt}\mathbf{G}(t) = -i\mathbf{H}\mathbf{G}(t), \quad (1.8)$$

$$\mathbf{G}(t) = e^{-it\mathbf{H}}, \quad (1.9)$$

where  $\mathbf{H}$  is the Hamiltonian. Otherwise the time-translation generator is less directly related to the algebraic generator, though it retains the same anti-Hermitian structure. More specifically we have

$$\mathbf{H}(t) = \int_0^1 d\eta e^{-i\eta\Theta(t)} \dot{\Theta}(t) e^{+i\eta\Theta(t)}, \quad (1.10)$$

which is a sum of unitarily transformed Hermitian operators and is thus Hermitian. This correspondence between the mathematical characterization of the (all-time) algebraic and (instantaneous) dynamical generators also holds for classical dynamics, but not for open systems.

In classical mechanics we begin with a manifold  $M$  in which our system positions  $X$  live. Momentum  $P$  is then identified with cotangent space  $T_X M$ , and phase space  $(X, P)$  with the cotangent bundle  $T^*M$  or symplectic manifold (when equipped with a phase-space volume form). Pure states are described by phase-space points, whereas mixed states are described by probability distributions of said points. Just as the unitary evolution of quantum mechanics is described by isomorphisms between Hilbert spaces, the symplectomorphic evolution of classical mechanics is described by isomorphisms between symplectic manifolds. The generators of these maps are also Hamiltonian, though of course, in the classical sense.

The extension of this mathematical structure to open-system dynamics is denoted in Fig. 1.1. The characterization of such maps and generators is well known.

	<b>Quantum</b>	<b>Classical</b>	
<b>Closed</b>	Unitary	Symplectomorphic	<b>Group</b>
	Hamiltonian	Hamiltonian	<b>Algebra</b>
<b>Open</b>	CP: Choi	Stochastic Matrices	<b>Group</b>
	Lindblad-GKS	Quasi-Positive Diagonally-Dominant	<b>Algebra</b>

Figure 1.1: Closed-system versus open-system evolution in classical and quantum mechanics.

For classical mechanics one considers Markov chains, which are positive (P) maps between discrete state vectors. In quantum mechanics one must additionally consider completely-positive (CP) maps between states, as explained and described in Sec. 1.2.2. In the classical context, all positive maps are completely positive and therefore we may speak of complete positivity as the more universal property.

Open systems present a severe problem for phenomenologists who wish to explore the non-Markovian regime (non-Markovian will be defined more explicitly in Sec. 1.3). Consider classical or quantum mechanical maps valid for all states at  $t = 0$ .

$$\mathcal{G}(t) : \rho(0) \rightarrow \rho(t) = \mathcal{G}(t) \rho(0). \quad (1.11)$$

These are completely-positive maps and their algebraic generators  $\Phi(t)$ , as determined by  $\mathcal{G}(t) = e^{\Phi(t)}$ , satisfy well-known characterizations. However, the (instantaneous) dynamical generators are given by the difference between two completely-

positive generators (CP-CP).

$$\mathcal{G}(t + dt, t) = \mathcal{G}(t + dt) \mathcal{G}^{-1}(t), \quad (1.12)$$

$$= e^{\Phi(t+dt) - \Phi(t) + \mathcal{O}(dt^2)} \quad (1.13)$$

Therefore the dynamical generators (when applied to arbitrary states) can be non-completely positive if the induced diffusion, decoherence, and other irreversible processes do not proceed uniformly in time. A Venn diagram of these three categories of dynamical generators is given in Fig. 1.2. Unfortunately, the vast majority of CP-CP generators do not provide (all-time) completely-positive evolution and the characterization of non-Markovian CP generators requires time-integrated solutions. We give a perturbative characterization in Sec. 2.2.1.3.

Consider, for instance, a purely diffusive map  $\mathcal{G}_\tau$  illustrated in Fig. 1.3. The related anti-diffusive map  $\mathcal{G}_\tau^{-1}$  is not completely positive, in that there are states for which it would map their covariance to a negative quantity. However, anti-diffusion can proceed after a sufficient amount of diffusion, so that the entire map is completely positive for any initial state.

## 1.2.2 Complete Positivity

*Positive maps* (P) for a system S will translate the physical states of system S (positive-definite, trace-unity density matrices in the product Hilbert space of S) strictly into physical states. *Completely positive maps* (CP) for a system S will additionally provide positive maps for S+A, where A is an arbitrary ancillary system. Completely positive maps are therefore compatible with entanglement to additional

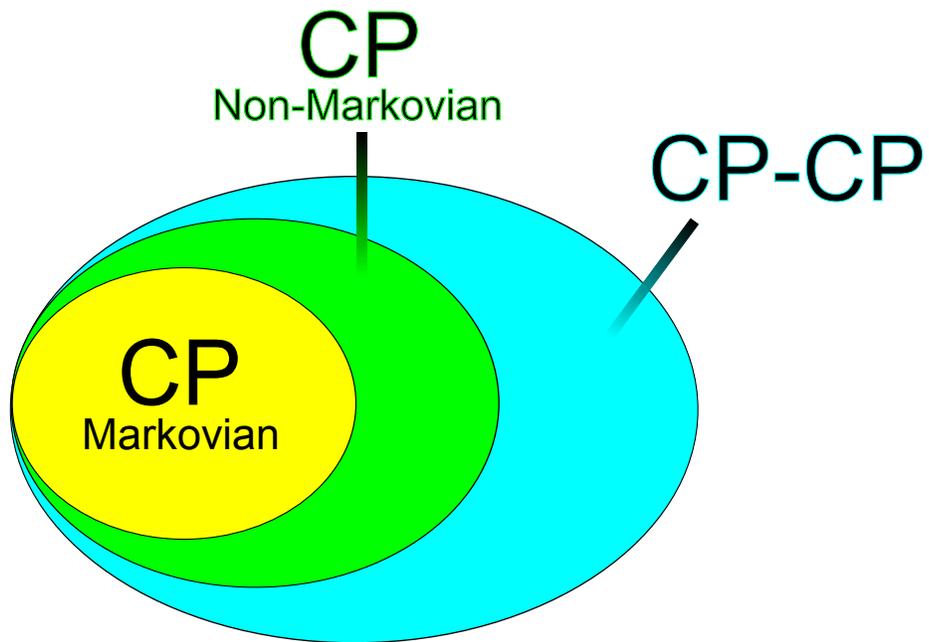


Figure 1.2: Classification of dynamical generators: Markovian completely-positive generators  $\subset$  non-Markovian completely-positive generators  $\subset$  the difference of two (Markovian) completely-positive generators.

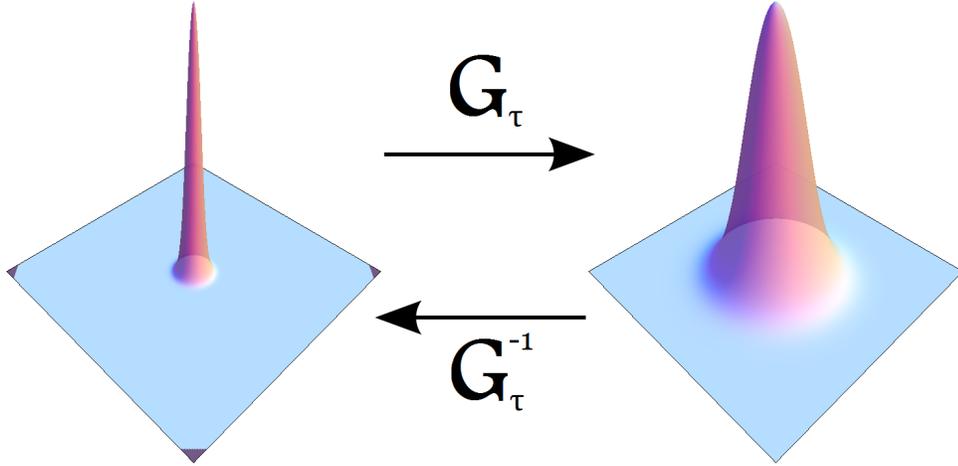


Figure 1.3: Diffusive and anti-diffusive maps.

degrees of freedom not included in the system. Any physical theory of quantum mechanics must take all input states and map them in a completely positive manner into the future. We will momentarily ignore the subtleties and disagreement [126, 139] with this commonly accepted wisdom. Also note that for classical distributions it is easy to prove that all positive maps are completely-positive maps, and so this is of no concern to classical mechanics.

Given an initial-time propagator  $\mathcal{G}(t) : \rho(0) \rightarrow \rho(t)$  which maps initial states into the future, and which can be represented via some basis in Hilbert space

$$\rho(t) = \mathcal{G}(t) \{ \rho(0) \} , \quad (1.14)$$

$$\mathcal{G}_{ij; i'j'}(t) = \langle i | \mathcal{G}(t) \{ |i'\rangle\langle j'| \} | j \rangle , \quad (1.15)$$

preservation of normalization and Hermiticity dictates the relations

$$\sum_i \mathcal{G}_{ii;i'j'}(t) = \delta_{i'j'}, \quad (1.16)$$

$$\mathcal{G}_{ij;i'j'}(t) = \mathcal{G}_{j'i;j'j}^*(t). \quad (1.17)$$

while non-complete positivity requires the positivity of all bi-quadratic forms

$$\langle \psi | \mathcal{G}(t) \{ |\phi\rangle\langle\phi| \} | \psi \rangle \geq 0, \quad (1.18)$$

$$(\psi_i \phi_{i'}^*)^* \mathcal{G}_{ij;i'j'}(t) (\psi_j \phi_{j'}^*) \geq 0, \quad (1.19)$$

where  $\psi$  and  $\phi$  are Hilbert space vectors. Determination of non-complete positivity is an NP-Hard problem [108]. *Choi's theorem* [35], proves that completely-positive maps are determined by the positivity of all quadratic forms

$$\Psi_{ii'}^* \mathcal{G}_{ij;i'j'}(t) \Psi_{jj'} \geq 0, \quad (1.20)$$

which is (formally) a simple matter of linear algebra to resolve. Note that the propagator's indices are interpreted differently from when ordinarily used as a transition matrix.

$$|i'\rangle\langle j'| \rightarrow |i\rangle\langle j| \quad (\text{Transition Matrix}), \quad (1.21)$$

$$|j\rangle\langle j'| \rightarrow |i'\rangle\langle i| \quad (\text{Choi Matrix}). \quad (1.22)$$

The eigen-value decomposition of the positive-definite *Choi matrix* is known as the *Kraus representation* [98].

Just as Choi's theorem characterizes the Lie group elements  $\mathcal{G}(t)$  of all valid quantum maps, the theorem due to Lindblad [106] and Gorini, Kossakowski and

Sudarshan [76], in its most general application, characterizes the associated Lie algebra. Specifically, given the exponential representation

$$\mathcal{G}(t) = e^{\Phi(t)}, \quad (1.23)$$

then with the assumption of a semi-group structure, i.e.  $e^{\eta\Phi(t)}$  is also a group element for  $\eta > 0$ , the algebraic generator must be of *Lindblad form*:

$$\Phi \rho = \underbrace{-i[\Theta, \rho]}_{\text{unitary}} + \underbrace{\sum_{ij} \Delta_{ij} \left( \mathbf{e}_i \rho \mathbf{e}_j^\dagger - \frac{1}{2} \{ \mathbf{e}_j^\dagger \mathbf{e}_i, \rho \} \right)}_{\text{decoherent}}, \quad (1.24)$$

where  $\Theta$  is a Hermitian operator and  $\Delta$  is a Hermitian and positive-definite coefficient matrix, with  $\mathbf{e}_i$  a particular basis of representation. Such algebraic generators and the dynamics arising when the Liouvillian appearing in the master equation has Lindblad form have been extensively studied [97, 45, 46, 48, 6, 1, 14, 90, 107, 151]. Due to non-commutativity, the time-local master equation is in general not directly determined by  $\dot{\Phi}(t)$ .

$$\dot{\mathcal{G}}(t) = \mathcal{L}(t) \mathcal{G}(t), \quad (1.25)$$

$$\mathcal{L}(t) = \int_0^1 d\eta e^{+\eta\Phi(t)} \dot{\Phi}(t) e^{-\eta\Phi(t)}, \quad (1.26)$$

and therefore not all valid master equations must be of (time-dependent) Lindblad form

$$\frac{d}{dt} \rho = \mathcal{L} \rho = -i[\mathbf{H} + \mathbf{V}, \rho] + \sum_{ij} \mathcal{D}_{ij} \left( \mathbf{e}_i \rho \mathbf{e}_j^\dagger - \frac{1}{2} \{ \mathbf{e}_j^\dagger \mathbf{e}_i, \rho \} \right), \quad (1.27)$$

where the Hermitian operator  $\mathbf{V}$  arises with the existence of the non-unitary  $\mathcal{D}$  and therefore  $\mathbf{H} + \mathbf{V}$  should not be identified with the system Hamiltonian. This

class inequivalence between the *time-translation generators*  $\mathcal{L}(t)$  and the *algebraic generators*  $\Phi(t)$  does not exist for unitary transformations, where both share the same adjoint symmetry. However, following the generalization of Choi’s theorem on merely Hermitian-preserving maps, one can prove that any Hermitian and trace-preserving time-local master equation must have a *pseudo-Lindblad form*  $\mathcal{L}(t)$  with merely Hermitian  $\mathcal{D}$ . The super-operator  $\mathcal{D}$  is generally referred to as the *dissipator*. The dissipation generated by the dissipator is that of states and not of energy; its signature distinguishes between decoherent and recoherent evolution. The dissipator and unitary generator  $\mathbf{V}$  together contain both ordinary dissipation and diffusion. In general,  $\mathbf{V}$  should not be considered a renormalization of  $\mathbf{H}$ .

The “state dissipation” generated by the dissipator can be given a more precise geometrical meaning. For any distance  $D$  on the space of density operators which is constructed from a *monotonic* metric (e.g. trace distance or Bures distance), then CP evolution cannot cause any such distances to expand [128].

$$D[\mathcal{G}(t) \rho_1, \mathcal{G}(t) \rho_2] \leq D[\rho_1, \rho_2]. \quad (1.28)$$

From this result it is easy to prove that positive-definite dissipators contract the state-space volume, whereas negative-definite dissipators expand the state-space volume (as they appear to be time-reversed contractions). This is very much analogous to how phase-space volume is contracted in dissipative classical mechanics.

Testing for the complete positivity of a time-dependent master equation involves a bit more effort than a determination of  $\mathcal{D}(t) > \mathbf{0}$  in the pseudo-Lindblad  $\mathcal{L}(t)$ . A Lindblad form master equation is sufficient but not necessary. In Sec. 2.2.1.3

we prove that all microscopically-derived second-order master equations are completely positive to second order. We show that the corresponding *weak test for complete positivity* in an arbitrary (time-local) master equation is that the pseudo-dissipator is not necessarily positive-definite for all times but for all running averages while in the interaction picture:

$$\int_0^t d\tau \underline{\mathcal{D}}(\tau) \geq \mathbf{0}. \quad (1.29)$$

A similar analysis exists to check for higher-order consistency.

### 1.3 Markovian versus Non-Markovian Dynamics

The environment acts as a source of random noise influencing the system. This noise has correlations (memory) which reflect the natural timescales of the environment, e.g. the temperature of a thermal reservoir. Such noise is said to be *colored* and the process *non-Markovian*. In the *Markovian limit* the timescales of the environment are taken to be much shorter than the timescales of the system. Such noise is said to be *white* and the process *Markovian*; its timescales cannot be resolved by the system. *Quantum noise* correlations are complex, containing both real noise and dissipation (see Sec. 2.5.2). Thus we say that for quantum noise, the Markovian limit corresponds to all such memories being of insignificant duration.

An open-system master equation is a linear map for the reduced density matrix

generated via

$$\frac{d}{dt}\boldsymbol{\rho} = \mathcal{L}\{\boldsymbol{\rho}\}, \quad (1.30)$$

$$\mathcal{L}_0\{\boldsymbol{\rho}\} = [-i\mathbf{H}, \boldsymbol{\rho}], \quad (1.31)$$

where, in our open system formalism,  $\mathbf{H}$  denotes the free system Hamiltonian and  $\mathcal{L}_0$  the free system Liouville operator. The master equation is said to have *time-local*, *time-convolutionless*, or *Markovian representation* if  $\mathcal{L}(t)$  does not reference the past history of states  $\boldsymbol{\rho}(\tau)$  for  $\tau < t$ . A Markovian representation does not necessarily imply Markovian dynamics. Non-Markovian processes can readily admit Markovian representations for their coarsegrained behavior, e.g. the exact QBM master equation of HPZ [84] discussed in Chapter 3. Moreover, for any non-Markovian master equation of the form

$$\frac{d}{dt}\boldsymbol{\rho}(t) = \int_0^t d\tau \mathcal{K}(t-\tau) \boldsymbol{\rho}(\tau), \quad (1.32)$$

with invertible propagator  $\mathcal{G}(t) : \boldsymbol{\rho}(0) \rightarrow \boldsymbol{\rho}(t)$ , there is a corresponding time-local master equation

$$\frac{d}{dt}\boldsymbol{\rho}(t) = \mathcal{L}(t) \boldsymbol{\rho}(t), \quad (1.33)$$

where  $\mathcal{L}(t) = \dot{\mathcal{G}}(t) \mathcal{G}^{-1}(t)$  trivially and in this case  $\mathcal{G}(t)$  and thus the master equation can be formally determined via Laplace transformation. Eq. (1.32) and (1.33) are fully equivalent and can both generate non-Markovian dynamics despite any Markovian representation; all memory effects are encoded in the full time dependence of the master equation coefficients themselves.

Given a time-local master equation, an  $\mathcal{L}$  constant in time is said to be *homogeneous* while an  $\mathcal{L}(t)$  variable in time is said to be *inhomogeneous*. Some authors in physics refer to this distinction as the Markovian/non-Markovian distinction as such processes can lead to this kind of evolution. However, as before, these properties are not equivalent and therefore we will avoid such terminology. For time-dependent master equations which asymptote into a homogeneous form, this is known as the *stationary limit* of the master equation. A well-defined stationary limit is also not specific to Markovian processes, e.g. the exact QBM master equation for a regulated Ohmic coupling, Sec. 3.7. Nor does a stationary limit imply Markovian dynamics therein, as can be seen by a damped quantum oscillator's response to forces, Sec. 3.4.4.

In the context of quantum open systems, Markovian processes generally result in *Lindblad equations*. Lindblad equations generate completely-positive evolution for all states at all times. However not all master equations which generate completely-positive evolution are of Lindblad form, and not all Lindblad equations arise from Markovian processes. The relationship between Markov, Lindblad, and complete positivity will be discussed more thoroughly in Sec. 1.2.2. A conflation of the stochastic process and representation often results in properties of Markovian dynamics being incorrectly applied to master equations of Markovian representation.

### 1.3.1 Quantum Regression Theorem

For a closed system, or open system driven by Markovian processes, all multi-time correlations of the system can be generated by master equation  $\mathcal{L}(t)$ , or more specifically its super-adjoint  $\mathcal{L}^\dagger(t)$ , Sec. 2.4.1. This is what is known as the *Quantum Regression Theorem* (QRT), e.g. for two-time correlations we have

$$\langle \dot{\mathbf{X}}_1(t_1) \mathbf{X}_2(t_2) \rangle = \langle \{ \mathcal{L}^\dagger(t_1) \mathbf{X}_1 \}(t_1) \{ \mathbf{X}_2 \}(t_2) \rangle. \quad (1.34)$$

But for a non-Markovian process, the open-system master equation cannot generate all multi-time correlations despite whatever *representation* the master equation may have. This is, in fact, a defining characteristic of the Markovian process and has nothing to do with quantum mechanics specifically. Classically speaking, the Markov property can be defined in terms of probabilities of events or expectation values of observables. Extending this definition to the expectation values of quantum observables, the Markov property is rather synonymous with the QRT. Other definitions of the (quantum) Markov property may not maintain classical correspondence with what is well understood to be Markovian and non-Markovian.

The non-Markovian corrections to the QRT have been known for almost three decades [145]. This was first reported via the projection operator method and has recently been duplicated and expanded upon with stochastic Schrödinger-equation techniques [49]. These corrections do not vanish in the weak coupling regime, but strictly in the white-noise limit. In Sec. 2.4 we rederive the non-Markovian corrections to the QRT in a simple perturbative fashion and express them in a form which demonstrates their inherent non-Markovian character.

### 1.3.2 Piecemeal Master Equations

As a Markovian process is without memory, its response is somewhat system agnostic. This allows for the piecemeal construction of master equations. Let us consider the reduced density matrices of systems A and B coupled to identical dissipative environments. We then have the open-system master equations

$$\frac{d}{dt}\rho_A = -\imath[\mathbf{H}_A, \rho_A] + \delta\mathcal{L}_A\{\rho_A\}, \quad (1.35)$$

$$\frac{d}{dt}\rho_B = -\imath[\mathbf{H}_B, \rho_B] + \delta\mathcal{L}_B\{\rho_B\}, \quad (1.36)$$

where the Hamiltonians are those of the free systems and the corrections to the Liouvillian are introduced via interaction with the dissipative environment. In the Hamiltonian formalism one can simply add two Hamiltonians and arrive at another Hamiltonian, though one might be motivated to fix the energy spectrum through renormalization. One cannot do this with non-Markovian Liouvillians, e.g. given some subsystem coupling  $\mathbf{H}_{AB}$  one cannot simply add dissipative terms.

$$\frac{d}{dt}\rho_{A+B} \neq -\imath[\mathbf{H}_A + \mathbf{H}_B + \mathbf{H}_{AB}, \rho_{A+B}] + \delta\mathcal{L}_A\{\rho_{A+B}\} + \delta\mathcal{L}_B\{\rho_{A+B}\}, \quad (1.37)$$

The above (incorrect) master equation is in general completely different from the correct open-system master equation as derived from first principles. For non-Markovian processes, the environmental contributions have a nontrivial dependence (due to memory effects) upon the systems' dynamics through their couplings. If one changes the system Hamiltonian, then one must also change the environmental contributions to be compatible with the history these new terms will create. This is how memory exhibits itself in a time-local representation. Moreover, one must

also take into account whether or not the dissipative environments are separate or shared. If the dissipative environment is shared then the two subsystems can interact via environmental back-reaction, even in the Markovian regime. This effect is also missed when simply combining the Liouville operators.

In a general non-Markovian master equation, the invalidity of the above incorrect master equation would be readily apparent as it would likely violate positivity, uncertainty, etc. Positivity violation will not occur when adding Lindblad terms. But if one has a non-Markovian Lindblad equation, such as given by the rotating-wave approximation, then the mistake has only become more subtle and therefore more dangerous. The master equation might be completely positive, but it does not correspond to the dynamics of the physical system considered.

The above example of interacting systems coupled to an environment should not be surprising. If the environment is thermal, then to zeroth-order in the system-environment interaction, the systems should relax to a Boltzmann state which includes the system-system interaction Hamiltonian. Obviously this can only happen if the master equation coefficients are aware of the system-system interaction. However, this same mistake is often applied in more subtle ways. For instance, if the system is driven by an external force, then due to non-Markovian response the driving terms in the Hamiltonian theory and in the open-system master equation are generally inequivalent (see Sec. 3.4.4). This is to compensate for the difference between the actual nonlocal response of the system and the superficial time-local representation of the master equation.

This issue has also been commented on in the context of cavity QED. The

often-used master equation includes the Hamiltonian for the atom, intracavity field, and atom-field interaction, but the dissipator used is exactly that of an empty cavity with dissipation plus that of an atom spontaneously emitting into empty space, so that the situation is just that depicted in 1.37. And, indeed, if one begins instead with the atom-cavity system and derives the microscopic master equation using the standard technique [22], one finds that the master equation has a different dissipative term [137, 135]. As explained in [135] if the spectrum of environmental noise is sufficiently flat then the difference is suppressed, which explains the success of the standard cavity QED master equation. But not so otherwise, which is something often overlooked.

### 1.3.3 Physical Limitations of the Lindblad Master Equation

Any time-local master equation which preserves the trace and complete positivity for every time translation  $t_1 \rightarrow t_2$  given arbitrary state  $\rho(t_1)$  must be of Lindblad form. This is not exhaustive of all valid theories. Quantum open systems provide valid master equations which are very generally not of Lindblad form. In the simplest models of quantum open systems, one begins at some initial time  $t_0$  with an uncoupled and uncorrelated (factorized) system S and environment E. The system and environment are then coupled together via an interaction Hamiltonian and the environmental degrees of freedom are traced over to obtain the open-system dynamics of only S. It is a trivial matter to show that all initial system states  $\rho(t_0)$

will evolve in a completely positive manner.

$$\mathcal{G}(t, t_0) : \rho(t_0) \rightarrow \rho(t), \quad (1.38)$$

but the intermediate mappings

$$\mathcal{G}(t_2, t_1) = \mathcal{G}(t_2, t_0) \mathcal{G}(t_1, t_0)^{-1}, \quad (1.39)$$

are not necessarily completely positive. The total volume of possible physical states can shrink such that an arbitrary intermediate state  $\rho(t_1)$  can fall into the category of impossible physical states, or states which did not evolve from any physical state at  $t_0$ . Such a state can then evolve into the category of unphysical states and there is nothing fundamentally wrong with this; it is an irrelevant evolution which has nothing to do with any physical prediction of the theory. This is a typical feature of non-Markovian master equations; they can generate a less uniform decoherence than Lindblad equations.

In summary, completely-positive maps are much less useful outside of the Markovian regime, as one rarely has the all-time maps. “All-time” here meaning that the maps must describe all times wherein there was any correlation to anything. In the most pathological interpretation, such a map would have to describe the entire universe from its very birth. More typically, both mathematically and empirically, one only has information pertaining to the two-time maps of some limited set of states. These maps are not completely positive in the non-Markovian regime and not much is known about them in terms of mathematical characterization, therefore microscopically derived models are of the utmost importance.

## Chapter 2

### Perturbative Solution of Nonlinear Systems

#### 2.1 Overview

An open quantum system is a quantum system that interacts with some environment whose degrees of freedom have been coarse grained over (i.e., traced out). The dynamical environment provides a model of noise, dissipation, and decoherence far more general than Markovian (white noise) treatments which can be argued on purely phenomenological grounds. Specifically for the open state and corresponding single-time correlations, the open-system dynamics are described by a master equation governing the reduced density matrix  $\rho$ . Exact master equations for the stochastic dynamics of open quantum systems are, in general, out of reach. However, arbitrary-order perturbative master equations (in the system-environment interaction) can be derived in a variety of different ways [93, 20, 144] and find application in many branches of physics and chemistry [129, 32, 22, 94]. Multi-time correlations add an extra layer of complication in the non-Markovian regime (e.g. finite temperature, cutoff, etc.) as we shall discuss.

In the following chapters we report our formalism for quantum open systems which closely mirrors the more well-known canonical perturbation theory applied to Schrödinger's equation. In expanding upon this formalism, much of this work consist of review material, however a fair number of these results are novel and

require a thorough buildup of foundational material to derive and explain.

In Sec. 2.2 we develop the perturbative time-local master equation, which is fairly well-known in the literature, and focus primarily upon the second-order master equation. It is often suspected that these perturbative master equations cannot be employed for significant lengths of time, but we find this to be more specifically determined by the noise distribution. We also explicitly demonstrate how one can test for the complete positivity of a non-Markovian master equation which is not of Lindblad form. The microscopically derived master equation indeed passes this test at second order. Perturbative solutions are detailed for the asymptotically stationary and cyclo-stationary (periodic) master equation. Later in Sec. 2.3 we derive the dual time-nonlocal master equation and determine it to be equivalent, at least asymptotically.

In Sec. 2.4 we rederive the perturbative non-Markovian Quantum Regression Theorem (QRT) corrections in simple manner and regard their structure more carefully. It is shown that, even at late times when the master equation has settled down into its stationary limit, the dynamics of the system remain non-Markovian as they should.

In Sec. 2.5 we give some categorization of the fundamental object of all second-order open-system dynamics, regardless of formalism: the environmental correlation function. Highlighted are the decomposition into fluctuations and dissipation, and their relation. Some passing mention is given to the correlation function's important role in categorizing *decoherence strength* and our newly discovered *fluctuation-dissipation inequality*, both of which will be covered more thoroughly in the next

chapter. Finally in Sec. 2.5.3 we discuss thermal reservoirs. The correspondences between the fluctuation-dissipation, KMS relations, Boltzmann distribution, and detailed balance are drawn.

## 2.2 Time-Local Master Equations

### 2.2.1 Perturbative Master Equations

We would like to consider a fairly general microscopic model of quantum dissipation: that of a quantum system, environment, and system-environment interaction all separable in the Hamiltonian.

$$\frac{d}{dt}\boldsymbol{\rho}_C(t) = \mathcal{L}_C(t)\boldsymbol{\rho}_C(t) = [-i\mathbf{H}_C(t), \boldsymbol{\rho}_C(t)] , \quad (2.1)$$

$$\mathbf{H}_C(t) = \mathbf{H}_F(t) + \mathbf{H}_I(t) , \quad (2.2)$$

where C subscript quantities refer to the (combined) closed system + environment, F subscript quantities refer to the free system + environment without interaction, and I subscript quantities refer to the system-bath interaction. S and E subscript quantities will refer to system and environment quantities respectively; ambiguous quantities will always refer to the system as the vast majority of this work is with regards to the open system. We will now proceed with a derivation of the open-system dynamics, under the assumption that we may treat the effect of the bath upon the system perturbatively. At least initially we will take the initial state of the system + bath to be uncorrelated,  $\boldsymbol{\rho}_C(0) = \boldsymbol{\rho}_S(0) \otimes \boldsymbol{\rho}_E(0)$ , and uncoupled for all previous times. Without abandoning the linear master-equation formalism, the

existence of initial-time system-environment correlations are considered in Sec. 4.3.

Rotating Eq. (2.1) from the Schrödinger picture to the interaction (Dirac) picture, the equivalent integral equation of motion is

$$\underline{\mathcal{G}}_{\text{C}}(t) = \mathbf{1} + \int_0^t d\tau \underline{\mathcal{L}}_{\text{I}}(\tau) \underline{\mathcal{G}}_{\text{C}}(\tau), \quad (2.3)$$

where the interaction-picture propagator and super-operators are defined

$$\mathcal{G}_{\text{C}}(t) = \mathcal{G}_{\text{F}}(t) \underline{\mathcal{G}}_{\text{C}}(t), \quad (2.4)$$

$$\mathcal{L}_{\text{I}}(t) = \mathcal{G}_{\text{F}}(t) \underline{\mathcal{L}}_{\text{I}}(t) \mathcal{G}_{\text{F}}^{-1}(t). \quad (2.5)$$

This integral equation is directly amenable to perturbation theory via a Neumann series. Tracing over the bath we then have

$$\underline{\mathcal{G}}(t) = \mathbf{1} + \int_0^t d\tau_1 \langle \underline{\mathcal{L}}_{\text{I}}(\tau_1) \rangle_{\text{E}} + \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \langle \underline{\mathcal{L}}_{\text{I}}(\tau_1) \underline{\mathcal{L}}_{\text{I}}(\tau_2) \rangle_{\text{E}} + \dots, \quad (2.6)$$

for the open-system propagator. Note that this perturbative series has secular behavior and is of little direct use. But given some level of approximation to the propagator, we can then extract an approximate open-system Liouvillian via the relation  $\mathcal{L}(t) = \dot{\mathcal{G}}(t) \mathcal{G}^{-1}(t)$  or equivalently

$$\mathcal{L}(t) = \mathcal{L}_0(t) + \mathcal{G}_0(t) \dot{\underline{\mathcal{G}}}(t) \underline{\mathcal{G}}^{-1}(t) \mathcal{G}_0^{-1}(t). \quad (2.7)$$

Assuming the odd moments of the bath vanish, which is always true for Gaussian noise, we only require the perturbative expansions

$$\dot{\underline{\mathcal{G}}}(t) = \dot{\underline{\mathcal{G}}}_2(t) + \dot{\underline{\mathcal{G}}}_4(t) + \dots, \quad (2.8)$$

$$\underline{\mathcal{G}}^{-1}(t) = \mathbf{1} - \underline{\mathcal{G}}_2(t) + \dots, \quad (2.9)$$

where the inverse propagator can also be generated via Neumann series, though it will involve a double summation. Ordinary perturbation in the coupling will then yield the following results expressed succinctly in the interaction picture:

$$\underline{\mathcal{L}}_2(t) = \int_0^t d\tau \langle \underline{\mathcal{L}}_1(t) \underline{\mathcal{L}}_1(\tau) \rangle_{\text{E}} , \quad (2.10)$$

$$\underline{\mathcal{L}}_4(t) = \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 \langle \underline{\mathcal{L}}_1(t) \underline{\mathcal{L}}_1(\tau_1) \underline{\mathcal{L}}_1(\tau_2) \underline{\mathcal{L}}_1(\tau_3) \rangle_{\text{E}} - \underline{\mathcal{L}}_2(t) \int_0^t d\tau \underline{\mathcal{L}}_2(\tau) , \quad (2.11)$$

and for stationary reservoirs the Liouville operators can be easily expressed in the Schrödinger picture as

$$\mathcal{L}_2(t) = \int_0^t d\tau \langle \mathcal{L}_1(t) \underline{\mathcal{L}}_1(\tau, t) \rangle_{\text{E}} , \quad (2.12)$$

$$\mathcal{L}_4(t) = \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 \langle \mathcal{L}_1(t) \underline{\mathcal{L}}_1(\tau_1, t) \underline{\mathcal{L}}_1(\tau_2, t) \underline{\mathcal{L}}_1(\tau_3, t) \rangle_{\text{E}} - \mathcal{L}_2(t) \int_0^t d\tau \underline{\mathcal{L}}_2(\tau, t) , \quad (2.13)$$

where two-time interaction-picture operators are given by

$$\underline{\mathcal{L}}(t_1, t_2) \equiv \mathcal{G}_{\text{F}}(t_1, t_2)^{-1} \mathcal{L}(t_1) \mathcal{G}_{\text{F}}(t_1, t_2) , \quad (2.14)$$

and the two-time propagator is given by

$$\mathcal{G}(t_1, t_2) = \mathcal{G}(t_1) \mathcal{G}^{-1}(t_2) . \quad (2.15)$$

Equivalent perturbative expansions of the open-system master equation have been derived by projection operator techniques [93], series expansion of the influence functional [20], and coherent unraveling of the influence functional [144].

### 2.2.1.1 The Second-Order Master Equation

To specify the second-order master equation in Eq. (2.10), we will first expand the interaction Hamiltonian into a sum of separable couplings.

$$\mathbf{H}_I(t) = \sum_n \mathbf{L}_n(t) \otimes \mathbf{l}_n(t), \quad (2.16)$$

with Hermitian system coupling variables  $\mathbf{L}_n(t)$  and collective environment coupling variables  $\mathbf{l}_n(t)$ . The environmental variables should be assumed to be completely non-stationary, in that they are off-diagonal in the free energy basis of the environment. Any stationary environmental coupling would commute with the free bath Hamiltonian and could be effectively absorbed into the free system Hamiltonian at this order. The second-order master equation can be expressed

$$\mathcal{L}_2 \rho = \sum_{nm} [\mathbf{L}_n, \rho (\mathbf{A}_{nm} \diamond \mathbf{L}_m)^\dagger - (\mathbf{A}_{nm} \diamond \mathbf{L}_m) \rho], \quad (2.17)$$

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

$$(\mathbf{A}_{nm} \diamond \mathbf{L}_m)(t) \equiv \int_0^t d\tau \alpha_{nm}(t, \tau) \{ \mathcal{G}_0(t, \tau) \mathbf{L}_m(\tau) \}, \quad (2.18)$$

in terms of the (multivariate) environmental correlation function

$$\alpha_{nm}(t, \tau) \equiv \langle \mathbf{l}_n(t) \mathbf{l}_m(\tau) \rangle_E, \quad (2.19)$$

which will be discussed more thoroughly in Sec. 2.5. For now simply note that the correlation function is Hermitian and positive definite. The pseudo-Lindblad coefficients of this master equation can then be expressed

$$\mathbf{V} \equiv \frac{1}{2i} \sum_{nm} [\mathbf{L}_n (\mathbf{A}_{nm} \diamond \mathbf{L}_m) - (\mathbf{A}_{nm} \diamond \mathbf{L}_m)^\dagger \mathbf{L}_n], \quad (2.20)$$

for the unitary generator and for the dissipator

$$\mathcal{D}_{ii':jj'} = \sum_{nm} \left\{ \langle i | \mathbf{L}_n | i' \rangle \overline{\langle j | (\mathbf{A}_{nm} \diamond \mathbf{L}_m) | j' \rangle} + \langle i | (\mathbf{A}_{nm} \diamond \mathbf{L}_m) | i' \rangle \overline{\langle j | \mathbf{L}_n | j' \rangle} \right\}, \quad (2.21)$$

where the dissipator has been evaluated in some basis  $\mathbf{e}_{ii'} = |i\rangle\langle i'|$  given representation (1.27). Though the second-order master equation is not of Lindblad form, in Sec. 2.2.1.3 we prove that it generates dynamics which are completely positive to second order.

### 2.2.1.2 Validity of the Late-Time Limit

The second-order Liouvillian  $\mathcal{L}_2(t)$  can have a well defined late-time limit for many reservoirs. But because of the convergence of  $\mathcal{L}_2(t)$ , this necessarily implies that the  $\mathcal{L}_2^2$  contribution to  $\mathcal{L}_4(t)$  in Eq. (2.11) has the potential to give rise to an  $\mathcal{O}(t)$  secular term. Even assuming an asymptotic limit for the lower-order perturbative master-equation coefficients, secularly-evolving higher-order terms could invalidate taking the late-time limit after the perturbative expansion. We will argue that the second-order master equation is exempt from worry, and that higher-order perturbative master equations can also be acceptable if (1) the environment is Gaussian (in its cumulants) to that order of perturbation and (2) the correlation function is sufficiently localized.

First we must express the fourth-order Liouvillian strictly in terms of time-

ordered integrals.

$$\begin{aligned}
\underline{\mathcal{L}}_4(t) &= \int_0^t d\tau_1 \int_0^{\tau_1} d\tau_2 \int_0^{\tau_2} d\tau_3 [\langle \underline{\mathcal{L}}_I(t) \underline{\mathcal{L}}_I(\tau_1) \underline{\mathcal{L}}_I(\tau_2) \underline{\mathcal{L}}_I(\tau_3) \rangle_E \\
&\quad - \langle \underline{\mathcal{L}}_I(t) \underline{\mathcal{L}}_I(\tau_1) \rangle_E \langle \underline{\mathcal{L}}_I(\tau_2) \underline{\mathcal{L}}_I(\tau_3) \rangle_E - \langle \underline{\mathcal{L}}_I(t) \underline{\mathcal{L}}_I(\tau_2) \rangle_E \langle \underline{\mathcal{L}}_I(\tau_1) \underline{\mathcal{L}}_I(\tau_3) \rangle_E \\
&\quad - \langle \underline{\mathcal{L}}_I(t) \underline{\mathcal{L}}_I(\tau_3) \rangle_E \langle \underline{\mathcal{L}}_I(\tau_1) \underline{\mathcal{L}}_I(\tau_2) \rangle_E ] . \tag{2.22}
\end{aligned}$$

The previously discussed secular behavior would appear to be now located in the second term of this equation. In the two following terms, the arguments of the 2-time correlations are intertwined between correlations with respect to their order of integration. For a Gaussian environment, or an environment which is at least Gaussian to this order, the 4-time correlation can then be decomposed into a sum of 3 products of 2 2-time correlations. The result is difficult to express in standard notation because the super-operators do not commute. The integrand of the 4-time correlation becomes

$$\underbrace{\underline{\mathcal{L}}_I(t) \underline{\mathcal{L}}_I(\tau_1)} + \underbrace{\underline{\mathcal{L}}_I(\tau_2) \underline{\mathcal{L}}_I(\tau_3)} + \underbrace{\underline{\mathcal{L}}_I(t) \underline{\mathcal{L}}_I(\tau_1) \underline{\mathcal{L}}_I(\tau_2) \underline{\mathcal{L}}_I(\tau_3)} + \underbrace{\underline{\mathcal{L}}_I(t) \underline{\mathcal{L}}_I(\tau_1) \underline{\mathcal{L}}_I(\tau_2) \underline{\mathcal{L}}_I(\tau_3)} , \tag{2.23}$$

where the brackets denote the pairs of operators being traced over with respect to the environment, and we would refer the reader to Ref. [20] for a more thorough examination of arbitrary orders. Non-commutativity is not a problem with the first term of the decomposition, which precisely cancels the secular term in Eq. (2.22). This cancelation of secular terms will continue at higher orders of perturbation theory, as long as one can apply a Gaussian decomposition to the moments of the environment. Therefore Gaussian reservoirs, and possibly other reservoirs which ad-

mit analogous cumulant decomposition, can be effectively described by perturbative master equations even in the late-time limit.

The stability of Gaussian reservoirs bodes well for the second-order master equation even when the reservoir correlations are not exactly Gaussian. We can make a Gaussian approximation to the environmental influence [57] by truncating the influence phase to quadratic order. Because this perturbation is done in the influence phase as opposed to the influence functional, it should be good for long times as higher-order terms in the influence phase should be well controlled. This is essentially the saddle-point approximation for path integrals. From the Gaussian approximation we may then take a weak-coupling approximation, which as we have shown is also justified for long times, and we will arrive at the same second-order master equation that we would have gotten if we had never taken the Gaussian approximation. Therefore the second-order master equation can be safe in the late-time limit because it is also an effectively Gaussian approximation.

### 2.2.1.3 Complete Positivity

As explained in Sec. 1.2.2, application of the Lindblad-GKS theorem to test for complete positivity requires, not the time-translation generator, but the algebraic generator. Consider the Neumann series, Eq. (2.6), whose terms are given by

$$\underline{\mathcal{G}}_k(t) = \left\langle \prod_{i=1}^k \int_0^{\tau_{i-1}} d\tau_i \underline{\mathcal{L}}_I(\tau_i) \right\rangle_{\text{E}}, \quad (2.24)$$

where  $\tau_0 = t$ . This series can be contracted into the single exponential

$$\underline{\mathcal{G}}(t) = e^{\underline{\Phi}(t)}, \quad (2.25)$$

$$\mathcal{G}(t) = \mathcal{G}_0(t) e^{\underline{\Phi}(t)}, \quad (2.26)$$

where for symmetric noise, e.g. Gaussian, the perturbative generators can then be found to be

$$\underline{\Phi}_2(t) = \underline{\mathcal{G}}_2(t), \quad (2.27)$$

$$\underline{\Phi}_4(t) = \underline{\mathcal{G}}_4(t) - \frac{1}{2}\underline{\mathcal{G}}_2^2(t), \quad (2.28)$$

This is equivalent to solving the master equation via *Magnus series* [109] in the interaction picture. It should be noted that these Magnus-series solutions are slightly secular in time, as in general the Magnus series has a finite radius of convergence [19]. The second-order Magnus series will not asymptote to the second-order solution but to the corresponding RWA solution (if applicable), which lies between zeroth and second-order perturbation and is completely positive. These are not solutions that we would apply generally, and we will only be analyzing their semi-group structure.

The Magnus-series solution to the second-order master equation gives rise to the second-order algebraic generator

$$\underline{\Phi}(t) = \int_0^t d\tau \int_0^\tau d\tau' \langle \underline{\mathcal{L}}_I(\tau) \underline{\mathcal{L}}_I(\tau') \rangle_E + \mathcal{O}(\mathcal{L}_I^4), \quad (2.29)$$

in the interaction picture. In terms of the interaction Hamiltonian, the Lindblad coefficients of the algebraic generator  $\underline{\Phi}(t)$  are then

$$\underline{\Delta}_{i'i';jj'}(t) = \int_0^t d\tau \int_0^\tau d\tau' \langle \langle i | \underline{\mathbf{H}}_I(\tau) | i' \rangle \langle j' | \underline{\mathbf{H}}_I(\tau') | j \rangle \rangle_E, \quad (2.30)$$

given representation (1.27). With the interaction Hamiltonian expanded as a sum of separable operators, as per Eq. (2.16), the coefficients evaluate to

$$\Delta_{ii';jj'}(t) = \sum_{nm} \int_0^t d\tau \int_0^t d\tau' \langle i | \underline{\mathbf{L}}_m(\tau) | i' \rangle \alpha_{nm}(\tau', \tau) \overline{\langle j | \underline{\mathbf{L}}_n(\tau') | j' \rangle}, \quad (2.31)$$

in terms of the environmental correlation function. Both forms are positive-definite quadratic forms, therefore the second-order master equation must generate completely-positive maps to second order and the second-order Magnus-series solution  $\mathcal{G}(t) = \mathcal{G}_0(t) e^{\Phi_2(t)}$  happens to be *exactly* completely positive.

The algebraic generator for intermediate transitions is not generally of Lindblad form. For  $t_1 < t_2$ , the integration kernel for  $\Phi_2(t_1)$  in the quadratic form is effectively a leading principal minor of the corresponding integration kernel of  $\Phi_2(t_2)$ . Therefore, the intermediate transitions can only be ensured completely positive (for arbitrary states and couplings) given delta correlations or white noise. One can also have Lindblad master equations for specific system-environment couplings, such as the RWA-interaction, but this is a coupling dependent result.

Finally note that  $\Phi_2(t) = \int_0^t d\tau \mathcal{L}_2(\tau)$  and therefore the *weak test for complete positivity* in an arbitrary time-local master equation is

$$\int_0^t d\tau \mathcal{D}(\tau) \geq \mathbf{0}, \quad (2.32)$$

that the dissipator is on-average positive definite in the interaction picture. Higher-order tests for complete positivity would rely upon higher-order terms of the Magnus series.

#### 2.2.1.4 Exact Second-Order Master Equations

In Sec. 2.2.1.2, we have shown that a Gaussian noise cumulant decomposition causes the cancelation of potentially secular terms which would arise at fourth order and higher orders in the master equation. The remaining non-secular terms do not cancel because their operator, time, and trace ordering are not generally the same. However, there are situations where these higher-order terms do cancel, specifically when the system coupling operators are constant or at least effectively constant when integrated alongside environmental correlations. Such is the case when the noise is delta correlated, in which case the system operators do not have time to evolve under integration. Also if the system coupling operators commute with the system Hamiltonian, then they will be stationary in the interaction picture and will not evolve regardless of reservoir timescales. Therefore, *given Gaussian noise, the second-order master equation is exact for either a Markovian process or stationary coupling.*

For the Markovian process this property can also be seen most easily in the influence functional. The influence phase trivially resolves into a much more mundane second-order term. For stationary system couplings this property can also be seen in the stochastic Schrödinger equation and corresponding convolutionless master-equation formalism [144]. In this case the effective noise derivative can be exactly solved for, as it is completely lacking in dynamics.

### 2.2.1.5 The Born-Markov Approximation

The second-order master equation is both consistent with second-order perturbation in the system-environment coupling and consistent with the Markovian limit, which can be viewed as the zeroth-order limit of weak system-energy perturbation. In general, these are two radically different regimes. Related to these two regimes of validity, the second-order master equation is in agreement with the *Born-Markov approximation* and *Redfield equation* after time localization. The *Born approximation* is to assume the system and environment,  $\rho_C(t)$ , to remain factorized in time

$$\rho_C(t) \approx \rho_S(t) \otimes \rho_E(0) \quad (2.33)$$

with the bath insignificantly influenced by the system. This is a reasonable approximation in the weak-coupling regime. The *Markov Approximation* is then said to approximate bath correlations as delta correlations when integrated alongside system variables. When used together in deriving the open-system master equation these two approximations constitute the *Born-Markov Approximation* [22]. The often stated Markov approximation in the interaction picture is

$$\frac{d}{dt}\underline{\rho}(t) = \int_0^t d\tau \langle \underline{\mathcal{L}}_I(t) \underline{\mathcal{L}}_I(\tau) \underline{\rho}(\tau) \rangle_E, \quad (2.34)$$

$$\approx \int_0^t d\tau \langle \underline{\mathcal{L}}_I(t) \underline{\mathcal{L}}_I(\tau) \rangle_E \underline{\rho}(t), \quad (2.35)$$

and often the integration limit is taken to infinity, corresponding to the late-time dynamics. Note that the density matrix can be pulled out of the integral if the remaining kernel is a delta correlation. Finally, the stationary limit of the master equation coefficients often accompanies this approximation.

It may seem curious that the Born-Markov approximation is more generally consistent with the second-order (in the system-environment interaction) master equation, even when far from the Markovian regime such as for zero-temperature reservoirs with moderate cutoff frequencies. In general, a Markovian approximation is not reasonable outside of the Markovian or near-Markovian regime. Indeed if one were to apply the very same Markov approximation to remaining system variables still inside the integral of Eq. (2.35), then the result would be incorrect outside of the Markovian regime. It is specifically in the Born-Markov approximation, that the Markov approximation is consistent with lowest-order perturbation theory and does not require highly localized correlations. The nonlocal effects can only arrive at higher-order perturbation regardless of bath correlation timescales. This is a typical occurrence in integro-differential equations which are only perturbatively nonlocal.

## 2.2.2 Stationary Master Equations

In this section we consider the case where all Hamiltonian terms are constant in time. The second-order operator ( $\mathbf{A}_{nm} \diamond \mathbf{L}_m$ ) in Eq. (2.18) is reduced to quadrature in the energy basis where it is given by a Hadamard product.

$$\langle \omega_i | \mathbf{A}_{nm} \diamond \mathbf{L}_m | \omega_{i'} \rangle = \langle \omega_i | \mathbf{A}_{nm} | \omega_{i'} \rangle \langle \omega_i | \mathbf{L}_m | \omega_{i'} \rangle , \quad (2.36)$$

with the  $\mathbf{A}_{nm}$  operator defined

$$\langle \omega_i | \mathbf{A}_{nm} | \omega_{i'} \rangle \equiv A_{nm}(\omega_{ii'}) , \quad (2.37)$$

$$A_{nm}(t; \omega) = \int_0^t d\tau \alpha_{nm}(t, \tau) e^{-\omega(t-\tau)} , \quad (2.38)$$

where  $\omega_{ii'} = \omega_i - \omega_{i'}$ . If the correlation function is sufficiently localized in time, then these coefficients will have a stationary limit.

It will be useful to decompose  $A_{nm}$  into its Hermitian and anti-Hermitian parts in the noise index

$$\text{He}[A_{nm}(t; \omega)] = \frac{1}{2} [A_{nm}(t; \omega) + A_{mn}^*(t; \omega)] , \quad (2.39)$$

$$\text{An}[A_{nm}(t; \omega)] = \frac{1}{2i} [A_{nm}(t; \omega) - A_{mn}^*(t; \omega)] , \quad (2.40)$$

where for a univariate correlation function these will reduce to the real and imaginary parts respectively. Next we define the pseudo-stationary correlation

$$\boldsymbol{\alpha}_t(\Delta t) = \begin{cases} \boldsymbol{\alpha}(t+\Delta t, t) & \Delta t < 0 \\ \boldsymbol{\alpha}(t, t-\Delta t) & 0 < \Delta t \end{cases} , \quad (2.41)$$

with Hermiticity relation  $\boldsymbol{\alpha}_t(\Delta t) = \boldsymbol{\alpha}_t^\dagger(-\Delta t)$  and where for truly stationary noise one simply has  $\boldsymbol{\alpha}_t(\Delta t) = \boldsymbol{\alpha}(\Delta t)$ . All of the following calculations may then proceed as if we have a stationary correlation. E.g. the characteristic function, or power spectral density, is defined to be

$$\tilde{\boldsymbol{\alpha}}_t(\omega) = \int_{-\infty}^{+\infty} d\tau e^{-i\omega\tau} \boldsymbol{\alpha}_t(\tau) . \quad (2.42)$$

The full-time master equation coefficients can now be represented

$$\text{He}[A_{nm}(t; \omega)] = \frac{1}{2} \delta_t(\omega) * \tilde{\alpha}_{nm}(\omega) , \quad (2.43)$$

$$\text{An}[A_{nm}(t; \omega)] = -\mathcal{P}_t \left[ \frac{1}{\omega} \right] * \delta_t(\omega) * \tilde{\alpha}_{nm}(\omega) , \quad (2.44)$$

with  $*$  the appropriate Fourier convolution in frequency space and the late-time

limiting delta function and Cauchy principal value defined

$$\delta_t(\omega) \equiv \frac{\sin(\omega t)}{\pi\omega}, \quad (2.45)$$

$$\mathcal{P}_t \left[ \frac{1}{\omega} \right] \equiv \frac{1 - \cos(\omega t)}{\omega}. \quad (2.46)$$

Assuming a sufficiently smooth and localized correlation function, the late-time coefficients will then be

$$\text{He}[A_{nm}(\omega)] = \frac{1}{2} \tilde{\alpha}_{nm}(\omega), \quad (2.47)$$

$$\text{An}[A_{nm}(\omega)] = -\mathcal{P} \left[ \frac{1}{\omega} \right] * \tilde{\alpha}_{nm}(\omega), \quad (2.48)$$

and thus the late-time coefficients obey a Kramers-Kronig relation and are causal response functions. Bochner's theorem dictates that positive-definite correlation functions in the frequency domain will arise from stationary (and positive-definite) correlation functions in the time domain. Therefore, for stationary correlations and with some assumptions of continuity, the Hermitian coefficients will comprise a positive-definite matrix in the late-time limit.

Perhaps more useful computationally is the Laplace representation.

$$\hat{\alpha}_t(s) = \int_0^\infty d\tau e^{-s\tau} \alpha_t(\tau). \quad (2.49)$$

In the Laplace domain, Eq. (2.38) is merely a frequency shift of the correlation function before the time integration.

$$\hat{A}_{nm}(s; \omega) = \frac{1}{s} \hat{\alpha}_{nm}(s + i\omega), \quad (2.50)$$

and from the final value theorem we have the late-time coefficients

$$A_{nm}(\omega) = \hat{\alpha}_{nm}(i\omega). \quad (2.51)$$

This is generally the fastest method for obtaining the late-time coefficients, assuming one has obtained functions with analytic continuation into the complex plane. From the Kramers-Kronig relation, the late-time coefficients should be analytic in the upper half of the complex plane.

Finally, the pseudo-Lindblad master-equation coefficients can be represented

$$\mathcal{D}_{ii';jj'} = \sum_{nm} \langle \omega_i | \mathbf{L}_m | \omega_{i'} \rangle \mathcal{A}_{ii';jj'} \overline{\langle \omega_j | \mathbf{L}_n | \omega_{j'} \rangle}, \quad (2.52)$$

in terms of the kernel

$$\mathcal{A}_{ii';jj'} \equiv A_{nm}(\omega_{ii'}) + A_{mn}^*(\omega_{jj'}), \quad (2.53)$$

all evaluated in the energy basis  $\mathbf{e}_{ii'} = |\omega_i\rangle\langle\omega_{i'}|$ . In the *rotating-wave approximation* one only keeps the terms stationary in the interaction picture of the free system:

$$\mathcal{D}_{ii';ii'} = \sum_{nm} \langle \omega_i | \mathbf{L}_m | \omega_{i'} \rangle 2 \text{He}[A_{nm}(\omega_{ii'})] \overline{\langle \omega_i | \mathbf{L}_n | \omega_{i'} \rangle}, \quad (2.54)$$

and

$$\mathcal{D}_{ii;jj} = \sum_{nm} \langle \omega_i | \mathbf{L}_m | \omega_i \rangle 2 \text{He}[A_{nm}(0)] \overline{\langle \omega_j | \mathbf{L}_n | \omega_j \rangle}. \quad (2.55)$$

as well as the corresponding stationary contributions of  $\mathbf{V}$ . These terms are a quadratic form on the correlation function. If one has a correlation function which is stationary, at least in the late-time limit, then the RWA coefficients will take on a Lindblad form. Therefore the rotating-wave approximation constitutes a Lindblad-projection of the master equation. Correspondence between the RWA-Lindblad master equation and perturbation theory will be discussed in Sec. 4.4.

### 2.2.2.1 Second-Order Solutions

Given sufficiently-localized environment correlations, the master-equation coefficients asymptote primarily within the environment timescales and secondarily within the free-system timescales. We give some exact results for linear systems in Chapter 3 where this behavior can be seen explicitly. The short-time relaxation, which occurs on the order of the environment cutoff frequency, can be particularly violent and the master equation coefficients are said to *jolt*. Such jolts are result of pathological initial conditions where the system environment are initially factorized, yet subsequent evolution occurs with finite interaction strength. In the context of linear master equations, avoidance of jolts by the preparation of correlated initial states or by turning on the interaction gradually are discussed in Sec. 4.3.

Therefore given some properly correlated or coupled environment, the dissipative portion of the master equation should asymptote to its stationary value smoothly and (for weak coupling) within timescales which are much shorter than those of its effects. Upon this initial relaxation, the system evolves in a manner invariant to time translations

$$\lim_{t > \tau \gg 0} \mathcal{G}(t, \tau) = e^{(t-\tau)\mathcal{L}(\infty)}. \quad (2.56)$$

Therefore in the weak-coupling regime it typically suffices to consider the dynamics generated by the stationary limit of the master equation. As with the time-independent Schrödinger equation, one can use canonical perturbation theory to compute the stationary propagator  $e^{t\mathcal{L}}$ . Full-time solutions can also be calculated analytically, and to within some perturbative order, via Fer expansion [56].

Canonical perturbation theory is applied specifically to the eigen-value problem

$$\mathcal{L} \mathbf{o}_{ij} = f_{ij} \mathbf{o}_{ij}, \quad (2.57)$$

where the eigen-values  $f_{ij}$  and corresponding right eigen-operators  $\mathbf{o}_{ij}$  of  $\mathcal{L}$  can be expanded perturbatively in powers of the coupling as

$$\mathbf{o}_{ij} = |\omega_i\rangle\langle\omega_j| + \delta\mathbf{o}_{ij} + \dots, \quad (2.58)$$

$$f_{ij} = -i\omega_{ij} + \delta f_{ij} + \dots, \quad (2.59)$$

here to second order, where  $\omega_{ij} = \omega_i - \omega_j$ . We will assume no resonance or near resonance in the energy-level splittings, though it will be more-or-less clear how to apply degenerate perturbation theory to these cases. By construction, the zeroth-order terms are set correctly. The second-order terms are set by the corresponding order of terms in the master equation.

$$\langle\omega_{i'}|\mathcal{L}_2\{|\omega_i\rangle\langle\omega_j|\}\rangle|\omega_{j'}\rangle = -i(\omega_{ij} - \omega_{i'j'})\langle\omega_{i'}|\delta\mathbf{o}_{ij}|\omega_{j'}\rangle + \delta f_{ij}\delta_{ij;i'j'}. \quad (2.60)$$

Evaluating the components of this equation yields the non-degenerate corrections.

$$\langle\omega_{i'}|\delta\mathbf{o}_{ij}|\omega_{j'}\rangle = \frac{\langle\omega_{i'}|\mathcal{L}_2\{|\omega_i\rangle\langle\omega_j|\}\rangle|\omega_{j'}\rangle}{-i(\omega_{ij} - \omega_{i'j'})}, \quad (2.61)$$

$$\delta f_{ij} = \langle\omega_i|\mathcal{L}_2\{|\omega_i\rangle\langle\omega_j|\}\rangle|\omega_j\rangle, \quad (2.62)$$

where  $\omega_{ij} \neq \omega_{i'j'}$ . The second-order timescales  $f_{ij}$  are determined strictly by the RWA coefficients, Eq. (2.54)-(2.55). The non-degenerate perturbative frequency

shifts are found to be

$$\begin{aligned}
\delta f_{ij} &= \sum_{nm} \langle \omega_i | \mathbf{L}_m | \omega_i \rangle 2 \operatorname{He}[A_{nm}(0)] \overline{\langle \omega_j | \mathbf{L}_n | \omega_j \rangle} \\
&\quad - \sum_{nmk} \langle \omega_k | \mathbf{L}_m | \omega_i \rangle A_{nm}(\omega_{ki}) \overline{\langle \omega_k | \mathbf{L}_n | \omega_j \rangle} \\
&\quad - \sum_{nmk} \langle \omega_k | \mathbf{L}_m | \omega_j \rangle A_{mn}^*(\omega_{kj}) \overline{\langle \omega_k | \mathbf{L}_n | \omega_j \rangle}. \tag{2.63}
\end{aligned}$$

This relation reveals that the Hermitian part of  $A_{nm}(\omega)$  only gives rise to the real timescales, i.e. growth and decay, while the anti-Hermitian part only gives rise to imaginary timescales. Upon completing the square with the first term, as we shall do in Eq. (2.72), one can see that  $\operatorname{He}[A_{nm}(\omega)]$  (and thus  $\tilde{\alpha}(\omega)$  at late time) being positive definite will force the real timescales to be negative. This decay corresponds to the decoherence of (perturbatively) off-diagonal components of the density matrix while in the energy basis.

Even when far from resonance, the unperturbed eigen-operators are always degenerate along the diagonal where  $\omega_{ii} = 0$  and the previous perturbation theory only applies to the off-diagonal entries. For the diagonal evolution, we need the linear combinations of diagonal entries that branch under perturbation. The correct diagonal combination

$$\mathbf{p} = \sum_i p_i |\omega_i\rangle\langle\omega_i|, \tag{2.64}$$

with second-order eigen-frequency  $\delta f$  must satisfy the characteristic equation

$$\langle \omega_i | \mathcal{L}_2\{\mathbf{p}\} | \omega_i \rangle = \delta f \langle \omega_i | \mathbf{p} | \omega_i \rangle, \tag{2.65}$$

This is also a well defined eigen-value problem. Written more conveniently in matrix

notation we have

$$\mathbf{W} \vec{\mathbf{p}} = \delta f \vec{\mathbf{p}}, \quad (2.66)$$

with the characteristic matrix and diagonal vector defined

$$\llbracket \mathbf{W} \rrbracket_{ij} = \langle \omega_i | \mathcal{L}_2 \{ |\omega_j\rangle \langle \omega_j| \} | \omega_i \rangle, \quad (2.67)$$

$$\llbracket \vec{\mathbf{p}} \rrbracket_i \equiv \langle \omega_i | \mathbf{p} | \omega_i \rangle, \quad (2.68)$$

and where the characteristic matrix evaluates to

$$\llbracket \mathbf{W} \rrbracket_{ij} = \sum_{nm} \langle \omega_i | \mathbf{L}_m | \omega_j \rangle 2 \text{He}[A_{nm}(\omega_{ij})] \overline{\langle \omega_i | \mathbf{L}_n | \omega_j \rangle}, \quad (2.69)$$

for  $i \neq j$  and for the diagonals

$$\llbracket \mathbf{W} \rrbracket_{ii} = - \sum_{k \neq i} \langle \omega_k | \mathbf{W} | \omega_i \rangle. \quad (2.70)$$

Note that away from resonance  $\mathbf{W}$  receives no contribution from the anti-Hermitian coefficients and is encapsulated by the RWA coefficients. Associated with this characteristic equation is the Pauli master equation

$$\frac{d}{dt} \vec{\mathbf{p}} \approx \mathbf{W} \vec{\mathbf{p}}, \quad (2.71)$$

though it should be noted that this neglects the second-order corrections to the eigen-operators, of which the second-order off-diagonal perturbations are obtainable directly from the second-order master equation.

For  $n$  energy levels, then there is an order  $n$  characteristic polynomial that needs to be factored. Directly solving the master equation would have been order  $n^2$ . The zero temperature limit is a particularly simple case to solve for.  $\mathbf{W}$  is upper

triangular in accord with the lack of thermal activation; eigen-values are determined by the diagonal entries and eigen-vectors by simple matrix inversion.

The diagonal entries  $[\mathbf{W}]_{ii}$  denote maximal instantaneous relaxation rates for the perturbatively diagonal entries of the density matrix in the energy basis. The perturbatively off-diagonal relaxation (decoherence) rates can be expressed

$$\text{Re}[\delta f_{ij}] = \frac{[\mathbf{W}]_{ii} + [\mathbf{W}]_{jj}}{2} - \sum_{nm} d\ell_m^{[ij]} \text{He}[A_{nm}(0)] \bar{d}\ell_n^{[ij]}, \quad (2.72)$$

given the matrix elements

$$d\ell_n^{[ij]} \equiv \langle \omega_i | \mathbf{L}_n | \omega_i \rangle - \langle \omega_j | \mathbf{L}_n | \omega_j \rangle, \quad (2.73)$$

and therefore the decoherence rates are strictly larger (more negative) than the (average) maximal diagonal decay rates. Aside from an overall Markovian limit, only the decoherence rates reference  $A_{nm}(0)$  or the Markovian coefficients. This gives the decoherence rates additional sensitivities to the infrared regime (e.g.  $1/f$  noise) which are not present among the stationary relaxation rates. *Decoherence can occur on timescales much more rapid than thermalization.*

It is not difficult to see from (2.69) that the columns of  $\mathbf{W}$  are not independent, as they all add up to zero. In fact they must do so to preserve the trace of the density matrix. Therefore there is always at least one state within the null space of  $\mathbf{W}$ . This is a stationary state with characteristic frequency  $\delta f = 0$ . Any stationary master equation must have a stationary state. Depending on the details of the model, there may be additional stationary states. These will also be energy states (to lowest order in the coupling) in accord with the “quantum limit of einselection” [125], but here as a simple consequence of perturbation theory.  $\mathbf{W}$  is weakly column diagonal

dominant given a positive definite  $\text{He}[A_{nm}(\omega)]$ , as will be the case for stationary reservoir in the late-time limit where  $\text{He}[A_{nm}(\omega)] = \frac{1}{2}\tilde{\alpha}_{nm}(\omega)$ . The Gershgorin circle theorem immediately implies that there can only be damped oscillations and that the damping rates strictly outpace the oscillation rates.

Finally we note that second-order diagonal perturbations to the operators  $\mathbf{p}$  in the degenerate subspace require the fourth-order Pauli master equation, as there will be corrections of the form  $\mathbf{W}_4/f_2$  because of degeneracy (see Sec. 4.2). *There is often a mistaken expectation that the second-order master equation should produce second-order solutions*, e.g. complete positivity to second order, but this is not the case. The range of approximations between zeroth and second-order perturbation can be organized as follows: zeroth-order limiting solutions, RWA solutions, solutions to the second-order master equation, and finally the second-order solutions. Each successive approximation contains more information than the last (see Sec. 4.4), however there can be a trade-off in positivity between the RWA and second-order master equation. Solutions to the second-order master equation without fourth-order Pauli equation can easily violate positivity at second order. This is particularly exacerbated by low temperature environments where the asymptotic state has a largely vanishing diagonal to zeroth-order and one lacks the proper second-order correction to the diagonals which are needed to accommodate the off-diagonal perturbations.

### 2.2.2.2 The Damping Basis

The damping basis [23] is simply the basis which diagonalizes  $\mathcal{L}$ . It is a basis of operators or matrices and not necessarily physical states or vectors, as in general the eigen-operators  $\mathbf{o}_{ij}$  cannot be expressed as an outer product of state vectors.  $\mathcal{L}$  is not necessarily Hermitian or normal in the ordinary sense of linear algebra, and so the left eigen-operators are not determined by the super-adjoint of the right eigen-operators. This is not to be confused with the fact that  $\mathcal{L}$  is Hermitian and normal in the sense of preserving the Hermiticity and trace of the density matrix. The master equation sense of Hermiticity implies that the eigen-system has an adjoint symmetry,  $\mathcal{L} \mathbf{o}_{ij}^\dagger = f_{ij}^* \mathbf{o}_{ij}^\dagger$ . The master equation sense of normality implies that the identity matrix is always a left eigen-matrix with eigen-value zero as

$$\frac{d}{dt} \text{Tr}[\boldsymbol{\rho}] = \text{Tr}[\mathbf{1} \mathcal{L} \boldsymbol{\rho}] = 0, \quad (2.74)$$

for all  $\boldsymbol{\rho}$ ; the corresponding right eigen-state to  $\mathbf{1}$  being a stationary state.

In dissipative quantum mechanics we must resort to calculating the dual, left eigen-operators (super-vectors)  $\mathbf{o}_{ij}^*$  such that  $\mathbf{o}_{ij}^* \mathcal{L} = \mathbf{o}_{ij}^* f_{ij}$  where  $f_{ij}$  is also the eigen-value of the corresponding right eigen-operator  $\mathbf{o}_{ij}$ . To clarify, left and right super-multiplication with super-vectors is defined as

$$\boldsymbol{\rho}_L^* \mathcal{L} \boldsymbol{\rho}_R = \sum_{ij;i'j'} \langle i | \boldsymbol{\rho}_L | j \rangle \langle i | \mathcal{L} \{ |i'\rangle \langle j'| \} | j \rangle \langle i' | \boldsymbol{\rho}_R | j' \rangle, \quad (2.75)$$

and the spectral decomposition of our open-system propagator can be represented by the sum of outer-products

$$e^{t\mathcal{L}} = \sum_{ij} e^{f_{ij}t} \mathbf{o}_{ij} \mathbf{o}_{ij}^*, \quad (2.76)$$

which acts upon states as

$$e^{t\mathcal{L}}\boldsymbol{\rho} = \sum_{ij} e^{f_{ij}t} \left( \sum_{i'j'} \langle i' | \mathbf{o}_{ij}^* | j' \rangle \langle i' | \boldsymbol{\rho} | j' \rangle \right) \mathbf{o}_{ij}. \quad (2.77)$$

The left eigen-operator  $\mathbf{o}_{ij}^*$  projects out the  $\mathbf{o}_{ij}$  component from  $\boldsymbol{\rho}$ . Perturbatively, the dual vectors can be written  $\mathbf{o}_{ij}^* = |\omega_i\rangle\langle\omega_j| + \delta\mathbf{o}_{ij}^* + \mathcal{O}(\mathcal{L}_1^4)$ , and the second-order terms of the left eigen-value equation are then

$$\langle\omega_i | \mathcal{L}_2 \{ |\omega_{i'}\rangle\langle\omega_{j'}| \} | \omega_j \rangle = -\imath(\omega_{ij} - \omega_{i'j'}) \langle\omega_i | \delta\mathbf{o}_{ij}^* | \omega_{j'} \rangle + \delta f_{ij} \delta_{ij;i'j'}. \quad (2.78)$$

As must be the case, the corresponding eigen-values are the same while the left eigen-operators become

$$\langle\omega_{i'} | \delta\mathbf{o}_{ij}^* | \omega_{j'} \rangle = \frac{\langle\omega_i | \mathcal{L}_2 \{ |\omega_{i'}\rangle\langle\omega_{j'}| \} | \omega_j \rangle}{-\imath(\omega_{ij} - \omega_{i'j'})}, \quad (2.79)$$

where  $\omega_{ij} \neq \omega_{i'j'}$ . Due to degeneracy, the left eigen-vectors of  $\mathbf{W}$  must be solved non-perturbatively. As for the non-degenerate eigen-vectors, their orthogonality relation perturbatively evaluates to

$$\mathbf{o}_{ij}^* \mathbf{o}_{i'j'} = \delta_{ij;i'j'} + \mathcal{O}(\mathcal{L}_1^4), \quad (2.80)$$

and therefore the eigen-basis is not only perturbatively orthogonal but also normalized to second order.

### 2.2.3 Cyclo-Stationary Master Equations

Here we will briefly discuss the applicability of second-order master equations to time-dependent problems by a simple analysis of asymptotically cyclo-stationary master equations. Such master equations can arise when stationary free-system dynamics are under the influence of cyclo-stationary correlations (defined

in Sec. 2.5.1.2) or when periodic free-system dynamics are under the influence of stationary correlations.

As a brief review of periodic systems, recall that Floquet's theory asserts that given a periodic Hamiltonian  $\mathbf{H}(t)$ , the free solutions take the form

$$\boldsymbol{\psi}(t) = \mathbf{P}_0(t) e^{-i\mathcal{H}t} \boldsymbol{\psi}(0), \quad (2.81)$$

for pure states in the Schrödinger picture, where  $\mathbf{P}_0(t)$  is a unitary operator with the same period as  $\mathbf{H}(t)$  and  $\mathcal{H}$  is the time-homogeneous pseudo-Hamiltonian whose eigen-values are the pseudo-energy levels. Effectively,  $\mathbf{P}_0(t)$  acts as a change of basis between the time-dependent Hamiltonian motion and the time-independent pseudo-Hamiltonian motion.  $\mathbf{P}_0(t)$  has its own Fourier decomposition in terms of the Hamiltonian frequency  $\Omega_H$  and induces a Fourier decomposition upon transformed operators

$$\mathbf{P}_0^\dagger(t) \mathbf{X} \mathbf{P}_0(t) = \sum_u \mathbf{X}_{[u]} e^{+iu\Omega_H t}, \quad (2.82)$$

with which one can calculate the full spectral decomposition of Heisenberg picture observables

$$\langle \omega_i | \underline{\mathbf{X}}(t) | \omega_j \rangle = \sum_u \langle \omega_i | \mathbf{X}_{[u]} | \omega_j \rangle e^{+i\omega_{ij}t} e^{+iu\Omega_H t}, \quad (2.83)$$

where  $|\omega\rangle$  here denotes the pseudo-energy basis. This spectral decomposition, whether for the system or environment, will allow for calculation of the second-order master equation coefficients in Eq. (2.18) in terms of the more mundane stationary coefficients in Eq. (2.38).

Given stationary environment correlations and periodic system dynamics, the

second-order operator

$$\langle \omega_i | \mathbf{P}_0^\dagger(t) (\mathbf{A}_{nm} \diamond \mathbf{L}_m) \mathbf{P}_0(t) | \omega_j \rangle , \quad (2.84)$$

can be decomposed into

$$\sum_u A_{nm}(\omega_{ij} + u\Omega_{\text{H}}) e^{+iu\Omega_{\text{H}}t} \langle \omega_i | \mathbf{L}_m^{[u]} | \omega_j \rangle , \quad (2.85)$$

here evaluated in the free concyclic picture of the system, which will turn out to be more useful than the Schrödinger picture.

Given stationary system dynamics yet cyclo-stationary environment correlations as resolved in Eq. (2.164), the second-order coefficients can be expressed by the Hadamard product formulas, Eq. (2.36)-(2.37), with the cyclo-stationary coefficients

$$A_{nm}(\omega) = \sum_{uv} A_{nm}^{[uv]}(\omega - v\Omega_{\text{H}}) e^{i(u-v)\Omega_{\text{H}}t} , \quad (2.86)$$

where the coefficients  $A_{nm}^{[uv]}(\omega)$  are calculated also according to Eq. (2.38), but with the stationary kernels  $\alpha_{[uv]}(t)$  given by Eq. (2.165).

In either case the mundane stationary coefficients  $A(\omega + u\Omega_{\text{H}})$  remain asymptotically stationary and therefore the full coefficients are asymptotically periodic. The cyclo-stationary master equation can therefore be represented with modulated stationary master equation coefficients.

### 2.2.3.1 Second-Order Solutions

Let us assume that we have an asymptotically and perturbatively cyclo-stationary master equation of the general form

$$\dot{\rho}(t) = -i[\mathcal{H}, \rho(t)] + \mathcal{L}_2(t)\{\rho(t)\}, \quad (2.87)$$

$$\mathcal{L}_2(t) = \mathcal{L}_2(t+\mathcal{T}), \quad (2.88)$$

where if our system had a periodic Hamiltonian, then we would have transformed into the free concyclic picture via  $\mathbf{P}_0(t)$

$$\rho(t) \rightarrow \mathbf{P}_0^\dagger(t) \rho(t) \mathbf{P}_0(t), \quad (2.89)$$

so that the dynamics are zeroth-order stationary and we now work with the pseudo-Hamiltonian and concyclic dissipator. If our system has stationary dynamics and the environment has cyclo-stationary correlations then we simply remain in the Schrödinger picture.

Floquet's theorem then asserts that there is a periodic transformation of the density matrix into a basis wherein the dynamics are generated by a time-independent pseudo-Liouville operator  $\mathfrak{L}$  where

$$\mathfrak{L} = \mathfrak{L}_0 + \mathfrak{L}_2 + \mathcal{O}(\mathcal{L}_1^4), \quad (2.90)$$

$$\mathfrak{L}_0 \rho = -i[\mathcal{H}, \rho]. \quad (2.91)$$

Determination of these perturbative corrections can be made by Magnus-Floquet series, detailed in App. A.3. The easiest terms to calculate are those stationary in the interaction picture, or the RWA coefficients. These terms are simply given by the

time average of the Liouvillian  $\langle \mathcal{L}(t) \rangle$  and are sufficient to determine the timescale perturbations and degenerate evolution as in the stationary master equation. For instance, the RWA coefficients corresponding to Eq. (2.86) are given by

$$\langle A_{nm}(\omega) \rangle = \sum_u A_{nm}^{[uu]}(\omega - u\Omega). \quad (2.92)$$

As with the stationary master equation, the RWA coefficients lack some information. Here, the pseudo-energy basis is perturbed into a pseudo damping basis and there is also some (perturbatively) periodic transformation between the cyclo-stationary Liouvillian and pseudo-Liouvillian.

For finite-temperature reservoirs, one can see that cyclo-thermalization will not ensue (non-trivially at least) as detailed balance (discussed in Sec. 2.5.3.3) is lost with the frequency shifts. The system will still evolve towards a cyclo-stationary state, only it will not generally be cyclo-thermal.

## 2.3 Time-Nonlocal Master Equations

Historically, the most general open-system master equations were, as first derived, of nonlocal form such as in the projection-operator formalism of Nakajima [120] and Zwanzig [165]. These nonlocal master equations would then be localized via Born-Markov approximation or a more careful perturbative analysis. Our simple derivation of the convolutionless master equation in Sec. 2.2.1 contained no appeal to a nonlocal form. Here we will demonstrate the dual non-secular perturbation theory which naturally produces a nonlocal master equation. We will also discuss similarities between the two *representations*.

### 2.3.1 Perturbative Master Equations

We will consider the same sort of system-environment-interaction decomposition, but instead of time-local master equations we consider time-nonlocal master equations of the form

$$\frac{d}{dt}\boldsymbol{\rho}_C(t) = \boldsymbol{\mathcal{K}}_C(t) * \boldsymbol{\rho}_C(t), \quad (2.93)$$

$$\boldsymbol{\mathcal{K}}_C(t) = \boldsymbol{\mathcal{K}}_F(t) + \boldsymbol{\mathcal{K}}_I(t), \quad (2.94)$$

where  $*$  denotes the Laplace convolution. Therefore we should expect a perturbative level of agreement between the two analyses for closed system + environment Liouvillians which are constant in time. In the Laplace frequency domain, we have an algebraic equation for the closed system + environment propagator

$$s\hat{\boldsymbol{\mathcal{G}}}_C(s) - \mathbf{1} = \hat{\boldsymbol{\mathcal{K}}}_C(s)\hat{\boldsymbol{\mathcal{G}}}_C(s), \quad (2.95)$$

which readily admits perturbative solutions via the Neumann series

$$\hat{\boldsymbol{\mathcal{G}}}(s) = \hat{\boldsymbol{\mathcal{G}}}_0(s) + \left\langle \hat{\boldsymbol{\mathcal{G}}}_F(s)\hat{\boldsymbol{\mathcal{K}}}_I(s)\hat{\boldsymbol{\mathcal{G}}}_F(s) \right\rangle_E + \left\langle \hat{\boldsymbol{\mathcal{G}}}_F(s)\left[\hat{\boldsymbol{\mathcal{K}}}_I(s)\hat{\boldsymbol{\mathcal{G}}}_F(s)\right]^2 \right\rangle_E + \dots, \quad (2.96)$$

$$\hat{\boldsymbol{\mathcal{G}}}_F(s) \equiv \left[ s - \hat{\boldsymbol{\mathcal{K}}}_F(s) \right]^{-1}, \quad (2.97)$$

after tracing over the environment. Again, the perturbative propagator is secular in nature, but we will only use it as an intermediate in the perturbative approximation of the open-system Liouville kernel.

$$\hat{\boldsymbol{\mathcal{K}}}(s) = s - \hat{\boldsymbol{\mathcal{G}}}(s)^{-1}. \quad (2.98)$$

For Gaussian noise we find the second and fourth-order kernels to be

$$\hat{\kappa}_2(s) = \hat{\mathcal{G}}_0(s)^{-1} \left\langle \hat{\mathcal{G}}_F(s) \left[ \hat{\kappa}_I(s) \hat{\mathcal{G}}_F(s) \right]^2 \right\rangle_E \hat{\mathcal{G}}_0(s)^{-1}, \quad (2.99)$$

$$\hat{\kappa}_4(s) = \hat{\mathcal{G}}_0(s)^{-1} \left\langle \hat{\mathcal{G}}_F(s) \left[ \hat{\kappa}_I(s) \hat{\mathcal{G}}_F(s) \right]^4 \right\rangle_E \hat{\mathcal{G}}_0(s)^{-1} - \hat{\kappa}_2(s) \hat{\mathcal{G}}_0(s) \hat{\kappa}_2(s). \quad (2.100)$$

For a stationary reservoir this simplifies to

$$\hat{\kappa}_2(s) = \left\langle \hat{\kappa}_I(s) \hat{\mathcal{G}}_F(s) \hat{\kappa}_I(s) \right\rangle_E, \quad (2.101)$$

$$\hat{\kappa}_4(s) = \left\langle \hat{\kappa}_I(s) \left[ \hat{\mathcal{G}}_F(s) \hat{\kappa}_I(s) \right]^3 \right\rangle_E - \hat{\kappa}_2(s) \hat{\mathcal{G}}_0(s) \hat{\kappa}_2(s). \quad (2.102)$$

### 2.3.2 The Second-Order Master Equation

For a stationary reservoir with time-local interaction, the second-order Liouville kernel can be expressed in terms of the time-local Liouville operator as

$$\kappa_2(t) = \dot{\mathcal{L}}_2(t) \mathcal{G}_0(t), \quad (2.103)$$

and for a time-local system Hamiltonian we can additionally express

$$\hat{\kappa}_2(s) \{|\omega_i\rangle\langle\omega_j|\} = s' \hat{\mathcal{L}}_2(s') \{|\omega_i\rangle\langle\omega_j|\} \Big|_{s'=s+\iota\omega_{ij}}, \quad (2.104)$$

in Laplace space. Using Eq. (2.50), we have the relation

$$s \hat{A}_{nm}(s; \omega) = \hat{\alpha}_{nm}(s + \iota\omega), \quad (2.105)$$

which reveals a particular shift-scale symmetry of the nonlocal master equation.

The Liouville kernel can therefore be expressed

$$\hat{\kappa}_2(s) \{|\omega_i\rangle\langle\omega_j|\} = s \hat{\mathcal{L}}_2(s + 2\iota\omega_{ij}) \{|\omega_i\rangle\langle\omega_j|\}. \quad (2.106)$$

In terms of the noise kernel  $\hat{\alpha}(s)$ , it is more clear that while the time-local master equation always has Markovian representation, the time-nonlocal master equation only has Markovian representation for the Markovian process, or at least when the process appears Markovian with respect to the freely-evolving system coupling operators.

Though the Liouville kernel is slightly more difficult to express than the time-local analog, the full-time solution is simply

$$\hat{\mathcal{G}}(s) = \left[ s - \hat{\mathcal{K}}(s) \right]^{-1}, \quad (2.107)$$

in the Laplace domain. Using the final-value theorem, the asymptotic state is

$$\rho(\infty) = \lim_{s \rightarrow 0} s \left[ s - \hat{\mathcal{K}}(s) \right]^{-1} \rho(0), \quad (2.108)$$

which, under some assumptions of convergence, is equivalent to

$$\rho(\infty) = \lim_{t \rightarrow \infty} e^{t\hat{\mathcal{K}}(0)} \rho(0). \quad (2.109)$$

Therefore the stationary operator  $\hat{\mathcal{K}}(0)$  must determine the asymptotic state just as the stationary operator  $\mathcal{L}(\infty)$  does. By comparison with either (2.104) or (2.106) we have

$$\hat{\mathcal{K}}_2(s) \{ |\omega_i\rangle\langle\omega_i| \} = s \hat{\mathcal{L}}_2(s) \{ |\omega_i\rangle\langle\omega_i| \}, \quad (2.110)$$

which is sufficient to ensure that the two operators share the same zeroth-order stationary states, and second-order corrections to any zeroth-order stationary state.

The late-time dynamics are slightly more involved to compare. Ideally, one would decompose the full-time propagator into its early and late-time behavior

$$\hat{\mathcal{G}}(s) = \hat{\mathcal{G}}_\infty(s) + \delta\hat{\mathcal{G}}_i(s), \quad (2.111)$$

where  $\hat{\mathcal{G}}_\infty(s)$  denotes the late-time local evolution which decays slowly with the perturbation, and  $\delta\hat{\mathcal{G}}_i(s)$  denotes the short-time nonlocal evolution which decays more rapidly with system and environment timescales. The asymptotically dominant  $\hat{\mathcal{G}}_\infty(s)$  would then correspond to  $e^{t\mathcal{L}(\infty)}$ , possibly modulo some amplitude and phase differences. This relation will be demonstrated once we have obtained solutions.

### 2.3.3 Second-Order Solutions

Our perturbative solutions will follow the spirit of canonical perturbation theory. First we inspect the structure of the exact solution, which takes the form

$$\hat{\mathcal{G}}(s) = \left[ s - \hat{\mathcal{K}}(s) \right]^{-1}, \quad (2.112)$$

and is a rational function of the Liouville kernel. Given the eigen-system  $\hat{k}_{ij}(s)$ ,  $\hat{\mathbf{o}}_{ij}(s)$  of  $\hat{\mathcal{K}}(s)$  such that

$$\hat{\mathcal{K}}(s) \hat{\mathbf{o}}_{ij}(s) = \hat{k}_{ij}(s) \hat{\mathbf{o}}_{ij}(s), \quad (2.113)$$

then the exact solutions can also be expressed

$$\hat{\mathcal{G}}(s) = \sum_{ij} \frac{1}{s - \hat{k}_{ij}(s)} \hat{\mathbf{o}}_{ij}(s) \hat{\mathbf{o}}_{ij}^*(s), \quad (2.114)$$

where  $\hat{\mathbf{o}}_{ij}^*(s)$  is the left eigen-matrix dual to  $\hat{\mathbf{o}}_{ij}(s)$  as described in Sec. (2.2.2.2). We will construct a perturbative solution from the perturbative eigen-system, which will constitute a rational approximation of the exact solution akin to a Padé approximant. Further note that, with  $\hat{\mathcal{K}}(s)$  perturbatively truncated, exact solutions

to the perturbative master equation are already rational approximants of the exact solutions, so there is no new kind of mangling in doing this.

The nature of such a perturbative solution is slightly different than for the time-local perturbation. The perturbative evaluation of  $e^{dt\mathcal{L}(t)}$  is non-secular, but may contain slight amplitude and phase discrepancy and can only be applied for  $\mathcal{L}(t)$  commuting or for small  $dt$ . The nonlocal perturbative solutions are effectively full time, but being a rational approximation in Laplace space they can introduce some slightly secular behavior. For instance, exact timescales can become duplicated with small (here fourth-order) error, giving rise to a very slow beat frequency. Regardless, we will show these perturbative solutions to be convergent with the correct asymptotics.

The second-order eigen-value problem constrains the non-degenerate perturbative corrections to be

$$\langle \omega_{i'} | \delta \hat{\mathbf{o}}_{ij}(s) | \omega_{j'} \rangle = \frac{\langle \omega_{i'} | \hat{\mathcal{K}}_2(s) \{ |\omega_i\rangle\langle\omega_j| \} | \omega_{j'} \rangle}{-\imath(\omega_{ij} - \omega_{i'j'})}, \quad (2.115)$$

for the operator corrections, where  $\omega_{ij} \neq \omega_{i'j'}$ , and

$$\delta \hat{k}_{ij}(s) = \langle \omega_i | \hat{\mathcal{K}}_2(s) \{ |\omega_i\rangle\langle\omega_j| \} | \omega_j \rangle. \quad (2.116)$$

for the eigen-value corrections. Given the similarity mentioned in Eq. (2.110), the “frequency” will be perturbed by a term analogous to the time-local formula. The eigen-value perturbation is particularly important to analyze, as the stability of our perturbative solution is dictated by its poles, or equivalently the roots of

$$s - \hat{k}_{ij}(s) = s + \imath\omega_{ij} - \delta \hat{k}_{ij}(s) + \dots, \quad (2.117)$$

having negative real part. Assuming the correlation function to be well regulated, these poles should occur as perturbations of the system frequencies and perturbations of scales which regulate the correlations, such as the cutoff. For well-regulated correlations our attention turns to the near-resonance poles,  $s \approx -\imath\omega_{ij}$ . We can actually solve for these poles perturbatively, they are simply

$$s_{ij} = -\imath\omega_{ij} + \delta\hat{k}_{ij}(-\imath\omega_{ij}) + \dots, \quad (2.118)$$

and are precisely the late-time eigen-values of the time-local master equation given the relation

$$\hat{\mathcal{K}}_2(-\imath\omega_{ij})\{|\omega_i\rangle\langle\omega_j|\} = \mathcal{L}_2(\infty)\{|\omega_i\rangle\langle\omega_j|\}, \quad (2.119)$$

$$\hat{\mathcal{K}}_2(\mathcal{L}_0) = \mathcal{L}_2(\infty), \quad (2.120)$$

where the operator-Laplace transform in the second equation is interpreted as resulting in the first equation. This general relation also reveals the asymptotic propagator from Eq. (2.114) as correctly being the rational approximant

$$\hat{\mathcal{G}}_\infty(s) = \left[ s - \hat{\mathcal{K}}(\mathcal{L}_0) \right]^{-1}, \quad (2.121)$$

to second order, assuming the degenerate dynamics also work out correctly.

### 2.3.3.1 The Pauli Master Equation

Just as for the time-local master equation, the nonlocal master equation is confronted with degeneracy in the stationary states. Here we must find the correct linear combination of diagonal matrices  $\hat{\mathbf{q}}$ , and associated vectors  $\vec{\mathbf{q}}_i = \langle\omega_i|\hat{\mathbf{q}}|\omega_i\rangle$ ,

which branch under perturbation

$$\hat{\mathbf{V}}(s) \vec{\mathbf{q}}(s) = \delta \hat{k}(s) \vec{\mathbf{q}}(s) , \quad (2.122)$$

$$\langle \omega_i | \hat{\mathbf{V}}(s) | \omega_j \rangle = \langle \omega_i | \hat{\mathcal{K}}_2(s) \{ | \omega_j \rangle \langle \omega_j | \} | \omega_i \rangle , \quad (2.123)$$

where from similarity (2.110) we have the correspondence

$$\hat{\mathbf{V}}(s) = s \hat{\mathbf{W}}(s) , \quad (2.124)$$

to the time-local Pauli master equation, Eq. (2.66). The asymptotic dynamics follow in accord with the time-local master equation but the full-time evolution is, in general, exceedingly complicated from this perspective. Full-time solutions may only be feasible in either the Markovian limit, where it is trivial, or in the zero temperature limit, where  $\mathbf{W}$  is upper triangular in accord with the lack of thermal activation.

## 2.4 Non-Markovian Quantum Regression Theorem

An open-system master equation provides a propagator  $\mathcal{G}(t)$  and super-adjoint propagator  $\mathcal{G}^\dagger(t)$  such that

$$\text{Tr} [\mathbf{X} \mathcal{G}(t) \{ \rho \}] = \text{Tr} [\mathcal{G}^\dagger(t) \{ \mathbf{X} \} \rho] . \quad (2.125)$$

Given this, one could construct a ‘‘Heisenberg picture’’ via  $\mathbf{X}(t) \equiv \mathcal{G}^\dagger(t) \{ \mathbf{X} \}$ . However, for multi-time correlations this ‘‘Heisenberg picture’’ does not necessarily have any relation to the Heisenberg picture of the microscopic model. This is very easy to see, as in general

$$\langle \mathbf{X}_1(t_1) \mathbf{X}_2(t_2) \rangle_{\text{E}} \neq \langle \mathbf{X}_1(t_1) \rangle_{\text{E}} \langle \mathbf{X}_2(t_2) \rangle_{\text{E}} . \quad (2.126)$$

For the closed system or an open system driven by Markovian processes, the Quantum Regression Theorem asserts that the dynamics of multi-time correlations can be generated via  $\mathcal{L}(t)$  or more specifically its super-adjoint  $\mathcal{L}^\dagger(t)$ . However this is a defining property of the Markovian process and will not hold true in the non-Markovian regime.

In this section we approach the category of non-Markovian dynamics which cannot be described by the regression theorem. First we derive the adjoint master equation, which is dual to the ordinary master equation. Following this simple derivation, that of the two-time correlations is more straightforward. Finally we regard observables of the environment.

### 2.4.1 The Adjoint Master Equation

First we would like to derive the (super) adjoint master equation for the dynamics of single-time operators

$$\mathbf{X}(t) = \langle \{\mathbf{X}\}(t) \rangle_{\mathbf{E}} = \left\langle \mathcal{G}_{\mathbf{C}}^\dagger(t) \{\mathbf{X}\} \right\rangle_{\mathbf{E}}. \quad (2.127)$$

As we already know the ordinary master equation, no new information will be gained in doing this, but the basic procedure will remain largely the same for multi-time correlations. The super-adjoint propagator satisfies the differential equation

$$\frac{d}{dt} \mathcal{G}_{\mathbf{C}}^\dagger(t) = \mathcal{G}_{\mathbf{C}}^\dagger(t) \mathcal{L}_{\mathbf{C}}^\dagger(t), \quad (2.128)$$

where here in the unitary theory of the system + environment we have

$$\mathcal{L}_{\mathbf{C}}^\dagger(t) \mathbf{X} = [+i \mathbf{H}_{\mathbf{C}}(t), \mathbf{X}], \quad (2.129)$$

and therefore our adjoint master equation can be expressed

$$\dot{\mathbf{X}}(t) = \mathcal{G}^\dagger(t) \left\{ \mathcal{L}_0^\dagger(t) \mathbf{X} \right\} + \left\langle \mathcal{G}_C^\dagger(t) \mathcal{L}_I^\dagger(t) \{ \mathbf{X} \} \right\rangle_E, \quad (2.130)$$

where we have used the fact that  $\mathcal{L}_E^\dagger(t) \{ \mathbf{X} \} = 0$  for system operators. We want to express the right-hand side in terms of single-time operators, therefore the second term is cast into the form of an adjoint super-operator

$$\dot{\mathbf{X}}(t) = \mathcal{G}^\dagger(t) \left\{ \mathcal{L}_0^\dagger(t) \mathbf{X} \right\} + \mathcal{G}^\dagger(t) \{ \delta \mathcal{L}^\dagger(t) \mathbf{X} \}, \quad (2.131)$$

$$\delta \mathcal{L}^\dagger(t) \equiv \left\langle \mathcal{G}_C^\dagger(t) \right\rangle_E^{-1} \left\langle \mathcal{G}_C^\dagger(t) \mathcal{L}_I^\dagger(t) \right\rangle_E \quad (2.132)$$

which will be calculated perturbatively. To do this, first we express the closed system propagator via the interaction picture where

$$\mathcal{G}_C^\dagger(t) = \underline{\mathcal{G}}_C^\dagger(t) \mathcal{G}_F^\dagger(t), \quad (2.133)$$

$$\mathcal{L}_I^\dagger(t) = \mathcal{G}_F^{-\dagger}(t) \underline{\mathcal{L}}_I^\dagger(t) \mathcal{G}_F^\dagger(t). \quad (2.134)$$

The interaction picture propagator then has the integral equation of motion

$$\underline{\mathcal{G}}_C^\dagger(t) = \int_0^t d\tau \underline{\mathcal{G}}_C^\dagger(\tau) \underline{\mathcal{L}}_I^\dagger(t), \quad (2.135)$$

which is directly amenable to perturbation theory via Neumann series expansion.

Ordinary perturbation theory then reveals the lowest order correction

$$\underline{\mathcal{L}}_I^\dagger(t) = \int_0^t d\tau \left\langle \underline{\mathcal{L}}_I^\dagger(\tau) \underline{\mathcal{L}}_I^\dagger(t) \right\rangle_E, \quad (2.136)$$

and one can see that this is indeed the super-adjoint of Eq. (2.10). More specifically,

for the sum of separable couplings the super-adjoint master equation resolves to

$$\mathcal{L}_2^\dagger(t) \mathbf{X} = \sum_{nm} \{ (\mathbf{A}_{nm} \diamond \mathbf{L}_m)^\dagger [\mathbf{X}, \mathbf{L}_n] + [\mathbf{L}_n, \mathbf{X}] (\mathbf{A}_{nm} \diamond \mathbf{L}_m) \}. \quad (2.137)$$

## 2.4.2 Two-Time Correlations

For two-time and other multi-time correlations, one should note that there will be no assurance of a quantum regression theorem (QRT):

$$\left\langle \dot{\mathbf{X}}_1(t_1) \mathbf{X}_2(t_2) \right\rangle_{\mathbf{E}} \neq \left\langle \{ \mathcal{L}^\dagger(t_1) \mathbf{X}_1 \}(t_1) \{ \mathbf{X}_2 \}(t_2) \right\rangle_{\mathbf{E}}, \quad (2.138)$$

given a non-Markovian process. Here we look for perturbative corrections to the standard QRT formula. After application of second-order perturbation theory, of the same form as was used for the adjoint master equation, the standard QRT terms on the right-hand side are seen to match the zeroth-order terms and most of the second-order terms of the left-hand side. The second-order remainder is such that

$$\begin{aligned} \left\langle \dot{\mathbf{X}}_1(t_1) \mathbf{X}_2(t_2) \right\rangle_{\mathbf{E}} &= \left\langle \{ \mathcal{L}^\dagger(t_1) \mathbf{X}_1 \}(t_1) \{ \mathbf{X}_2 \}(t_2) \right\rangle_{\mathbf{E}} \\ &+ \int_0^{t_2} d\tau \left\langle \underline{\mathcal{L}}_1^\dagger(t_1) \{ \underline{\mathbf{X}}_1(t_1) \} \underline{\mathcal{L}}_1^\dagger(\tau_2) \{ \underline{\mathbf{X}}_2(t_2) \} \right\rangle_{\mathbf{E}}, \end{aligned} \quad (2.139)$$

and it is also revealed, but not directly shown here, that it is imperative to cast the standard QRT terms inside a single environmental trace, as to cancel certain highly secular terms.

A general non-secular perturbative fit to this expression is not so obvious, so we will resort to a more specific model. For a constant Hamiltonian with separable coupling to a stationary reservoir, the second-order non-Markovian corrections to the QRT evaluate to

$$- \sum_{nm} [\underline{\mathbf{L}}_n(t_1), \underline{\mathbf{X}}_1(t_1)] \left[ \int_0^{t_2} d\tau \alpha_{nm}(t_1, \tau) \underline{\mathbf{L}}_m(\tau), \underline{\mathbf{X}}_2(t_2) \right], \quad (2.140)$$

in agreement with Ref. [49]. The two-time integral

$$\int_0^{t_2} d\tau \alpha_{nm}(t_1, \tau) \underline{\mathbf{L}}_m(\tau) = \mathcal{G}_0^\dagger(t_1) \{ (\mathbf{A}_{nm} \diamond \mathbf{L}_m)(t_1, t_2) \}, \quad (2.141)$$

can be expressed in terms of ordinary second-order operators as

$$(\mathbf{A}_{nm} \diamond \mathbf{L}_m)(t_1, t_2) \equiv (\mathbf{A}_{nm} \diamond \mathbf{L}_m)(t_1) - (\mathbf{A}_{nm} \diamond \mathbf{L}_m)(t_1 - t_2), \quad (2.142)$$

and it should be noted that this non-Markovian correction requires no renormalization as it involves the difference between two ordinary second-order operators. The most natural non-secular expression to fit (2.140) to is

$$- \sum_{nm} \{[\mathbf{L}_n, \mathbf{X}_1]\}(t_1) \left[ \{(\mathbf{A}_{nm} \diamond \mathbf{L}_m)(t_1, t_2)\}(t_1), \{\mathbf{X}_2\}(t_2) \right], \quad (2.143)$$

which is obviously non-Markovian and cannot be expressed as a sum of two operators evaluated at two times. Note that, although it was necessary to place the standard QRT terms inside a single environmental trace, at least here at second order, the corrections may be considered as a product of single-time operators or a collective multi-time correlation. Higher-order perturbation theory might reveal which representation is superior (in the stronger-coupling regime) as second-order perturbation did for Eq. (2.139).

These non-Markovian corrections do not vanish in the weak coupling limit (second-order) but, from Eq. (2.142), if  $t_1 > t_2$  they will vanish in the Markovian limit. More generally, the two-time operator vanishes in the softer limit of  $t_1 \gg t_2$ , as compared to the system and environment timescales, much in the same manner that the master equation takes its stationary limit. The stationary limit of this expression does not merely require late times, but also a similarly lengthy span of time between the two times of the correlation. So, even at late times when one has a time-homogeneous master equation, the non-Markovian property does not

go away. This should not be surprising as the noise process does not stop being non-Markovian.

### 2.4.3 Environmental Excitations

Here we consider the dynamics of environment mode excitations in terms of the reduced system dynamics. Consider a spectral decomposition of the environment modes

$$\mathbf{H}_E = \sum_i \varepsilon_i, \quad (2.144)$$

where  $\varepsilon |\varepsilon\rangle = \varepsilon |\varepsilon\rangle$ . As there are a continuum of modes, inspecting one individual mode would be rather meaningless as it can only exchange an infinitesimal amount of energy with the system. Instead we will inspect the dynamics of some finite range of modes

$$\mathbf{E}_\chi = \sum_i \chi(\varepsilon_i) \varepsilon_i, \quad (2.145)$$

where  $0 \leq \chi(\varepsilon) \leq 1$  is an indicator function with finite support; it indicates the range of modes to be inspected. The expectation value for  $\mathbf{E}_\chi$  will be infinite for a reservoir, but  $\dot{\mathbf{E}}_\chi$  will be finite corresponding to the finite amount of energy exchanged with the system. This energy exchange can be reduced to the single-time expectation value

$$\left\langle \dot{\mathbf{E}}_\chi(t) \right\rangle_E = \mathcal{G}^\dagger(t) \{ \wp_\chi(t) \}, \quad (2.146)$$

in terms of the operator

$$\wp_\chi(t) \equiv \left\langle \mathcal{G}_C^\dagger(t) \right\rangle_E^{-1} \left\langle \mathcal{G}_C^\dagger(t) \mathcal{L}_1^\dagger(t) \{ \mathbf{E}_\chi \} \right\rangle_E, \quad (2.147)$$

which is amenable to non-secular perturbation theory. The reduced system operator  $\wp_\chi$  denotes the power delivered to the environment modes as indicated by the function  $\chi(\varepsilon)$ . At second order in the coupling and for a stationary environment, the reservoir power becomes

$$\underline{\wp}_\chi(t) = \int_0^t d\tau \left\langle \underline{\mathcal{L}}_1^\dagger(\tau) \underline{\mathcal{L}}_1^\dagger(t) \{ \mathbf{E}_\chi \} \right\rangle_{\mathbf{E}}. \quad (2.148)$$

For a system Hamiltonian constant in time and separable coupling to the environment, after integration by parts and some manipulation, the reservoir power can be expressed

$$\wp_\chi = -\mathcal{L}_\chi^\dagger \{ \mathbf{H} \} - \left[ \frac{d}{dt} + \mathcal{L}_0^\dagger \right] \{ 2 \mathbf{V}_\chi \}, \quad (2.149)$$

where all  $\chi$  indicated values are calculated with the noise kernel

$$\tilde{\alpha}_\chi(\omega) \equiv \tilde{\alpha}(\omega) [\chi(+\omega) + \chi(-\omega)], \quad (2.150)$$

which only references environment modes indicated by  $\chi(\varepsilon)$ . Consistent with second-order perturbation, we can also express the reservoir power

$$\wp_\chi = - \left[ \frac{d}{dt} + \mathcal{L}_\chi^\dagger \right] \{ \mathbf{H} + 2 \mathbf{V}_\chi \}. \quad (2.151)$$

The first term of the reservoir power is simply the (not yet renormalized) system power loss whereas the remaining term can be confirmed to belong to the interaction energy. The  $\mathbf{V}$  here is the same as that of the Lindblad or pseudo-Lindblad form master equation given by Eq. (2.20) and is the extent of what is renormalizable for the system. Renormalization cannot affect the form of this expression, in its entirety, as for any simple counter term  $\mathcal{L}_R$  we have  $\mathcal{L}_R^\dagger \{ \mathbf{E}_\chi \} = 0$  identically.

Any “renormalization” of the system power must be a reidentification of interaction power here. More intuitively, any power due to renormalization energy and power renormalization of the system energy always cancel at second order.

$$\mathcal{L}_0\{\mathbf{H}_R\} + \mathcal{L}_R\{\mathbf{H}\} = \mathbf{0}. \quad (2.152)$$

Eq. (2.151) has revealed that  $2\mathbf{V}$ , modulo some renormalization, is the effective interaction energy. The prefactor of 2 in the interaction power is somewhat unfortunate; a prefactor of unity would have implied the reservoir power to be naturally cutoff insensitive. Because of its intrinsic time dependence, which would reveal bare environmental correlations after differentiation, one can see that it also jolts near the initial time. This must be another artifact of the uncorrelated initial conditions.

In the late-time regime, in which the transient terms have vanished and all remaining terms have reached their asymptotic values, the system power operator  $\mathcal{L}^\dagger\{\mathbf{H}\}$  will not vanish, but once the system has completely relaxed then its expectation value will vanish. The remaining operator, which describes energy exchange between the environment and interaction, will also not vanish as the system Hamiltonian does not generally commute with  $\mathbf{V}$ . But once the system is fully relaxed, the interaction power expectation value will vanish at second order. This is because the system relaxes approximately to an energy mixture which approximately commutes with the Hamiltonian.

We can inspect a reservoir mode of energy approximately  $\varepsilon_0$  but with some narrow but non-vanishing resolution  $\delta\varepsilon$ . In computing the full-time coefficients,

$A_\chi(t; \omega)$ ,  $\frac{1}{\delta\varepsilon}$  is a new timescale for which we must wait for the coefficients to asymptote. The  $\text{An}[A_\chi(\omega)]$  coefficients will only pick up what is indicated by  $\chi$  in their integral and are therefore small like  $\delta\varepsilon$ . The  $\text{He}[A_\chi(\omega)]$  coefficients retain their full strength, but only for  $\omega \approx \varepsilon_0$ . These are the predominant late-time terms for the inspection of reservoir modes of fairly precise frequency.

## 2.5 Environmental Correlations

### 2.5.1 Classification of Correlations

#### 2.5.1.1 Positivity and Decoherence

The (multivariate) environmental correlation function, first defined in Eq. (2.19), is Hermitian in the sense of

$$\boldsymbol{\alpha}(t, \tau) = \boldsymbol{\alpha}^\dagger(\tau, t), \quad (2.153)$$

and also positive definite in the sense of

$$\int_0^t d\tau_1 \int_0^t d\tau_2 \mathbf{f}^\dagger(\tau_1) \boldsymbol{\alpha}(\tau_1, \tau_2) \mathbf{f}(\tau_2) \geq 0, \quad (2.154)$$

for all vector functions  $\mathbf{f}(t)$  indexed by the noise. All quantum correlations are at least *nonlocally decoherent*: any resultant algebraic dissipator will be positive definite for all time,  $\boldsymbol{\Delta}(t) > \mathbf{0}$ . This property is required for completely-positive time evolution as proven in Sec. 2.2.1.3. Nonlocal decoherence only implies that there is more net decoherent evolution than recoherent evolution, as per Eq. (2.63). The more strict property of decoherence at every instant in time,  $\dot{\boldsymbol{\Delta}}(t) > \mathbf{0}$ , will

only generally be satisfied by delta correlations which exhibit *local decoherence*. Such correlations will always produce a Lindblad master equation, as can be inferred from Eq. (2.52). However some very restricted classes of system-environment interactions, such as the RWA-interaction Hamiltonian (see Sec. 4.4), can be constrained by their coupling to be instantaneously decoherent. This characterizes the class of master equations which are of naturally Lindblad form and yet non-Markovian in their dynamics.

Finally we note these stochastic processes are partially ordered in their *decoherence strength*. Given two correlation functions, one can sometimes order them  $\alpha_+(t, \tau) > \alpha_-(t, \tau)$  according to the positivity relation (2.154). For instance, the set of univariate Markov processes is totally ordered by the scalar magnitude of the respective delta correlations, e.g.  $2\delta(t-\tau) > 1\delta(t-\tau)$ . This idea is given more consideration, including nontrivial examples, in Sec. 4.7.

### 2.5.1.2 Time-Dependence

*Stationary* correlations are defined by their invariance to time translation

$$\alpha(t, \tau) = \alpha(t-\tau), \quad (2.155)$$

and can produce asymptotically stationary (time-constant) master equations. Such correlations are produced when the environment is in an initially stationary state

$$\rho_E(0) = \sum_i p_E(\varepsilon_i) |\varepsilon_i\rangle\langle\varepsilon_i|, \quad (2.156)$$

where  $|\varepsilon\rangle$  denotes the energy basis of the environment and  $p_E(\varepsilon)$  are its stationary probabilities at the initial time. Furthermore the coupling operators must be

constant in time, resulting in

$$\alpha_{nm}(t, \tau) = \sum_{ij} p_E(\varepsilon_i) \langle \varepsilon_i | \mathbf{L}_n | \varepsilon_{ij} \rangle \overline{\langle \varepsilon_i | \mathbf{L}_m | \varepsilon_{ij} \rangle} e^{+i\varepsilon_j(t-\tau)}. \quad (2.157)$$

Comparing to the mode sum

$$\boldsymbol{\alpha}(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega e^{+i\omega t} \tilde{\boldsymbol{\alpha}}(\omega), \quad (2.158)$$

the accompanying characteristic function can be readily identified as

$$\tilde{\alpha}_{nm}(\omega) \propto 2\pi \sum_i p_E(\varepsilon_i) \langle \varepsilon_i | \mathbf{L}_n | \varepsilon_i - \omega \rangle \overline{\langle \varepsilon_i | \mathbf{L}_m | \varepsilon_i - \omega \rangle}, \quad (2.159)$$

where the underscored proportionality here is strictly in reference to the continuum limit of the reservoir which relates environmental mode sums to integrals given the infinitesimal strength of individual environmental mode couplings. This can be more rigorously defined through the use of a finite spectral-density function in place of the infinitesimal environment couplings.

Also of note are *Quasi-stationary* correlations such as of the form

$$\boldsymbol{\alpha}(t, \tau) = \boldsymbol{\alpha}_S(t-\tau) + \boldsymbol{\delta}\boldsymbol{\alpha}_{NS}(t+\tau), \quad (2.160)$$

where  $\boldsymbol{\alpha}_S(t-\tau)$  denotes a stationary correlation while  $\boldsymbol{\delta}\boldsymbol{\alpha}_{NS}(t+\tau)$  is an additional non-stationary contribution. Such correlations will result from linear coupling to an environment with non-stationary initial state. Other kinds of quasi-stationary correlations can result from quadratic coupling, etc. Due to their highly oscillatory behavior in the late-time limit, given the Fourier representation

$$\boldsymbol{\delta}\boldsymbol{\alpha}_{NS}(t+\tau) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\varepsilon e^{+i\varepsilon(t+\tau)} \boldsymbol{\delta}\tilde{\boldsymbol{\alpha}}_{NS}(\varepsilon), \quad (2.161)$$

the non-stationary dynamical contributions typically lose effect therein. Therefore quasi-stationary correlations can produce an asymptotically stationary master equation with equivalent asymptotics as generated by their corresponding stationary projection  $\alpha_S$ .

*Cyclo-stationary* correlations are defined by their invariance to periodic translations

$$\alpha(t_1, t_2) = \alpha(t_1 + \mathcal{T}, t_2 + \mathcal{T}), \quad (2.162)$$

and can produce asymptotically cyclo-stationary (periodic) master equations. Such correlations are produced when the environment is in an initially stationary state and its coupling operators are periodic in time.

$$\mathbf{l}_n(t) = \sum_u \mathbf{l}_n^{[u]} e^{+iu\Omega_H t}, \quad (2.163)$$

where  $\Omega_H = \frac{2\pi}{\mathcal{T}}$  is the interaction period. The correlation function  $\alpha(t_1, t_2)$  can then be expressed

$$\sum_{uv} \alpha_{[uv]}(t_1 - t_2) e^{+i\frac{u+v}{2}\Omega_H(t_1 - t_2)} e^{+i\frac{u-v}{2}\Omega_H(t_1 + t_2)}, \quad (2.164)$$

in terms of the stationary kernels

$$\alpha_{nm}^{[uv]}(t) = \sum_{ij} p_E(\varepsilon_i) \langle \varepsilon_i | \mathbf{l}_n^{[u]} | \varepsilon_j \rangle \overline{\langle \varepsilon_i | \mathbf{l}_m^{[v]} | \varepsilon_j \rangle} e^{+i\varepsilon_{ij}t}. \quad (2.165)$$

and such that the non-stationary factors of the full correlation function are more obviously periodic. Equivalent correlations are produced when the environment is in an initially cyclo-stationary state and its coupling operators are constant in time. I.e. the bath modes have Floquet-normal-form solutions such that the coupling

operators admit the spectral decomposition

$$\underline{\mathbf{1}}_n(t) = \sum_{ij,u} \langle \varepsilon_i | \underline{\mathbf{1}}_n^{[u]} | \varepsilon_{ij} \rangle e^{+i\varepsilon_j t} e^{+iu\Omega_H t}, \quad (2.166)$$

where  $|\varepsilon\rangle$  denotes the pseudo-energy basis of the environment associated with the Floquet-normal-form solutions and  $\Omega_H$  is the period of the environment Hamiltonian.

## 2.5.2 Correlation Function Decomposition

Second-order correlation functions can always be decomposed into a real noise kernel and dissipation kernel.

$$\underbrace{\boldsymbol{\alpha}(t, \tau)}_{\text{complex noise}} = \underbrace{\boldsymbol{\nu}(t, \tau)}_{\text{noise}} + i \underbrace{\boldsymbol{\mu}(t, \tau)}_{\text{dissipation}}, \quad (2.167)$$

where the Hermiticity stated in Eq. (2.153) leads to the relations

$$\boldsymbol{\nu}(t, \tau) \equiv \frac{1}{2} [\boldsymbol{\alpha}(t, \tau) + \boldsymbol{\alpha}^T(\tau, t)], \quad (2.168)$$

$$\boldsymbol{\mu}(t, \tau) \equiv \frac{1}{2i} [\boldsymbol{\alpha}(t, \tau) - \boldsymbol{\alpha}^T(\tau, t)]. \quad (2.169)$$

The role of each kernel can be inferred from the *influence functional* [57, 26] and *quantum Langevin equation* [67]. The noise kernel  $\boldsymbol{\nu}$  appears in the influence kernel as the correlation of an ordinary real stochastic source, whereas the dissipation kernel  $\boldsymbol{\mu}$  alone would produce a purely homogeneous (though not generally positivity-preserving) evolution.

For stationary correlations  $\boldsymbol{\alpha}(t-\tau)$  with characteristic function (Fourier transform)  $\tilde{\boldsymbol{\alpha}}(\omega)$ , the real noise and damping kernels are then Hermitian in both noise

index and frequency argument

$$\tilde{\gamma}(\omega) = \tilde{\gamma}^\dagger(\omega) = \tilde{\gamma}^*(-\omega), \quad (2.170)$$

$$\tilde{\nu}(\omega) = \tilde{\nu}^\dagger(\omega) = \tilde{\nu}^*(-\omega). \quad (2.171)$$

One implication is that, in the noise index, their real symmetric parts are even functions of the frequency while their imaginary anti-symmetric parts are odd functions of the frequency. More importantly, from Bochner's theorem both  $\tilde{\alpha}(\omega)$  and  $\tilde{\nu}(\omega)$  are positive-definite for all frequencies. Again the damping kernel  $\tilde{\gamma}(\omega)$  may be positive definite, negative definite, or indefinite.

The two kernels naturally decompose the second-order operators, Eq. (2.18), into their Hermitian and anti-Hermitian parts, in the ordinary sense of Hilbert space operators.

$$\mathbf{A}_{nm} = \underbrace{\mathbf{N}_{nm}}_{\text{diffusion}} + i \underbrace{\mathbf{M}_{nm}}_{\text{dissipation}}, \quad (2.172)$$

$$(\mathbf{N}_{nm} \diamond \mathbf{L}_m)(t) \equiv \int_0^t d\tau \nu_{nm}(t, \tau) \{ \mathcal{G}_0(t, \tau) \mathbf{L}_m(\tau) \}, \quad (2.173)$$

$$(\mathbf{M}_{nm} \diamond \mathbf{L}_m)(t) \equiv \int_0^t d\tau \mu_{nm}(t, \tau) \{ \mathcal{G}_0(t, \tau) \mathbf{L}_m(\tau) \}, \quad (2.174)$$

The second-order master equation can then be expressed entirely in terms of Hermitian operators

$$\mathcal{L}_2 \rho = - \sum_{nm} [\mathbf{L}_n, i \{ (\mathbf{M}_{nm} \diamond \mathbf{L}_m), \rho \} + [(\mathbf{N}_{nm} \diamond \mathbf{L}_m), \rho]]. \quad (2.175)$$

Here the noise coefficients describe diffusion while the dissipation coefficients describe dissipation (or the opposite), renormalization, and other homogeneous dynamics.

The correlation function  $\boldsymbol{\alpha}(t, \tau)$  is positive definite and therefore the noise kernel  $\boldsymbol{\nu}(t, \tau)$  must also be positive definite. The dissipation kernel  $\boldsymbol{\mu}(t, \tau)$  is not positive definite, but it is related to the damping kernel  $\boldsymbol{\gamma}(t, \tau)$ , which is given by

$$\boldsymbol{\mu}(t, \tau) = -\frac{\partial}{\partial \tau} \boldsymbol{\gamma}(t, \tau), \quad (2.176)$$

and can be positive definite, negative definite, or indefinite. For non-stationary noise, Eq. (2.176) is an incomplete definition, and constructing a positive-definite damping kernel may require additional considerations. Assuming relation (2.176) is sufficient, the dissipation kernel coefficients can then be expressed

$$\underbrace{(\mathbf{M}_{nm} \diamond \mathbf{L}_m)(t)}_{\text{dissipation}} = \underbrace{(\boldsymbol{\Gamma}_{nm} \diamond \dot{\mathbf{L}}_m)(t)}_{\text{damping}} - \underbrace{\gamma_{nm}(t, t) \mathbf{L}_m(t)}_{\text{renormalization}} + \underbrace{\gamma_{nm}(t, 0) \{\boldsymbol{\mathcal{G}}_0(t) \mathbf{L}_m(0)\}}_{\text{slip}}, \quad (2.177)$$

in terms of damping kernel coefficients

$$(\boldsymbol{\Gamma}_{nm} \diamond \dot{\mathbf{L}}_m)(t) \equiv \int_0^t d\tau \gamma_{nm}(t, \tau) \left\{ \boldsymbol{\mathcal{G}}_0(t, \tau) \dot{\mathbf{L}}_m(\tau) \right\}, \quad (2.178)$$

$$\dot{\mathbf{L}}_m(t) \equiv +i [\mathbf{H}(t), \mathbf{L}_m(t)] + \frac{\partial}{\partial t} \mathbf{L}_m(t), \quad (2.179)$$

which can be used to place Eq. (2.175) into a form much like the QBM master equation. Note that if the  $\mathbf{L}_n(t)$  do not commute, then Eq. (2.179) is to be taken as a definition and is only perturbatively the actual time derivative in the (post-trace) Heisenberg picture. The slip is a transient effect, a result of the factorized initial conditions, and will be avoided by the preparation of a properly correlated initial state. The renormalization is a long-lasting shift of the system Hamiltonian which would diverge in the limit of local or simple damping.

### 2.5.2.1 Renormalization

The “renormalization” terms identified in Eq. (2.177) generally contains the highest frequency sensitivities of the environment and can be mitigated by a counter term when coupling the system and environment

$$\mathbf{H}_{\text{ren}}(t) = \sum_{nm} \gamma_{nm}(t, t) \mathbf{L}_n(t) \mathbf{L}_m(t), \quad (2.180)$$

where  $\gamma_{nm}(t, t)$  was assumed to be symmetric. Our argument for renormalization of this term specifically is to match the free-system dynamics to the homogeneous dynamics (sans damping) of the interacting system. This can be more clearly seen by expressing the equation of motion of system observables (2.137), or adjoint master equation, in terms of the noise and damping kernels.

Representing the influence via linear coupling to a linear environment (harmonic oscillators), this kind of renormalization is also equivalent to redefining the environment potentials

$$\mathbf{H}_{\text{E}} + \mathbf{H}_{\text{I}} = \sum_i \left[ \frac{1}{2m_i} \mathbf{p}_i^2 + \frac{m_i \varepsilon_i^2}{2} \mathbf{q}_i^2 \right] - \sum_{ij} c_{ij} \mathbf{q}_i \mathbf{L}_j, \quad (2.181)$$

to be in equilibrium around the system

$$\mathbf{H}_{\text{E}} + \mathbf{H}_{\text{I}} + \mathbf{H}_{\text{R}} = \sum_i \left[ \frac{1}{2m_i} \mathbf{p}_i^2 + \frac{m_i \varepsilon_i^2}{2} \left( \mathbf{q}_i - \sum_j \frac{c_{ij} \mathbf{L}_j}{m_i \varepsilon_i^2} \right)^2 \right], \quad (2.182)$$

which can also be viewed as enforcing a kind of global gauge invariance between the system and environment. If the  $\mathbf{L}_n$  correspond to system positions, then this more specifically corresponds to enforcing a global translation invariance. Furthermore from (2.182) one can see that “completing the square” of the environment potential is

the minimal renormalization required to ensure a lower bound in the energy spectra for all coupling strengths.

### 2.5.2.2 Classification of Damping Kernels

Environments with positive-definite damping kernels are *damping* or *resistive* environments, while those with negative-definite damping kernels are *amplifying*. If the coupling variables  $\mathbf{L}_n$  are position variables, the damping terms correspond to forces linear in momentum. Stationary correlations are the easiest to dissect. Their dissipation and damping kernels are related by

$$\tilde{\boldsymbol{\mu}}(\varepsilon) = \imath\varepsilon \tilde{\boldsymbol{\gamma}}(\varepsilon), \quad (2.183)$$

and from the definition of the dissipation kernel in Eq. (2.169) and the double Hermiticity in Eq. (2.170)-(2.171), the damping kernel will be most-generally positive or negative definite if we have a strict inequality between positive and negative energy argued correlation functions.

$$\tilde{\boldsymbol{\alpha}}(-|\omega|) > \tilde{\boldsymbol{\alpha}}^*(+|\omega|) \quad (\text{Damping}), \quad (2.184)$$

$$\tilde{\boldsymbol{\alpha}}(-|\omega|) < \tilde{\boldsymbol{\alpha}}^*(+|\omega|) \quad (\text{Amplifying}). \quad (2.185)$$

Using Eq. (2.159), one can show that damping environments result when the initial environmental state probability  $p_E(\varepsilon)$  is a monotonically decreasing function of the environment energy. Amplifying environments result from monotonically increasing functions or *population inversion*. The most common example of each being positive and negative temperature reservoirs.

Now we must validate this classification of damping kernels. Given our damping representation of the master-equation coefficients and the adjoint master equation (2.137), one can determine the dynamics of the system energy (power) to be

$$\mathcal{L}^\dagger \mathbf{H} = - \left\{ \dot{\mathbf{L}}_n, \left( \Gamma_{nm} \diamond \dot{\mathbf{L}}_m \right) \right\} + \imath \left[ \dot{\mathbf{L}}_n, \left( \mathbf{N}_{nm} \diamond \mathbf{L}_m \right) \right], \quad (2.186)$$

where we have neglected any power generated by the slip and time-dependence intrinsic to the coupling operators. Using the zeroth-order solution  $\boldsymbol{\rho}(t) = \mathcal{G}_0(t) \boldsymbol{\rho}(0)$  and symmetries of the damping kernel, the second-order expectation value for energy lost through damping can be represented

$$\int_0^t d\tau \langle \mathcal{L}_\gamma^\dagger(\tau) \mathbf{H}(\tau) \rangle = - \int_0^t d\tau_1 \int_0^t d\tau_2 \sum_{nm} \gamma_{nm}(\tau_1, \tau_2) \text{Tr} \left[ \dot{\mathbf{L}}_n(\tau_1) \boldsymbol{\rho}(0) \dot{\mathbf{L}}_m(\tau_2) \right], \quad (2.187)$$

which will be strictly dissipative for a positive-definite damping kernel. This expression also contrasts *nonlocal damping* to *local damping*. Evaluation with a delta correlated damping kernel yields damping which is strictly dissipative at every instant of time whereas nonlocal damping is only assured to be net dissipative.

### 2.5.2.3 Fluctuation-Dissipation Relations and Inequality

From the definitions of the noise and damping kernels, Eq. (2.167)-(2.169), one can easily prove the (stationary) *fluctuation-dissipation inequality*:

$$\tilde{\nu}(\omega) \geq \pm \omega \tilde{\gamma}(\omega). \quad (2.188)$$

To prove this one simply notes that the noise kernel is the sum of two positive-definite kernels whereas the dissipation kernel is given by their difference. The essential point

is that if there is any damping, or amplification, there will be quantum noise and Eq. (2.188) determines its lower bound. This is quite a departure from classical physics where noise can be made to vanish in the zero temperature limit. The FDI and its relation to the Heisenberg uncertainty principle is discussed more thoroughly in Sec. 4.6.

For the case of one collective environment coupling, it is sufficient to define a fluctuation dissipation relation

$$\tilde{\nu}(\omega) = \tilde{\kappa}(\omega) \tilde{\gamma}(\omega), \quad (2.189)$$

$$\tilde{\kappa}(\omega) \equiv \frac{\tilde{\nu}(\omega)}{\tilde{\gamma}(\omega)}, \quad (2.190)$$

with  $\tilde{\kappa}(\omega)$  the FDR integration kernel which relates fluctuations to dissipation. For multivariate noise one could use the symmetrized product

$$\tilde{\nu}(\omega) = \frac{1}{2} [\tilde{\kappa}(\omega) \tilde{\gamma}(\omega) + \tilde{\gamma}(\omega) \tilde{\kappa}(\omega)], \quad (2.191)$$

which would ensure  $\tilde{\kappa}(\omega)$  to be positive definite if  $\tilde{\gamma}(\omega)$  is, in accord with this being a (continuous) Lyapunov equation [18]. Inequality (2.188) can then be restated as

$$\tilde{\kappa}(\omega) \geq |\omega|, \quad (2.192)$$

for damping environments. Typically  $\tilde{\kappa}(\omega)$  will contain dependence upon the precise nature of environment couplings  $\mathbf{l}_n(t)$ , in that if one changes the couplings then the FDR also changes.

### 2.5.3 Thermal Correlations

In this section we describe many of the important properties of thermal reservoirs and their corresponding correlations. We demonstrate the equivalences between *the* fluctuation-dissipation relation, Boltzmann distribution, KMS relation [100, 111], and detailed balance in the master equation.

#### 2.5.3.1 The Fluctuation Dissipation Relation

As explained in Sec. 2.5.2.3, a fluctuation-dissipation relation can be almost any (possibly tautological) relation between the noise and damping kernels, though such relations are somewhat constrained by quantum mechanics. However, if the FDR is to be independent of precisely how the system and environment are coupled, or  $\mathbf{I}_n$ , then one can work out from Eq. (2.159) that the FDR kernel  $\boldsymbol{\kappa}$  must be a scalar quantity, directly related to the initial state of the environment by way of

$$\frac{\tilde{\kappa}(\omega)}{\omega} = \frac{p_{\mathbf{E}}(\varepsilon - \omega) + p_{\mathbf{E}}(\varepsilon)}{p_{\mathbf{E}}(\varepsilon - \omega) - p_{\mathbf{E}}(\varepsilon)}, \quad (2.193)$$

for all  $\varepsilon$ . But this implies the functional relation

$$p_{\mathbf{E}}(\varepsilon - \omega) = \left[ \frac{\frac{\tilde{\kappa}(\omega)}{\omega} + 1}{\frac{\tilde{\kappa}(\omega)}{\omega} - 1} \right] p_{\mathbf{E}}(\varepsilon), \quad (2.194)$$

where the  $\omega$  translations can factor out. This factorization property is unique to exponential functions, therefore only the thermal distribution  $p_{\mathbf{E}}(\varepsilon) \propto e^{-\beta\varepsilon}$  can produce a fluctuation dissipation relation which is generally coupling independent.

We then have the thermal FDR kernel

$$\tilde{\kappa}(\omega) = \omega \coth\left(\frac{\omega}{2T}\right), \quad (2.195)$$

which must be maintained no matter how the system is coupled to the environment. Returning all dimensionful constants reveals the high-temperature and semi-classical FDR

$$\lim_{\hbar \rightarrow 0} \tilde{\kappa}(\omega) = \lim_{T \gg \omega} \tilde{\kappa}(\omega) = 2T. \quad (2.196)$$

### 2.5.3.2 KMS Relations

Here we will detail the form of correlations that all thermal reservoirs generate.

We simply assume that our ideal reservoir is in a thermal state

$$\rho_B = \frac{1}{Z} \sum_k e^{-\frac{\varepsilon_k}{T}} |\varepsilon_k\rangle\langle\varepsilon_k|, \quad (2.197)$$

$$Z \equiv \sum_k e^{-\frac{\varepsilon_k}{T}}. \quad (2.198)$$

Next we perform the change of variables  $\varepsilon_k \rightarrow \varepsilon_k + \frac{\omega}{2}$  on Eq. (2.159) to expose the inherent symmetry in all thermal coefficients.

$$\tilde{\alpha}_{nm}(\omega) \propto \frac{2\pi}{Z} e^{-\frac{\omega}{2T}} \sum_k e^{-\frac{\varepsilon_k}{T}} \left\langle \varepsilon_k + \frac{\omega}{2} \left| \mathbf{l}_n \right| \varepsilon_k - \frac{\omega}{2} \right\rangle \overline{\left\langle \varepsilon_k + \frac{\omega}{2} \left| \mathbf{l}_m \right| \varepsilon_k - \frac{\omega}{2} \right\rangle}, \quad (2.199)$$

where the last factor defines a positive-definite matrix (in the noise index) which is Hermitian in both the noise index and frequency argument. The damping and noise kernels exhibit double Hermiticity [see Sec. (2.5.2)], whereas the thermal correlation function exhibits ordinary Hermiticity in the noise index and the thermal (a)symmetry

$$\tilde{\alpha}(+\omega) = \tilde{\alpha}^T(-\omega) e^{-\frac{\omega}{T}} = \tilde{\alpha}^*(-\omega) e^{-\frac{\omega}{T}}, \quad (2.200)$$

in its frequency argument.

### 2.5.3.3 Detailed Balance

Equilibrium states are stationary states, though not all stationary states are equilibrium states. To lowest order in the coupling, stationary states  $\mathbf{p}$  are diagonal in the energy basis. The stationary constraint is given by the Pauli master equation  $\mathbf{W}$  (2.66), and takes the form  $\mathbf{W} \vec{\mathbf{p}} = 0$ , where  $\vec{\mathbf{p}}$  denotes the diagonal entries of the stationary state. In terms of the environment correlations, this works out to be the vanishing of all sums

$$0 = \sum_{jnm} \langle \omega_i | \mathbf{L}_m | \omega_j \rangle [\tilde{\alpha}_{mn}(\omega_{ji}) p_i - \tilde{\alpha}_{nm}(\omega_{ij}) p_j] \overline{\langle \omega_i | \mathbf{L}_n | \omega_j \rangle}. \quad (2.201)$$

This constraint must be satisfied by any stationary state of the system. Moreover, if the thermal state is to be insensitive to the precise nature of the system coupling, then the more strict constraint

$$\frac{p_i}{p_j} = \frac{\tilde{\alpha}_{nm}(\omega_{ij})}{\tilde{\alpha}_{mn}(\omega_{ji})}, \quad (2.202)$$

must be satisfied; this stronger constraint of term-by-term cancellation is known as *detailed balance of the solutions*. If this condition is met then the system has reached equilibrium with its environment. If there isn't much detail in the system to be balanced, then Eq. (2.202) can be satisfied trivially. Such is the case with a system consisting of a single oscillator which can only asymptote into a Gaussian state with linear coupling (see Sec. 4.6). But for detailed balance to be attainable in any system, certain transitivity properties must be present in the environmental correlations, e.g.

$$\frac{\tilde{\alpha}_{nm}(\omega_{ij})}{\tilde{\alpha}_{mn}(\omega_{ji})} = \frac{\tilde{\alpha}_{nm}(\omega_{ik}) \tilde{\alpha}_{nm}(\omega_{kj})}{\tilde{\alpha}_{mn}(\omega_{ki}) \tilde{\alpha}_{mn}(\omega_{jk})}, \quad (2.203)$$

which is known as *detailed balance of the master equation* or equivalently *detailed balance of the correlations*. Transitivity is a unique property of the exponential function, and therefore only Maxwell-Boltzmann states

$$\boldsymbol{\rho}_T \propto e^{-\beta\mathbf{H}}, \quad (2.204)$$

and thermal correlations, Eq. (2.200), can non-trivially satisfy this constraint for asymptotic states which are insensitive to the precise nature of the system-environment coupling. Given that thermal reservoirs generate thermal correlations, it can be said that the thermal state is the only state which is *self-replicating*. Any other stationary reservoir will induce a stationary state upon the system which does not generally resemble that of the environment.

Furthermore we can also say something about dynamics of thermalization with an old proof from the theory of Markov chains and Pauli master equations. Notice that given detailed balance,  $\mathbf{W}$  is similar to a real, symmetric matrix

$$\mathbf{W} = \boldsymbol{\rho}_T^{+\frac{1}{2}} \mathbf{S} \boldsymbol{\rho}_T^{-\frac{1}{2}}. \quad (2.205)$$

Being real and symmetric,  $\mathbf{S}$  must have real eigen-values. Given the similarity transform,  $\mathbf{W}$  must share the same real eigen-values. The physical implication is that in this weak coupling limit, relaxation generated by  $\mathbf{W}$  is purely decay without oscillation. This property will not hold when near resonance.

### 2.5.3.4 Properties of Thermal Correlations

From any of the above expressions, all thermal correlations and noise kernels can be expressed

$$\tilde{\alpha}(\omega) = \tilde{\gamma}(\omega) \omega \left[ \coth\left(\frac{\omega}{2T}\right) - 1 \right], \quad (2.206)$$

$$\tilde{\nu}(\omega) = \tilde{\gamma}(\omega) \omega \coth\left(\frac{\omega}{2T}\right), \quad (2.207)$$

in terms of the damping kernel  $\gamma$ . Alternatively the thermal correlation and damping kernel can be expressed

$$\tilde{\alpha}(\omega) = \tilde{\nu}(\omega) \left[ 1 - \tanh\left(\frac{\omega}{2T}\right) \right], \quad (2.208)$$

$$\tilde{\gamma}(\omega) = \tilde{\nu}(\omega) \frac{1}{\omega} \tanh\left(\frac{\omega}{2T}\right), \quad (2.209)$$

in terms of the noise kernel  $\nu$ .

For positive frequencies larger than the temperature the correlations vanish, but not necessarily for large negative frequencies. This is the asymmetry between thermal activation and freezing and its lopsidedness is most apparent in the zero-temperature limit.

$$\lim_{T \rightarrow 0} \tilde{\alpha}(\omega) = 2|\omega| \lim_{T \rightarrow 0} \tilde{\gamma}(\omega) \quad (\omega < 0). \quad (2.210)$$

The *Markovian regime* (complex white noise) corresponds both local damping and high temperature (local FDR kernel). The nomenclature of “thermal noise” as being  $f^0$  is not entirely correct.  $f^0$  correlations are Markovian and high temperature is necessary but not sufficient to be in the Markovian regime. Non-Markovian thermal noise is perfectly capable of being  $1/f^n$  noise. The Markovian regime is reached

when the system time scales are much slower than those of the environment, so that we can take the small-frequency approximation

$$\lim_{\omega \rightarrow 0} \tilde{\gamma}(\omega) = \tilde{\gamma}_0, \quad (2.211)$$

$$\lim_{\omega \rightarrow 0} \tilde{\alpha}(\omega) = 2\tilde{\gamma}_0 T. \quad (2.212)$$

These are the coefficients one would see in the Markov-Lindblad master equation as this is the limit in which the reservoir time scales are much more rapid than the system time scales.

Occasionally in the literature, zero-temperature correlations with simple dissipation,  $\tilde{\mu}(\omega)$  constant in frequency or  $1/f$  damping, are also referred to as being “white noise”. In this case, the noise kernel  $\nu$  is local and thus the real noise is white, however the environment correlations are nonlocal and thus the complex noise which unravels the system evolution is not white. This “white noise” does not strictly correspond to the Markovian regime. Corresponding to the lack of thermal activation and the lack of negative energy modes in the environment, these correlations only appear white in certain respects. The upper left plot in Fig. 2.1 shows high-temperature (complex) white noise, while the lower right plot shows low temperature “white noise”. For low-temperature noise the small  $T$ , a very long time scale, determines the scale of drastic change between positive and negative frequency behavior. Low-temperature “white noise” generally requires an infrared cutoff to be suitable for positive temperature. High-temperature white noise generally requires an ultraviolet cutoff to be suitable for finite temperature.

Finally, given a simple damping kernel, it is convenient to specify the convo-

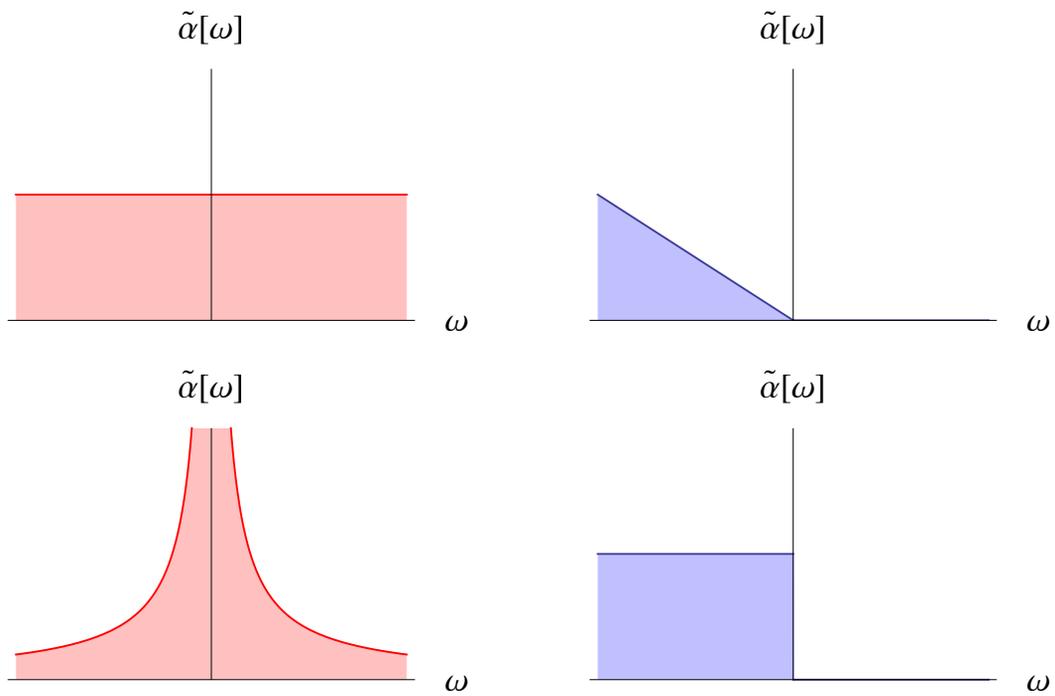


Figure 2.1: Environmental correlations  $\tilde{\alpha}(\omega)$  for high-temperature (left) and low-temperature (right) reservoirs, given simple damping  $\tilde{\gamma}(\omega)$  (top) and simple dissipation  $\tilde{\mu}(\omega)$  (bottom).

lution in (2.48) for our stationary coefficients using the rational expansion of the hyperbolic cotangent

$$\coth\left(\frac{\varepsilon}{2T}\right) = \frac{2T}{\varepsilon} + \frac{2}{\pi} \sum_{k=1}^{\infty} \frac{\frac{\varepsilon}{2\pi T}}{k^2 + \left(\frac{\varepsilon}{2\pi T}\right)^2}, \quad (2.213)$$

so that with proper choice of damping kernel, this contour integral may be evaluated via the residue theorem; we remember that the  $\pm\omega$  poles are regulated away and do not count towards the residue theorem.

Alternatively, given a simple noise kernel, it is convenient to specify the coefficients using the rational expansion of the hyperbolic tangent

$$\tanh\left(\frac{\varepsilon}{2T}\right) = \frac{2}{\pi} \sum_{k=1}^{\infty} \frac{\frac{\varepsilon}{2\pi T}}{\left(k - \frac{1}{2}\right)^2 + \left(\frac{\varepsilon}{2\pi T}\right)^2}. \quad (2.214)$$

## 2.6 Discussion

We have given a relatively simple perturbative formalism for the analysis of open-system dynamics and addressed concerns such as late-time convergence and complete positivity. Master equations not of Lindblad form (thus describing non-Markovian dynamics) are often viewed with a level of suspicion as they would seem to lack some vital structure necessary to ensure positivity. We have detailed specifically how this information is encoded in the time dependence of all microscopically derived coefficients. Moreover, one also has to temper expectations of complete positivity to the relevant perturbative order of accuracy generated by an inexact master equation. It is not well known that the second-order master equation can only provide the diagonal components of the density matrix to zeroth-order at late times [60].

Therefore when solving the second-order master equation, late-time positivity will only be preserved to zeroth-order in the late-time regime.

It is often suspected that these perturbative master equations cannot be employed for significant lengths of time, and it is certainly true that the late-time and weak-coupling limits do not always commute. However the master equation can be suitable for late-time, order-by-order in the coupling, provided that the functional distribution of noise is Gaussian (as to provide cancelation of certain potentially secular terms) and that the environmental correlations are sufficiently localized. Moreover, one must also be wary of what order of accuracy the master equation is actually capable of providing at late times. The late-time accuracy is strictly less than the early-time accuracy (see Sec. 4.2). There has also been some question in the literature raised as to the merits of the time-local versus non-local representation. As we have demonstrated, both representations share the same order of accuracy, only their higher-order errors manifest differently.

In Sec. 2.4 we reported upon non-Markovian corrections to the quantum regression theorem (QRT). The existence of such corrections have been previously noticed, even in the weak-coupling regime. Our newfound expressions make it manifest that non-Markovian dynamics of the system do not cease at late times, even if the master equation has relaxed to its stationary limit. In many calculations in the literature, the early-time dynamics of the master equation are discarded and its late-time stationary limit is employed, even for obtaining multi-time correlations. This does not correspond to an accurate calculation of the multi-time dynamics, even to only second-order system-environment interaction.

In Sec. 2.5 we made a careful decomposition of the environmentally-induced fluctuations and dissipation at second order (this characterizes all orders for Gaussian noise, see Chapter 3). Correspondences were worked out for deriving the fluctuation-dissipation relation (FDR): from first principles as in the KMS relation, from environment observable invariance in the FDR, from system observable invariance in the asymptotics, and from detailed balance.

## Chapter 3

### Exact Solution of Linear Systems

#### 3.1 Introduction

Quantum Brownian motion (QBM) of an oscillator coupled to a thermal bath of quantum oscillators has been extensively studied as a canonical model for open quantum systems as it is highly solvable and yet there is a considerable amount of insight that one can learn from it. Here we continue the lineage of work on QBM starting with the influence functional path-integral method of Feynman and Vernon [57] used by Caldeira and Leggett [27] to derive a master equation for high temperature and local damping (Ohmic), which corresponds to the Markovian regime. Also of note were the solutions in Grabert, Schramm and Ingold [77], particularly for local dissipation, though they did not derive a master equation. Following this, Caldeira, Cerdeira and Ramaswamy [25] derived the Markovian master equation for the system with weak coupling to an Ohmic bath, which was claimed to be valid at arbitrary temperature (see Sec. 3.7.3 for a critique of this claim). At the same time Unruh and Zurek [147] derived a more complete and general master equation that incorporated finite-temperature colored noise, but with local damping. Later, in a path-integral calculation from first principles, Hu, Paz and Zhang (HPZ) [84] derived a master equation for a general environment (arbitrary temperature and spectral density), barring certain subtle errors in the coefficients, which lead to in-

accurate treatment of the nonlocal dissipation cases, as we will discuss. After that, this equation has been rederived by a number of authors. Halliwell and Yu [80] exploited the phase-space transformation properties of the Wigner function for the full system plus environment and derived a Fokker-Planck equation corresponding to the HPZ equation. Calzetta, Roura and Verdaguer (CRV) [30, 29] derived it using a stochastic description for open quantum systems based on Langevin equations, whereas Ford and O’Connell [69] employed a somewhat related method via the quantum Langevin equation [67] and also obtained the solution to the HPZ equation for a Gaussian wave-packet. In [64, 66] we (Fleming, Roura and Hu) determined the precise form of the HPZ master equation coefficients and pointed out a problem with earlier derivations for nonlocal dissipation (Sec. 3.4.3.1). We additionally found concise and efficient solutions to the master equation with a number of exact nonperturbative analytical results (Sec. 3.6). Finally, we easily extended the theory to that of a system of multiple oscillators bilinearly coupled amongst themselves and to the bath in an arbitrary fashion while acted upon by external forces (Sec. 3.4.4).

There is also a cautionary tale to be found in this long lineage of work on QBM. The classical and quantum Brownian motion problem only differ (in the phase-space representation) by the fluctuation-dissipation relation (FDR) of their environmental influences. With a microscopically derived Langevin equation and quantum FDR in hand, one could have easily and correctly derived the HPZ equation in less than a minute, if one only had the (correct) non-Markovian classical Fokker-Planck equation to compare to. After making this proclamation many times, I was eventually answered by Prof. Eric Lutz, with a 1976 reference to Adelman’s work

on non-Markovian classical Brownian motion [2]. Indeed, the preceding lineage of work on QBM (1989-2010) could have been largely duplicated, at its core, with only minor adjustments to Adelman’s seven pages. However, the critical mistake which we discovered does provide an important lesson on the inequivalence between Markovian dynamics and Markovian representations, and this mistake would have likely been made at some point regardless.

Our previous work followed the approach of CRV Refs. [30, 29], making use of a stochastic description whose central element is a Langevin equation for the dynamics of the open quantum system. Here we will derive an equivalent Langevin equation, but instead of relying upon path integrals we will work from system + environment the Fokker-Planck equation and its corresponding “characteristics”, which provide the stochastic unraveling in place of the stochastic paths of the path integral. This method offers an efficient mathematical tool for obtaining all the quantum properties of the system.

One of our key contributions was uncovering a significant shortcoming of earlier results for the master equation coefficients. We point out a subtlety involving boundary conditions for solutions of integro-differential equations and explain how certain properties that hold for ordinary differential equations are not true for non-local dissipation. These properties had always been employed erroneously, in one way or another, when deriving the expressions for the master equation coefficients, even those which were then evaluated numerically. This long-standing error could have deep implications for regimes where the effects of nonlocal dissipation are significant and one should be cautious with all results for those cases reported in the

literature.

Taking into account the aspect mentioned in the previous paragraph, and using our compact formulation, we have provided a relatively simplified expression for the correct master equation. Moreover, one can also obtain the general solution to the master equation in terms of the matrix propagator of a linear integro-differential equation, and see that at late times it tends to a Gaussian state completely characterized by its covariance matrix. For meromorphic damping kernels, and many others, we are able to reduce the calculation of this covariance matrix to a simple contour integral and obtain exact nonperturbative results for finite cutoff and arbitrarily strong coupling. This includes examples of Ohmic, sub-Ohmic and supra-Ohmic environments; and from this late-time covariance one can immediately obtain the late-time diffusion coefficients as well.

In addition, working with Laplace transforms and then transforming back to time domain, we manage to find the exact solutions for the propagators associated with the integro-differential equations corresponding to Ohmic, sub-Ohmic and supra-Ohmic environments with a finite cutoff. This enables us to gain very valuable information on the dynamics of the system. For instance, for an Ohmic environment one can show that using the local approximation for the propagator is a valid approximation in the large cutoff limit, which makes it possible to obtain relatively manageable analytic results for the diffusion coefficients at all times. Furthermore, the exact solution of a specific sub-Ohmic environment reveals that long-time correlations (due to excessive coupling with IR modes of the environment) give rise to contributions to the propagator that decay at late times like power laws.

This invalidates the use of an effectively local description at late times, whose contributions decay exponentially, and provides a clear example of a situation where nonlocal dissipation needs to be properly dealt with. Finally, studying the exact solutions for some particular supra-Ohmic environment we also find significant non-local effects which are due in this case to the UV regulator function. This leads to a marked cutoff sensitivity of the momentum covariance that had not been noticed before.

## 3.2 Overview

In Sec. 3.3 we begin with a review of the classical evolution of density functions. This provides an extremely suitable perspective for quantum Brownian motion while in the phase-space or Wigner representation, which we review in the following sections. The key framework providing the stochastic description for an open quantum system in terms of a Langevin equation and its compact phase-space formulation is introduced in Sec. 3.4.1, and with this a very simple derivation of the general solution for the state evolution of the system is given in Sec. 3.4.3. The problems with previous derivations are pointed out in Sec. 3.4.2.1 and the correct derivation of the master equation is given in Sec. 3.4.3.1.

The general solution of the master equation is employed in Sec. 3.5 to discuss general properties of the state evolution of the QBM subsystem, tending to a Gaussian stationary state at late times. In addition, a generic discussion of late-time dynamics is provided in Sec 3.4.6. A very simple and intuitive picture of

environment-induced decoherence in terms of the reduced Wigner function can be directly extracted, which could easily be made quantitative and precise. Following this, initial-time jolts and slips are discussed.

In Sec. 3.7 we find the exact nonlocal propagator for an Ohmic environment with finite cutoff and identify a new regime at ultra-strong coupling. We provide exact nonperturbative results for the late-time thermal covariance and full-time results for the diffusion coefficients in the large cutoff limit. Explicit examples of sub-Ohmic and supra-Ohmic spectral functions are considered in the following sections, for which the exact propagator is computed and dominant contributions from nonlocal dissipation effects are found (of IR origin in one case and UV in the other). Finally, in Sec. 3.9 we summarize our results and discuss their main implications as well as possible applications.

### 3.3 Phase-Space Review

#### 3.3.1 Classical Evolution

Let  $\mathbf{Z} = (X, P, \dots)$  denote our collective phase-space coordinates. Let  $\rho[\mathbf{Z}; t]$  be the probability distribution associated with phase-space coordinate value  $\mathbf{Z}$  at time  $t$ . For Hamiltonian motion, and even more generally, the distribution must evolve according to classical trajectories:

$$\rho[\mathbf{Z}(t); t] = \rho[\mathbf{Z}(0); 0], \quad (3.1)$$

where  $\mathbf{Z}(t)$  is a classical trajectory which satisfies some classical equations of motion

$$\frac{d}{dt}\mathbf{Z}(t) = \mathcal{F}[\mathbf{Z}(t); t], \quad (3.2)$$

with initial conditions  $\mathbf{Z}(0)$ . For example, in one dimension:

$$\frac{d}{dt}X(t) = \frac{P(t)}{M}, \quad (3.3)$$

$$\frac{d}{dt}P(t) = F[X(t); t]. \quad (3.4)$$

In terms of the initial conditions, future values of the distribution can also be generated with a trajectory unraveling where one sums over all initial weights at locations  $\mathbf{Z}(0)$  which evolve to  $\mathbf{Z}$  in time  $t$ .

$$\rho[\mathbf{Z}, t] = \langle \delta[\mathbf{Z} - \mathbf{Z}(t)] \rangle_{\mathbf{Z}(0)}, \quad (3.5)$$

$$\langle \cdots \rangle_{\mathbf{Z}(0)} \equiv \int d\mathbf{Z}(0) \cdots \rho[\mathbf{Z}(0), 0]. \quad (3.6)$$

One can then derive the *Liouville equation* by considering the total derivative

$$\frac{d}{dt}\rho[\mathbf{Z}(t); t] = \frac{d}{dt}\rho[\mathbf{Z}(0); 0] = 0, \quad (3.7)$$

and so

$$\frac{\partial}{\partial t}\rho[\mathbf{Z}(t); t] + \dot{\mathbf{Z}}(t)^T \nabla_{\mathbf{Z}(t)} \rho[\mathbf{Z}(t); t] = 0. \quad (3.8)$$

Therefore we have the corresponding partial differential equation

$$\frac{\partial}{\partial t}\rho[\mathbf{Z}; t] = -\mathcal{F}[\mathbf{Z}; t]^T \nabla_{\mathbf{Z}} \rho[\mathbf{Z}; t]. \quad (3.9)$$

For example, in one dimension the Liouville equation is simply

$$\frac{\partial}{\partial t}\rho[X, P; t] = -\left\{ \frac{P}{M} \frac{\partial}{\partial X} + F[X; t] \frac{\partial}{\partial P} \right\} \rho[X, P; t]. \quad (3.10)$$

Vice versa, first-order PDE's can be solved by transforming them into a system of ODE's which govern the evolution of evolving coordinates. This is called the *method of characteristic curves*. Our characteristic curves here are the classical trajectories.

Quantum theory will differ in two respects: The corresponding states  $\rho \rightarrow W$  are no longer probability distributions but pseudo-distributions, which encode features such as coherence and respect the Heisenberg uncertainty principle. Secondly, the corresponding quantum master equation is not generally linear in derivatives, thus not admitting trajectories or “characteristics”.

### 3.3.1.1 Markovian Fokker-Planck equation

To give a simple example of classical open-system dynamics, one can consider the Fokker-Planck equation

$$\frac{\partial}{\partial t} \rho[X, P; t] = \left\{ -\frac{P}{M} \frac{\partial}{\partial X} - F(X) \frac{\partial}{\partial P} + 2\gamma_0 \frac{\partial}{\partial P} P + 2M\gamma_0 k_B T \frac{\partial^2}{\partial P^2} \right\} \rho[X, P; t], \quad (3.11)$$

The new environmentally-induced generators are

$$2\gamma_0 P \frac{\partial}{\partial P} \quad \text{damping force} \quad (3.12)$$

$$2\gamma_0 \quad \text{probability preservation (to counteract damping)} \quad (3.13)$$

$$2M\gamma_0 k_B T \frac{\partial^2}{\partial P^2} \quad \text{diffusion} \quad (3.14)$$

The forces alone are first order and would admit trajectories, but the diffusion generator is second-order and thus generates diffusion which smears the trajectories. Note that the diffusion coefficient contains both the temperature and the damping con-

stant. This proportionality is the classical (and Markovian) *fluctuation-dissipation relation* (FDR).

Let us express the Fokker-Plank equation

$$\frac{\partial}{\partial t} \rho[X, P; t] = \mathcal{L} \rho[X, P; t], \quad (3.15)$$

where  $\mathcal{L}$  is the Liouville operator or *Liouvillian*, then one can easily show

$$\mathcal{L} e^{-\frac{H(X,P)}{k_B T}} = 0 \quad (3.16)$$

$$H(X, P) \equiv \frac{P^2}{2M} + V(X) \quad (3.17)$$

and so the dissipation and diffusion work together as to relax the system into a Boltzmann thermal state.

### 3.3.2 Quantum States

Any mixed quantum state  $\rho$  (density matrix) can be expressed as a convex combination of orthogonal pure quantum states  $\psi_k$  (wave functions) as

$$\rho = \sum_k p_k |\psi_k\rangle\langle\psi_k|. \quad (3.18)$$

The density matrix has position representation  $\rho(X, X') = \langle X | \rho | X' \rangle$  and momentum representation  $\rho(P, P') = \langle P | \rho | P' \rangle$ . The two representations are related by a double Fourier transformation. But with a single Fourier transform one can also transform to the phase-space representation [81]:

$$W(X, P) = \frac{1}{2\pi\hbar} \int d\Delta e^{+i\frac{P\Delta}{\hbar}} \langle X - \Delta/2 | \rho | X + \Delta/2 \rangle. \quad (3.19)$$

$W$  denotes the *Wigner function*; it is considered *the* phase-space representation (and not *a* phase-space representation) of the density matrix  $\rho$  because of its many unique

and important properties. For instance, it has the same marginal distributions as the wave function

$$\int dP W(X, P) = |\psi(X)|^2 \quad (3.20)$$

$$\int dX W(X, P) = |\tilde{\psi}(P)|^2 \quad (3.21)$$

and thus it will have the same position and momentum moments (separately). The arguments  $X$  and  $P$  do not correspond to the position and momentum operators, but more correctly to their symmetrized moments. For example

$$\iint dX dP W(X, P) X P = \left\langle \frac{\mathbf{X P} + \mathbf{P X}}{2} \right\rangle. \quad (3.22)$$

The noncommuting information is still present, though it is not easily accessible in this representation. Similarly, the position representation also contains all momentum information and vice versa.

Unlike the classical phase-space distribution, the Wigner function is only a *pseudo-distribution*: it can take negative values. This is a typical feature of quantum states, for example consider the superposition of states in Fig 3.1. The Wigner function is also more restricted in how localized it can be. Singular distributions (which would correspond to individual classical trajectories) are strictly forbidden, as are any distributions which would violate the Heisenberg uncertainty principle.

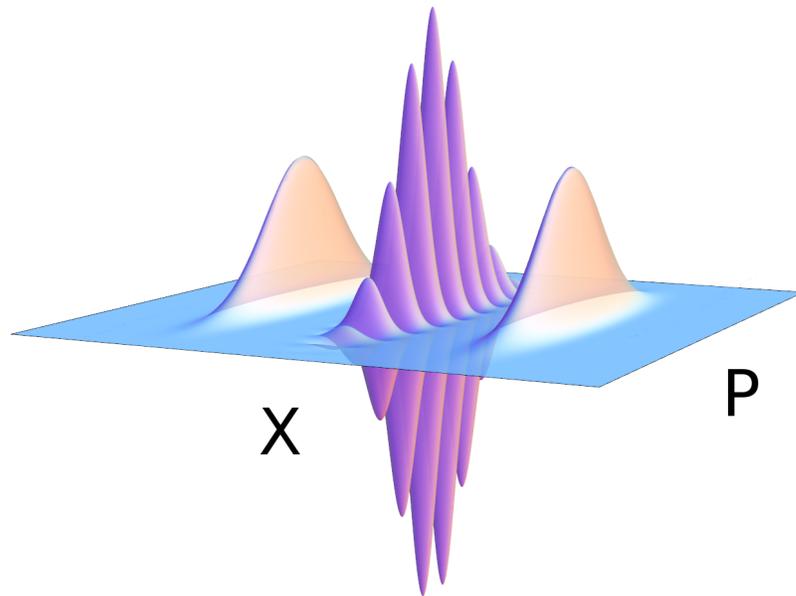


Figure 3.1: Wigner function corresponding to the coherent quantum superposition of two Gaussian wavefunctions in position space shifted by a distance. A classical mixture of two wavefunctions would lack the interference pattern.

### 3.3.3 Quantum Evolution

Fourier transforming the quantum master equation

$$\frac{d}{dt}\boldsymbol{\rho}(t) = -\frac{i}{\hbar} [\mathbf{H}, \boldsymbol{\rho}(t)] , \quad (3.23)$$

$$\mathbf{H} \equiv \frac{\mathbf{P}^2}{2M} + V(\mathbf{X}) , \quad (3.24)$$

one can derive the quantum Liouville equation

$$\frac{\partial}{\partial t} W[X, P; t] = \left\{ -\frac{P}{M} \frac{\partial}{\partial X} - F(X) \frac{\partial}{\partial P} + \sum_{\substack{n>1 \\ (\text{odd})}} \left( \frac{\hbar}{2i} \right)^{n-1} \frac{1}{n!} V^{[n]}(X) \frac{\partial^n}{\partial P^n} \right\} W[X, P; t] . \quad (3.25)$$

The terms zeroth order in  $\hbar$  are classical and would admit classical trajectories in their solution. The higher-order terms in  $\hbar$  appear with higher-order partial derivatives and thus this PDE no longer admits characteristic curves (unless the system is linear). The nonlinear derivatives do not generate diffusion, as in Eq. (3.11), but *quantum deformation*.

If the system potential is non-polynomial then the sum of quantum generators in Eq. (3.25) will not naturally truncate. Closed-form expressions are more naturally represented with nonlocal forces, e.g. with the classical to quantum substitution

$$F(X) \frac{\partial}{\partial P} W(X, P) \rightarrow \iint dX' dP' F(X - X') \delta_{\hbar}(X', P') \frac{\partial}{\partial P} W(X, P - P') , \quad (3.26)$$

in terms of the quantum pseudo-distribution function

$$\delta_{\hbar}(X, P) = -\frac{\text{Ci}\left(\frac{|XP|}{\hbar}\right)}{2\pi\hbar} , \quad (3.27)$$

where  $\text{Ci}(z)$  denotes the cosine integral function. This pseudo-distribution has the

limit

$$\lim_{\hbar \rightarrow 0} \delta_{\hbar}(X, P) = \delta(X) \delta(P), \quad (3.28)$$

and so this representation admits yet another definition of the classical limit. This pedagogically-appealing representation is a straightforward and simple consequence of basic results in [81], though we are not aware of any previous derivation of relation (3.27) in particular.

### 3.4 Quantum Brownian Motion

In quantum Brownian motion (QBM) we take the combined quantum system  $\mathbf{Z} = (\mathbf{X}, \mathbf{P})$  and environment  $\mathbf{z} = (\mathbf{x}, \mathbf{p})$  to be linear. Thus their closed-system dynamics appear classical in the phase-space representation. Quantum effects still arise in this theory due to the non-classical initial state. We consider the system + environment Hamiltonian with position-position coupling, as to model a damped oscillator.

$$\mathbf{H}_C = \mathbf{H}_S(\mathbf{X}, \mathbf{P}) + \mathbf{H}_E(\mathbf{x}, \mathbf{p}) + \mathbf{H}_I(\mathbf{X}, \mathbf{x}) + \mathbf{H}_R(\mathbf{X}), \quad (3.29)$$

$$\mathbf{H}_C = \frac{1}{2} (\mathbf{P}^T \mathbf{M}^{-1} \mathbf{P} + \mathbf{X}^T \mathbf{C} \mathbf{X}) + \frac{1}{2} (\mathbf{p}^T \mathbf{m}^{-1} \mathbf{p} + \mathbf{x}^T \mathbf{c} \mathbf{x}) - \mathbf{x}^T \mathbf{g} \mathbf{X} + \mathbf{H}_R(\mathbf{X}), \quad (3.30)$$

where  $\mathbf{M}$  denotes the system masses,  $\mathbf{C}$  denotes the system stiffness or spring constants,  $\mathbf{m}$  denotes the environment masses,  $\mathbf{c}$  denotes the environment spring constants, and  $\mathbf{g}$  denotes the system-environment coupling constants. It is known from the influence functional formalism [57], that such a model environment can emulate any source of Gaussian noise with proper choice of coupling. Furthermore, we do not

assume that the system spring constant  $\mathbf{C}$  is diagonalized, but only positive-definite, real, symmetric, etc..

Our choice of renormalization will be equivalent to inserting the entire system-environment interaction in the square of the potential.

$$\mathbf{H}_C = \frac{1}{2} (\mathbf{P}^T \mathbf{M}^{-1} \mathbf{P} + \mathbf{X}^T \mathbf{C} \mathbf{X}) + \frac{1}{2} \left( \mathbf{p}^T \mathbf{m}^{-1} \mathbf{p} + [\mathbf{x} - \mathbf{c}^{-1} \mathbf{g} \mathbf{X}]^T \mathbf{c} [\mathbf{x} - \mathbf{c}^{-1} \mathbf{g} \mathbf{X}] \right), \quad (3.31)$$

as this keeps the phenomenological system-system interaction from changing, regardless of what system-environment coupling  $\mathbf{g}$  we choose.

### 3.4.1 The Langevin Equation

Given that our theory is linear, the quantum Fokker-Planck equation of the closed system + environment is identical to the classical Fokker-Planck equation. Therefore the characteristic curves which solve our quantum system + environment are simply the classical trajectories. The quantum fluctuations induced by the environment will not enter until we coarse grain over the (quantum) state of the environment.

Our characteristic curves are therefore generated by the classical equations of motion

$$\mathbf{M} \ddot{\mathbf{X}}(t) + (\mathbf{C} + \delta \mathbf{C}) \mathbf{X}(t) = \mathbf{g}^T \mathbf{x}(t), \quad (3.32)$$

$$\mathbf{m} \ddot{\mathbf{x}}(t) + \mathbf{c} \mathbf{x}(t) = \mathbf{g} \mathbf{X}(t), \quad (3.33)$$

in terms of the system renormalization shift

$$\delta \mathbf{C} \equiv \mathbf{g}^T \mathbf{c}^{-1} \mathbf{g}. \quad (3.34)$$

We may solve the environment equation of motion in terms of the system trajectory as

$$\mathbf{x}(t) = \left( \mathbf{f}(t) \mathbf{m} \mathbf{x}_0 + \dot{\mathbf{f}}(t) \mathbf{p}_0 \right) + \mathbf{f}(t) * \mathbf{g} \mathbf{X}(t), \quad (3.35)$$

$$\mathbf{f}(t) \equiv \mathbf{m}^{-\frac{1}{2}} \frac{\sin(\boldsymbol{\omega} t)}{\boldsymbol{\omega}} \mathbf{m}^{-\frac{1}{2}}, \quad (3.36)$$

$$\boldsymbol{\omega}^2 \equiv \mathbf{m}^{-\frac{1}{2}} \mathbf{c} \mathbf{m}^{-\frac{1}{2}}, \quad (3.37)$$

where  $\mathbf{f}(t)$  is the free Green's function (position propagator) of the environment and  $*$  denotes the Laplace convolution

$$A(t) * B(t) = (A * B)(t) \equiv \int_0^t d\tau A(t-\tau) B(\tau), \quad (3.38)$$

which is equivalent to the  $\cdot$  product

$$A(t) \cdot B(t) = (A \cdot B)(t) \equiv \int_0^t d\tau A(\tau) B(\tau), \quad (3.39)$$

for stationary kernels. E.g., here we can define  $\mathbf{f}(t, \tau) = \mathbf{f}(t-\tau)$  such that  $\mathbf{f} * \mathbf{g} \mathbf{X} = \mathbf{f} \cdot \mathbf{g} \mathbf{X}$ . For the moment we have assumed the environment parameters to be constant in time. However, we can generalize the final result appropriately by considering all valid Gaussian influences.

Environment solutions may then be substituted back into the system equations of motion to obtain the Langevin equation

$$\mathbf{M} \ddot{\mathbf{X}}(t) + 2(\boldsymbol{\mu} \cdot \mathbf{X})(t) + (\mathbf{C} + \boldsymbol{\delta} \mathbf{C}) \mathbf{X}(t) = \boldsymbol{\xi}(t), \quad (3.40)$$

in terms of the dissipation kernel  $\boldsymbol{\mu}(t, \tau)$  and stochastic processes  $\boldsymbol{\xi}(t)$ . For constant linear coupling to an environment of harmonic oscillators, the dissipation kernel is

stationary  $\boldsymbol{\mu}(t, \tau) = \boldsymbol{\mu}(t - \tau)$  and these objects resolve to

$$\boldsymbol{\mu}(t) \equiv -\frac{1}{2} \mathbf{g}^T \mathbf{f}(t) \mathbf{g}, \quad (3.41)$$

$$\boldsymbol{\xi}(t) \equiv \mathbf{g}^T \left( \mathbf{f}(t) \mathbf{m} \mathbf{x}_0 + \dot{\mathbf{f}}(t) \mathbf{p}_0 \right). \quad (3.42)$$

Note that Eq. (3.40) is a classical-like Langevin equation which describes stochastic trajectories (coordinate unravelings) in the phase-space representation. This is different from the *quantum Langevin equation* [67], wherein one follows the same procedure for the Heisenberg equations of motion and obtains stochastic equations of motion for the system operators. The latter formulation is even possible for non-linear systems, though the stochastic equations of motion will no longer be exactly solvable.

The stochastic process  $\boldsymbol{\xi}$  has a Gaussian distributional with noise correlation

$$\boldsymbol{\nu}(t, \tau) = \langle \boldsymbol{\xi}(t) \boldsymbol{\xi}^T(\tau) \rangle_{\boldsymbol{\xi}}. \quad (3.43)$$

Coarse graining over the environment is equivalent to the stochastic noise average in this representation (and also equivalent that of [30], as derived from the path-integral formalism)

$$\langle \dots \rangle_{\boldsymbol{\xi}} = \frac{1}{\sqrt{2\pi \det(\boldsymbol{\nu})}} \int \mathcal{D}\boldsymbol{\xi} \dots e^{-\frac{1}{2} \boldsymbol{\xi}^T \cdot \boldsymbol{\nu}^{-1} \cdot \boldsymbol{\xi}}. \quad (3.44)$$

Inserting (3.42) into (3.44), the noise kernel resolves to

$$\boldsymbol{\nu}(t, \tau) = \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\coth\left(\frac{\omega}{2T}\right) \cos(\omega[t - \tau])}{2\omega} \mathbf{m}^{-\frac{1}{2}} \mathbf{g}, \quad (3.45)$$

for constant linear coupling to a thermal reservoir of harmonic oscillators. The noise kernel is stationary,  $\boldsymbol{\nu}(t, \tau) = \boldsymbol{\nu}(t - \tau)$ , because the environment is initially in a stationary state.

In general, for any Gaussian environment, and not necessarily the one we have specified here with constant mass and spring constants, the two kernels are constrained by the fact that the environment correlation or influence kernel

$$\boldsymbol{\alpha}(t, \tau) = \boldsymbol{\nu}(t, \tau) + \imath \boldsymbol{\mu}(t, \tau), \quad (3.46)$$

must be positive definite in addition to the noise kernel, as discussed in Sec. 2.5.1.1. The Langevin equation in Eq. (3.40) is written in such a way that it is completely general for any Gaussian influence.

Moreover, it is useful to turn to the phase-space representation, defined

$$\mathbf{Z} \equiv (\mathbf{X}, \mathbf{P}), \quad (3.47)$$

$$\boldsymbol{\Xi} \equiv (\mathbf{0}, \boldsymbol{\xi}), \quad (3.48)$$

$$\mathbf{N}(t, \tau) \equiv \begin{bmatrix} \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \boldsymbol{\nu}(t, \tau) \end{bmatrix}, \quad (3.49)$$

so that we can express the Langevin equation as the first-order integro-differential equation

$$\frac{d}{dt} \mathbf{Z}(t) + (\mathbf{H} \cdot \mathbf{Z})(t) = \boldsymbol{\Xi}(t), \quad (3.50)$$

where the nonlocal homogeneous generator is defined

$$\mathbf{H}(t, \tau) \equiv \begin{bmatrix} \mathbf{0} & -\delta(t-\tau) \mathbf{M}^{-1} \\ \delta(t-\tau)(\mathbf{C} + \boldsymbol{\delta}\mathbf{C}) + 2\boldsymbol{\mu}(t, \tau) & \mathbf{0} \end{bmatrix}. \quad (3.51)$$

### 3.4.1.1 The Damping Kernel

Dissipation may also be represented via the *damping kernel*, defined by the relation

$$\boldsymbol{\mu}(t, \tau) \equiv -\frac{\partial}{\partial \tau} \boldsymbol{\gamma}(t, \tau), \quad (3.52)$$

which straightforwardly resolves to the microscopic definition

$$\boldsymbol{\gamma}(t, \tau) = \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\cos(\boldsymbol{\omega}[t-\tau])}{2\boldsymbol{\omega}^2} \mathbf{m}^{-\frac{1}{2}} \mathbf{g}, \quad (3.53)$$

for constant linear coupling to an environment of simple harmonic oscillators, and in which case the damping kernel is positive definite in the sense of

$$\int_0^t d\tau_1 \int_0^t d\tau_2 \mathbf{V}^T(\tau_1) \boldsymbol{\gamma}(\tau_1, \tau_2) \mathbf{V}(\tau_2) \geq 0. \quad (3.54)$$

Integration by parts then relates their contributions to the Langevin equation.

$$\underbrace{2(\boldsymbol{\mu} \cdot \mathbf{X})(t)}_{\text{dissipation}} + \underbrace{\delta \mathbf{C} \mathbf{X}(t)}_{\text{renormalization}} = \underbrace{2(\boldsymbol{\gamma} \cdot \dot{\mathbf{X}})(t)}_{\text{damping}} + \underbrace{2\boldsymbol{\gamma}(t, 0) \mathbf{X}(0)}_{\text{slip}}. \quad (3.55)$$

The *slip* is a transient artifact of our factorized initial conditions and will be discussed further in Sec. 3.5.2.1. It is a highly-singular position-dependent impulse which rapidly distorts the initial state of the system soon after coupling to the environment.

In terms of the damping kernel, the Langevin equation can now be expressed

$$\mathbf{M} \ddot{\mathbf{X}}(t) + 2(\boldsymbol{\gamma} \cdot \dot{\mathbf{X}})(t) + \mathbf{C} \mathbf{X}(t) + 2\boldsymbol{\gamma}(t, 0) \mathbf{X}_0 = \boldsymbol{\xi}(t), \quad (3.56)$$

and so our choice of renormalization is justified by this equation of a nonlocally damped oscillator with phenomenological spring constant  $\mathbf{C}$ . If positive definite, as

will be the case for constant coupling to a thermal reservoir, the nonlocal damping truly generates dissipation, as can be seen when integrating each contribution to the Langevin equation upon multiplication with the velocity.

$$\dot{\mathbf{X}}^T(t) \cdot \mathbf{M} \ddot{\mathbf{X}}(t) = \frac{1}{2} \dot{\mathbf{X}}^T(t) \mathbf{M} \dot{\mathbf{X}}(t), \quad (3.57)$$

$$\dot{\mathbf{X}}^T(t) \cdot 2(\boldsymbol{\gamma} \cdot \dot{\mathbf{X}})(t) = (\dot{\mathbf{X}}^T \cdot \boldsymbol{\gamma} \cdot \dot{\mathbf{X}})(t), \quad (3.58)$$

$$\dot{\mathbf{X}}^T(t) \cdot \mathbf{C} \mathbf{X}(t) = \frac{1}{2} \mathbf{X}^T(t) \mathbf{C} \mathbf{X}(t). \quad (3.59)$$

These terms correspond to the kinetic energy, cumulative dissipated energy (a strictly non-negative quantity), and potential energy. If each contribution is positive, then the damping can never create a net increase of energy in the system.

### 3.4.1.2 Local Damping

Local damping is defined as

$$\boldsymbol{\gamma}(t-\tau) = 2\boldsymbol{\gamma}_0 \delta(t-\tau), \quad (3.60)$$

$$\tilde{\boldsymbol{\gamma}}(\omega) = 2\boldsymbol{\gamma}_0 \quad (3.61)$$

$$\hat{\boldsymbol{\gamma}}(s) = \boldsymbol{\gamma}_0 \quad (3.62)$$

which produces the local damping force

$$(\boldsymbol{\gamma} * \dot{\mathbf{X}})(t) = \boldsymbol{\gamma}_0 \dot{\mathbf{X}}(t), \quad (3.63)$$

receiving only half the weight of the distribution in the Laplace convolution. One can determine what microscopic model will produce such damping by inspecting Eq. (3.53) in the Fourier domain

$$\tilde{\boldsymbol{\gamma}}(\varepsilon) = \frac{\pi}{2} \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\delta(|\varepsilon| - \boldsymbol{\omega})}{\boldsymbol{\omega}^2} \mathbf{m}^{-\frac{1}{2}} \mathbf{g}, \quad (3.64)$$

and then relating this to Eq. (3.60), to find that local damping results from

$$\mathbf{I}(\varepsilon) \equiv \mathbf{g}^T \mathbf{m}^{-\frac{1}{2}} \frac{\delta(|\varepsilon| - \omega)}{2\omega} \mathbf{m}^{-\frac{1}{2}} \mathbf{g} = \frac{2}{\pi} \gamma_0 \varepsilon, \quad (3.65)$$

which is known as *Ohmic coupling*.  $\mathbf{I}(\omega)$  is known as the *spectral-density function* and it is equivalent to the damping kernel when defined in this manner.

$$\mathbf{I}(\omega) \equiv \frac{1}{\pi} \tilde{\gamma}(\omega) \omega. \quad (3.66)$$

Therefore, quite generally, finite damping results from infinitesimal coupling to an uncountable number of environment degrees of freedom.

Quantum mechanically, local damping will not produce a fully acceptable model, even when ignoring all transient behaviors. The lack of a U.V. cutoff, combined with the zero-point fluctuations of the quantum environment, will be shown to produce infinite diffusion. Instead of truly local damping, one typically considers a U.V. regulated Ohmic coupling of the form

$$\tilde{\gamma}(\omega) = 2\gamma_0 \chi(\omega/\Lambda), \quad (3.67)$$

where  $\Lambda$  denotes the U.V. cutoff and  $\chi : [0, \infty) \rightarrow [1, 0)$  denotes the regulator. The homogeneous evolution of a damped oscillator regulated in this manner will only differ from a simple damped oscillator by very small amounts, in terms of timescales, amplitude and phase, with corrections of the order  $\mathcal{O}(\Omega/\Lambda)$ . Furthermore, the regulated Ohmic coupling will only display a logarithmic cutoff sensitivity in its late-time diffusion. This cutoff sensitivity is not renormalizable (see Sec. 3.7.2) and can be physically motivated in the few models we consider.

### 3.4.1.3 The Fluctuation-Dissipation Relation

Restating some results of Sec. 2.5.3, we may compare the noise and damping kernels for constant coupling to a thermal environment which admits stationary kernels. We then have the fluctuation-dissipation relation (FDR), expressed most simply as

$$\tilde{\nu}(\omega) = \tilde{\kappa}(\omega) \tilde{\gamma}(\omega), \quad (3.68)$$

$$\tilde{\kappa}(\omega) \equiv \hbar\omega \coth\left(\frac{\hbar\omega}{2k_{\text{B}}T}\right), \quad (3.69)$$

with the Fourier transform defined

$$\tilde{f}(\omega) \equiv \int_{-\infty}^{+\infty} dt e^{-i\omega t} f(t). \quad (3.70)$$

$\tilde{\kappa}$  is the (quantum) FDR kernel which relates the fluctuations and dissipation. The classical or high-temperature FDR kernel is given by

$$\lim_{\hbar \rightarrow 0} \tilde{\kappa}(\omega) = \lim_{T \rightarrow \infty} \tilde{\kappa}(\omega) = 2k_{\text{B}}T, \quad (3.71)$$

which encapsulates the Einstein relation for Brownian motion. For local damping  $\gamma(t-\tau) = 2\gamma_0 \delta(t-\tau)$  and high temperature one has

$$\nu(t, \tau) = 4\gamma_0 k_{\text{B}}T \delta(t-\tau) \quad (3.72)$$

which describes *white noise* and more corresponds to the original models of Einstein and Langevin. But note that one can have nonlocal dissipation and a non-Markovian noise process even classically.

### 3.4.2 Evolution of Stochastic Trajectories

Let us now consider solutions the QBM Langevin equation (3.50):

$$\dot{\mathbf{Z}}(t) + (\mathbf{H} \cdot \mathbf{Z})(t) = \Xi(t), \quad (3.73)$$

expressed succinctly in the phase-space representation. For the moment we consider the most general regime wherein all coefficients may be time dependent and all memory kernels may be nonlocal and even non-stationary. All solutions can be represented

$$\mathbf{Z}(t) = \Phi(t) \mathbf{Z}(0) + (\Phi_i \cdot \Xi)(t), \quad (3.74)$$

where  $\Phi(t)$  is the single-time *homogeneous propagator* or transition matrix such that

$$\frac{d}{dt} \Phi(t) = -(\mathbf{H} \cdot \Phi)(t), \quad (3.75)$$

$$\Phi(0) = \mathbf{1}, \quad (3.76)$$

and  $\Phi_i(t, \tau)$  is the *initial-value propagator* such that

$$\frac{\partial}{\partial t} \Phi_i(t, \tau) = -(\mathbf{H} \cdot \Phi_i)(t, \tau), \quad (3.77)$$

$$\Phi_i(t, t) = \mathbf{1}. \quad (3.78)$$

It is important to note that, in general, the initial-value propagator is not so easily determined from the *two-time homogeneous propagator*:

$$\Phi(t, \tau) \equiv \Phi(t) \Phi^{-1}(\tau). \quad (3.79)$$

We will demonstrate this explicitly in Sec. 3.4.2.1 by characterizing these propagators in various regimes.

There is a history of mistakes in the QBM literature, for nonlocal damping, all related to the subtle difference between the various propagators of integro-differential equations. By definition, one can always relate the nonlocal dynamics to an equivalent local dynamics, e.g.

$$\mathcal{H}(t) \equiv -\dot{\Phi}(t) \Phi^{-1}(t), \quad (3.80)$$

and these are, in fact, the coefficients one sees in the time-local master equation. However, when the Langevin equation is nonlocal,  $\mathcal{H}(t)$  can only be used to generate the homogeneous propagation. With the introduction of identical forces, whether stochastic or external, into the previously equivalent equations

$$\dot{\Phi}(t) = -(\mathbf{H} \cdot \Phi)(t), \quad (3.81)$$

$$\dot{\Phi}(t) = -\mathcal{H}(t) \Phi(t), \quad (3.82)$$

the driven solutions will no longer be the same. As we demonstrated in [64], the external forces and diffusion present in the time-local master equation are not directly related to those in the Hamiltonian and Langevin equation. In fact, they are convoluted in such a manner as to produce the same driven solutions.

### 3.4.2.1 Characterization of the Propagators

Most simply, one can consider local damping and time-constant coefficients, in which case the Langevin equation simplifies to

$$\mathbf{H}(t, \tau) = \delta(t - \tau) \mathcal{H}, \quad (3.83)$$

$$\mathcal{H} = \begin{bmatrix} \mathbf{0} & -\mathbf{M}^{-1} \\ \mathbf{C} & 2\gamma_0 \mathbf{M}^{-1} \end{bmatrix}, \quad (3.84)$$

and the homogeneous propagator is then formally determined by the matrix exponential

$$\Phi(t) = e^{-t\mathcal{H}}, \quad (3.85)$$

and so we have a system of simple damped oscillators. In this case the initial-value propagator is given by

$$\Phi_i(t, \tau) = \Phi(t, \tau) = \Phi(t - \tau). \quad (3.86)$$

For local damping and time-dependent coefficients, the homogeneous propagator then must satisfy a parametric ODE.

$$\mathbf{H}(t, \tau) = \delta(t - \tau) \mathcal{H}(t), \quad (3.87)$$

$$\dot{\Phi}(t) = -\mathcal{H}(t) \Phi(t). \quad (3.88)$$

There is no general method for calculating  $\Phi(t)$  in closed form, but once determined the initial-value propagator is well-known to be given by

$$\Phi_i(t, \tau) = \Phi(t, \tau) \neq \Phi(t - \tau). \quad (3.89)$$

For nonlocal damping with a stationary damping kernel and otherwise time-constant coefficients, the Langevin equation can be formally solved by Laplace transformation

$$\mathbf{H}(t, \tau) = \mathbf{H}(t - \tau), \quad (3.90)$$

$$\hat{\Phi}(s) = [s + \hat{\mathbf{H}}(s)]^{-1}, \quad (3.91)$$

where the Laplace transform is defined

$$\hat{f}(s) \equiv \int_0^\infty dt e^{-st} f(t). \quad (3.92)$$

The initial-value propagator can also be determined by Laplace transform to be

$$\Phi_i(t, \tau) = \Phi(t - \tau) \neq \Phi(t, \tau), \quad (3.93)$$

as proven in Sec. 3.4.5. Thus, one can deduce that the more general relation must be highly nontrivial.

Finally we will propagate from future times. This is, in fact, where most previous derivations of the HPZ equation failed for nonlocal damping, as they relied upon  $\Phi$  being the “advanced” propagator given that it is the retarded propagator for constant coefficients. Working from the initial-value solutions (3.74) we have

$$\Phi(\tau, t) \mathbf{z}(t) = \Phi(\tau) \mathbf{z}(0) + \int_0^t d\tau' \Phi(\tau, t) \Phi_i(t, \tau') \Xi(\tau'), \quad (3.94)$$

$$= \mathbf{z}(\tau) - \int_0^\tau d\tau' \Phi_i(\tau, \tau') \Xi(\tau') + \int_0^t d\tau' \Phi(\tau, t) \Phi_i(t, \tau') \Xi(\tau'), \quad (3.95)$$

and so the final-value solutions are given by

$$\mathbf{z}(\tau) = \Phi(\tau, t) \mathbf{z}(t) - \int_0^t d\tau' \Phi_f(\tau, \tau') \Xi(\tau'), \quad (3.96)$$

in terms of the *final-value propagator*:

$$\Phi_f(\tau, \tau') = \Phi(\tau, t) \Phi_i(t, \tau') - \theta(\tau - \tau') \Phi_i(\tau, \tau'). \quad (3.97)$$

Note that  $\Phi(t, \tau) \neq \Phi(t - \tau)$  unless one has local damping, and so the final-value propagator does not reduce to the ordinary *advanced propagator*  $\theta(\tau' - \tau) \Phi(\tau, \tau')$  unless one has local damping. This means that, unlike with ordinary differential equations, when boundary conditions  $\mathbf{Z}(t)$  are specified at a final time  $t$ , there is no truly *advanced* propagator for the inhomogeneous solutions of the integro-differential equation. Such mathematical subtleties of final-value problems for integro-differential equations have been missed in the existing literature on the derivation of the master equation for QBM models and could lead to significant discrepancies whenever the nonlocal effects of dissipation are important.

### 3.4.2.2 Two-Time Correlations

Using the initial-value solution of the Langevin equation given by Eq. (3.74) and applying the Gaussian average defined by (3.44), it is straightforward to calculate quantum correlations between system observables at different times. For instance, the symmetrized two-point quantum correlation function for position and momentum operators is given by

$$\sigma(t_1, t_2) \equiv \langle \mathbf{Z}(t_1) \mathbf{Z}^T(t_2) \rangle_{\Xi}. \quad (3.98)$$

Applying our initial-value solution and the noise average results in

$$\boldsymbol{\sigma}(t_1, t_2) = \boldsymbol{\Phi}(t_1) \boldsymbol{\sigma}(0) \boldsymbol{\Phi}^T(t_2) + \boldsymbol{\sigma}_T(t_1, t_2), \quad (3.99)$$

$$\boldsymbol{\sigma}_T(t_1, t_2) = (\boldsymbol{\Phi}_1 \cdot \mathbf{N} \cdot \boldsymbol{\Phi}_1^T)(t_1, t_2). \quad (3.100)$$

The first term describes the oscillatory decay of the initial covariance, while the second term describes the emergence of a thermally-generated covariance. The single-time correlations are then given by the coincidence limit

$$\boldsymbol{\sigma}(t) = \boldsymbol{\sigma}(t, t), \quad (3.101)$$

$$\boldsymbol{\sigma}_T(t) = \boldsymbol{\sigma}_T(t, t). \quad (3.102)$$

### 3.4.3 Evolution of States

To determine the time evolution of the Wigner function, we express the evolution of the system and environment (Sec. 3.3.1) in the stochastic representation

$$W[\mathbf{Z}, t; \boldsymbol{\Xi}] = \langle \delta[\mathbf{Z} - \mathbf{Z}(t; \boldsymbol{\Xi})] \rangle_{\mathbf{Z}(0)}, \quad (3.103)$$

$$\langle \cdots \rangle_{\mathbf{Z}(0)} \equiv \int d\mathbf{Z}(0) \cdots W[\mathbf{Z}(0), 0], \quad (3.104)$$

with all noise dependence explicitly noted. These stochastic unravelings are then totaled in the noise average to determine the marginal state of the open system

$$W[\mathbf{Z}, t] = \langle W[\mathbf{Z}, t; \boldsymbol{\Xi}] \rangle_{\boldsymbol{\Xi}}, \quad (3.105)$$

$$\langle \cdots \rangle_{\boldsymbol{\Xi}} \equiv \frac{1}{\sqrt{2\pi \det(\mathbf{N})}} \int \mathcal{D}\boldsymbol{\Xi} \cdots e^{-\frac{1}{2} \boldsymbol{\Xi}^T \cdot \mathbf{N}^{-1} \cdot \boldsymbol{\Xi}}. \quad (3.106)$$

The state of the open system is therefore given by the double average

$$W[\mathbf{Z}, t] = \langle \langle \delta[\mathbf{Z} - \mathbf{Z}(t)] \rangle \rangle_{\mathbf{Z}(0), \boldsymbol{\Xi}}. \quad (3.107)$$

Next we introduce the Fourier transform of the Wigner function defined

$$\mathcal{W}(\mathbf{K}) \equiv \iint_{\mathbb{R}^{2N}} d\mathbf{Z} e^{-i\mathbf{K}^T \mathbf{Z}} W(\mathbf{Z}), \quad (3.108)$$

where  $\mathbf{K}$  is conjugate to  $\mathbf{Z}$ . As the Wigner function is a pseudo-distribution, its Fourier transform is a characteristic or moment generating function with the moment-generating property

$$i^n \frac{\partial^n \mathcal{F}\{f\}}{\partial K_j^n}(\mathbf{0}) = \iint d\mathbf{Z} Z_j^n f(\mathbf{Z}). \quad (3.109)$$

The characteristic function then trivially resolves to

$$\mathcal{W}[\mathbf{K}, t] = \int d\mathbf{z} e^{-i\mathbf{K}^T \mathbf{z}} \langle\langle \delta[\mathbf{Z} - \mathbf{Z}(t)] \rangle\rangle_{\mathbf{Z}(0), \Xi}, \quad (3.110)$$

$$= \left\langle \left\langle e^{-i\mathbf{K}^T \mathbf{z}(t)} \right\rangle \right\rangle_{\mathbf{Z}(0), \Xi}, \quad (3.111)$$

at which point we introduce our Langevin equation solutions from (3.74) to obtain

$$\mathcal{W}[\mathbf{K}, t] = \left\langle e^{-i\mathbf{K}^T \Phi(t) \mathbf{Z}(0)} \right\rangle_{\mathbf{Z}(0)} \left\langle e^{-i\mathbf{K}^T (\Phi_i \cdot \Xi)(t)} \right\rangle_{\Xi}. \quad (3.112)$$

The average over initial conditions reduces to the definition of the characteristic function of the initial state, but with homogeneously transformed coordinates. For the noise average, we can complete the square in the Gaussian functional integral. The final result can be expressed

$$\mathcal{W}[\mathbf{K}, t] = \mathcal{W}[\Phi^T(t) \mathbf{K}, 0] e^{-\frac{1}{2} \mathbf{K}^T \boldsymbol{\sigma}_T(t) \mathbf{K}}, \quad (3.113)$$

where  $\boldsymbol{\sigma}_T(t)$  is the thermal covariance in Eq. (3.100).

A straightforward interpretation of the solution immediately follows: the initial cumulants of the system experience damped oscillations due to the homogeneous

propagator  $\Phi(t)$  while a thermal covariance arises, smearing out details of the dissipating initial state. All initial states eventually evolve into a Gaussian state defined by the asymptotic thermal covariance. In the weak-coupling limit (zeroth order) this Gaussian state is Boltzmann. More generally, this state does not look like the thermal state of a free harmonic oscillator because of the non-vanishing interaction with the environment. More precisely it results from the thermal equilibrium state for the system + environment, which gives rise to a non-trivial correlation between them, and tracing out the environment.

Back in the ordinary phase-space representation, this simple product becomes a Fourier convolution. Convolution with a Gaussian function is known as a *Gaussian smearing*. Such an action smears away details smaller than the uncertainty of the covariance. This provides a natural mechanism for decoherence. Quantum features are represented in the Wigner function by  $\hbar$ -dependent interference patterns. A sizable thermal covariance would completely smear these details away.

### 3.4.3.1 Quantum Fokker-Plank Equation

The quickest method of calculating the master equation is from the solution (3.113) in the Fourier domain. We evaluate its time derivative to first obtain

$$\frac{d}{dt}\mathcal{W}[\mathbf{K}, t] = \left( \dot{\mathbf{K}}^T(t)\nabla_{\mathbf{K}(t)}\mathcal{W}[\mathbf{K}(t), 0] + \mathcal{W}[\mathbf{K}(t), 0]\frac{d}{dt} \right) e^{-\frac{1}{2}\mathbf{K}^T\boldsymbol{\sigma}_T(t)\mathbf{K}}, \quad (3.114)$$

$$\mathbf{K}(t) \equiv \Phi^T(t)\mathbf{K}. \quad (3.115)$$

Next we express the remaining  $\mathbf{K}$  in terms of  $\mathbf{K}(t)$  and evaluate the last time derivative. To transform the gradients into ordinary gradients, we note the relation

$$\nabla_{\mathbf{K}} f[\mathbf{K}(t)] = \Phi(t) \nabla_{\mathbf{K}(t)} f[\mathbf{K}(t)]. \quad (3.116)$$

After some additional manipulation, we are finally left with the master equation

$$\frac{\partial}{\partial t} \mathcal{W} = \{-\mathbf{K}^T \mathcal{H}(t) \nabla_{\mathbf{K}} - \mathbf{K}^T \mathbf{D}(t) \mathbf{K}\} \mathcal{W}, \quad (3.117)$$

$$\frac{\partial}{\partial t} W = \{\nabla_{\mathbf{Z}}^T \mathcal{H}(t) \mathbf{Z} + \nabla_{\mathbf{Z}}^T \mathbf{D}(t) \nabla_{\mathbf{Z}}\} W, \quad (3.118)$$

given the coefficient matrices

$$\mathcal{H}(t) \equiv -\dot{\Phi}(t) \Phi^{-1}(t), \quad (3.119)$$

$$\mathbf{D}(t) \equiv \frac{1}{2} \{\mathcal{H}(t) \boldsymbol{\sigma}_T(t) + \boldsymbol{\sigma}_T(t) \mathcal{H}^T(t) + \dot{\boldsymbol{\sigma}}_T(t)\}, \quad (3.120)$$

where  $\mathcal{H}$  are the homogeneous coefficients, which contain the time-local system spring constant and dissipation, and  $\mathbf{D}$  are the diffusion coefficients. These phase-space coefficients can be expressed with a more typical block-matrix representation as

$$\mathcal{H} = \begin{bmatrix} \mathbf{0} & -\mathbf{M}^{-1} \\ \mathbf{C}_L & 2\gamma_L \mathbf{M}^{-1} \end{bmatrix}, \quad (3.121)$$

$$\mathbf{D} = \begin{bmatrix} \mathbf{0} & -\frac{1}{2} \mathbf{D}_{\mathbf{XP}} \\ -\frac{1}{2} \mathbf{D}_{\mathbf{XP}}^T & \mathbf{D}_{\mathbf{PP}} \end{bmatrix}, \quad (3.122)$$

where  $\mathbf{C}_L$  is the effective system spring constant,  $\gamma_L$  is the time-local dissipation,  $\mathbf{D}_{\mathbf{PP}}$  is the regular diffusion, and  $\mathbf{D}_{\mathbf{XP}}$  is the anomalous diffusion, which is absent in the Markovian regime (but not necessarily the classical regime [2]). In general,

all of these coefficients can be time dependent. Previous descriptions labeled  $\mathbf{C}_L$  as a “renormalized” system spring constant, however this is rather misleading. The Langevin equation has already informed us that, in the renormalized theory, the spring constant of the open system is unchanged. Rather, the fact that  $\mathbf{C}_L \neq \mathbf{C}$  for nonlocal damping is a consequence of the time-local representation, not phenomenology (except in some late-time limits). This will become far more apparent in the case of sub-Ohmic coupling, Sec. 3.8.

Finally, if the master-equation coefficients have a well-defined stationary limit, then the asymptotic covariance  $\boldsymbol{\sigma}_T^\infty$  is uniquely determined by the Lyapunov equation

$$\mathcal{H}_\infty \boldsymbol{\sigma}_T^\infty + \boldsymbol{\sigma}_T^\infty \mathcal{H}_\infty^\text{T} = 2\mathbf{D}_\infty. \quad (3.123)$$

### 3.4.4 External Forces

In this section we consider the addition of an array of external forces  $\mathbf{F}(t)$  acting on the system oscillator. This is done by including the time-dependent potential

$$\mathbf{H}_{\text{ext}}(t) = -\mathbf{F}^\text{T}(t) \mathbf{X}(t), \quad (3.124)$$

which gives rise to the following additional phase-space source in the Langevin equation (3.73):

$$\mathbf{J}(t) = \begin{bmatrix} 0 \\ \mathbf{F}(t) \end{bmatrix}. \quad (3.125)$$

Following our derivation of solutions in Sec. 3.4.3, based on the solutions of the Langevin equation, can be straightforwardly generalized to this case and one obtains

the following result for the time evolution of the reduced Wigner function:

$$\mathcal{W}[\mathbf{K}, t] = \mathcal{W}[\Phi^T(t) \mathbf{K}, 0] e^{-\frac{1}{2} \mathbf{K}^T \sigma_T(t) \mathbf{K}} e^{-i \mathbf{K}^T \langle \mathbf{Z} \rangle_F(t)}, \quad (3.126)$$

with a driven mean  $\langle \mathbf{Z} \rangle_F(t)$  given by

$$\langle \mathbf{Z} \rangle_F(t) = (\Phi_i \cdot \mathbf{J})(t), \quad (3.127)$$

where  $\Phi_i(t, \tau)$  is the initial-value propagator, Sec. 3.4.2-3.4.2.1.

Following our master equation derivation in Sec. 3.4.3.1, it is easy to see that such a deterministic source in the Langevin equation simply adds a driving term to the master equation, which becomes

$$\frac{\partial}{\partial t} W = \{ \nabla_{\mathbf{Z}}^T \mathcal{H}(t) \mathbf{Z} - \nabla_{\mathbf{Z}}^T \mathbf{J}_{\text{eff}}(t) + \nabla_{\mathbf{Z}}^T \mathbf{D}(t) \nabla_{\mathbf{Z}} \} W, \quad (3.128)$$

where the effective force  $\mathbf{J}_{\text{eff}}(t)$  is given by

$$\mathbf{J}_{\text{eff}}(t) \equiv \mathbf{J}(t) + \int_0^t d\tau \left\{ \left[ \frac{d}{dt} + \mathcal{H}(t) \right] \Phi_i(t, \tau) \right\} \mathbf{J}(\tau), \quad (3.129)$$

and the difference would vanish for local damping. From this relation it is not terribly difficult to show that the local and nonlocal representations are equivalent in the sense

$$\langle \mathbf{z} \rangle_F(t) = \int_0^t d\tau \Phi(t, \tau) \mathbf{J}_{\text{eff}}(\tau) = \int_0^t d\tau \Phi_i(t, \tau) \mathbf{J}(\tau). \quad (3.130)$$

In addition to our discovery in Ref. [63], this discrepancy had also been noticed in Ref. [155]. Unfortunately they did not consider the time-dependence of the diffusion coefficients or they might have noticed the much greater problem.

### 3.4.5 Nonlocal Solutions

Here we give more detail to the solution expressed in Eq. (3.91) for a stationary damping kernel and otherwise constant coefficients. The position Langevin equation (3.56) can be expressed in the Laplace domain as

$$(s^2\mathbf{M} + 2s\hat{\gamma}(s) + \mathbf{C})\hat{\mathbf{X}}(s) = (s\mathbf{M}\mathbf{X}_0 + \mathbf{P}_0) + \hat{\boldsymbol{\xi}}(s), \quad (3.131)$$

and so the solutions are formally determined by inversion to be

$$\hat{\mathbf{X}}(s) = \hat{\mathbf{G}}(s) (s\mathbf{M}\mathbf{X}_0 + \mathbf{P}_0) + \hat{\mathbf{G}}(s) \hat{\boldsymbol{\xi}}(s), \quad (3.132)$$

$$\hat{\mathbf{G}}(s) = [s^2\mathbf{M} + 2s\hat{\gamma}(s) + \mathbf{C}]^{-1}. \quad (3.133)$$

Also useful will be the frequency representation

$$\hat{\mathbf{G}}(s) = \mathbf{M}^{-\frac{1}{2}} [s^2 + 2s\hat{\boldsymbol{\lambda}}(s) + \boldsymbol{\Omega}^2]^{-1} \mathbf{M}^{-\frac{1}{2}}, \quad (3.134)$$

$$\hat{\boldsymbol{\lambda}}(s) \equiv \mathbf{M}^{-\frac{1}{2}} \hat{\gamma}(s) \mathbf{M}^{-\frac{1}{2}}, \quad (3.135)$$

$$\boldsymbol{\Omega}^2 \equiv \mathbf{M}^{-\frac{1}{2}} \mathbf{C} \mathbf{M}^{-\frac{1}{2}}. \quad (3.136)$$

Back in the time domain we then have

$$\mathbf{X}(t) = \dot{\mathbf{G}}(t) \mathbf{M} \mathbf{X}_0 + \mathbf{G}(t) \mathbf{P}_0 + (\mathbf{G} * \boldsymbol{\xi})(t). \quad (3.137)$$

$\mathbf{G}(t)$  satisfies the initial boundary conditions

$$\mathbf{G}(0) = \mathbf{0}, \quad (3.138)$$

$$\dot{\mathbf{G}}(0) = \mathbf{M}^{-1}, \quad (3.139)$$

and fully determines the initial-value propagator. We can express phase-space solutions

$$\mathbf{Z}(t) = \Phi(t) \mathbf{Z}_0 + (\Phi * \Xi)(t), \quad (3.140)$$

$$\Phi(t) = \begin{bmatrix} \dot{\mathbf{G}}(t) \mathbf{M} & \mathbf{G}(t) \\ \mathbf{M} \ddot{\mathbf{G}}(t) \mathbf{M} & \mathbf{M} \dot{\mathbf{G}}(t) \end{bmatrix}. \quad (3.141)$$

and so the initial-value phase-space propagator is given by  $\Phi_1(t, \tau) = \Phi(t - \tau)$ .

The thermal covariance defined in Sec. 3.4.2.2, which determines the remainder of the system correlations, can then be broken down into the block-matrix correlations

$$\sigma_T^{\mathbf{X}\mathbf{X}}(t_1, t_2) = (\mathbf{G} \cdot \boldsymbol{\nu} \cdot \mathbf{G})(t_1, t_2), \quad (3.142)$$

$$\sigma_T^{\mathbf{P}\mathbf{X}}(t_1, t_2) = \mathbf{M}(\dot{\mathbf{G}} \cdot \boldsymbol{\nu} \cdot \mathbf{G})(t_1, t_2), \quad (3.143)$$

$$\sigma_T^{\mathbf{X}\mathbf{P}}(t_1, t_2) = (\mathbf{G} \cdot \boldsymbol{\nu} \cdot \dot{\mathbf{G}})(t_1, t_2) \mathbf{M}, \quad (3.144)$$

$$\sigma_T^{\mathbf{P}\mathbf{P}}(t_1, t_2) = \mathbf{M}(\dot{\mathbf{G}} \cdot \boldsymbol{\nu} \cdot \dot{\mathbf{G}})(t_1, t_2) \mathbf{M}. \quad (3.145)$$

### 3.4.5.1 Rational Damping: Pseudo-Modes

If one has a rational damping kernel in the Laplace domain, or even meromorphic for now, then the propagator will also be rational (or meromorphic) in the Laplace domain. Therefore it can be decomposed into simple fractions which correspond to *pseudo-modes*:

$$\hat{\mathbf{G}}(s) = \mathbf{M}^{-\frac{1}{2}} \left[ \sum_k \frac{\frac{1}{f_k}}{s - f_k} \mathbf{U}_k \mathbf{U}_k^\dagger \right] \mathbf{M}^{-\frac{1}{2}}, \quad (3.146)$$

$$\mathbf{G}(t) = \mathbf{M}^{-\frac{1}{2}} \left[ \sum_k \frac{e^{f_k t}}{f_k} \mathbf{U}_k \mathbf{U}_k^\dagger \right] \mathbf{M}^{-\frac{1}{2}}, \quad (3.147)$$

assuming no repeated roots, and where the  $\mathbf{U}_k$  form an overcomplete basis given the initial condition  $\dot{\mathbf{G}}(0) = \mathbf{M}^{-1}$ , symmetry and the uniqueness of solutions. These are not true modes and the  $\mathbf{U}_k$  are not orthonormal because for nonlocal damping the pseudo-modes outnumber the system degrees of freedom. However, they will be associated with non-physical modes in a higher-dimensional linear system in the following section.

From the frequency representation of Eq. (3.134) and the Langevin equation, the pseudo-mode decomposition must satisfy

$$\left[ s^2 + 2s \hat{\lambda}(s) + \Omega^2 \right] \left[ \sum_k \frac{1}{s - f_k} \mathbf{U}_k \mathbf{U}_k^\dagger \right] = \mathbf{1}, \quad (3.148)$$

for all  $s$ . Taking the limit  $s \rightarrow f_k$  reveals a necessary condition for convergence, the characteristic equation:

$$\left[ f^2 + 2f \hat{\lambda}(f) + \Omega^2 \right] \mathbf{U} = \mathbf{0}, \quad (3.149)$$

which is a nonlinear eigen-value equation. Canonical-like perturbation theory applied to this equation will be in agreement with results from the second-master equation (Chapter 2), but this method is much more efficient in calculating these particular frequencies to higher order as this method does not require any integration.

### 3.4.5.2 Rational Damping: Extended Phase Space

We can transform the nonlocal problem into an effectively time-local and time-homogeneous one, only in a higher-dimensional extension of phase space. The spe-

cific problem that we will work out will be the regulated Ohmic coupling

$$\hat{\gamma}(s) = \frac{\gamma_0}{1 + \frac{s}{\Lambda}}, \quad (3.150)$$

where the Ohmic-limiting damping matrix  $\gamma_0$  and the cutoff matrix  $\Lambda$  commute and are both positive definite. This damping kernel is a rational function of order  $[0/1]$  in  $s$  and corresponds to

$$\tilde{\gamma}(\omega) = \frac{2\gamma_0}{1 + \left(\frac{\omega}{\Lambda}\right)^2}, \quad (3.151)$$

in the Fourier domain. For this to be a valid coupling, one only has to ensure that  $\tilde{\gamma}(\omega) > \mathbf{0}$  for all  $\omega$  and we have done so.

All homogeneous trajectories must satisfy the Langevin equation

$$[s^2\mathbf{M} + \mathbf{C} + 2s\hat{\gamma}(s)] \hat{\mathbf{X}}(s) = s\mathbf{M}\mathbf{X}_0 + \mathbf{P}_0, \quad (3.152)$$

which, by the initial value theorem, can be shown to yield what will be our *initial-value constraints*

$$\mathbf{X}(0) = \mathbf{X}_0, \quad (3.153)$$

$$\mathbf{M}\dot{\mathbf{X}}(0) = \mathbf{P}_0, \quad (3.154)$$

$$\mathbf{M}\ddot{\mathbf{X}}(0) = - \underbrace{[\mathbf{C} + 2\Lambda\gamma_0]}_{\mathbf{C}_{\text{bare}}} \mathbf{X}_0, \quad (3.155)$$

where the initial acceleration will always appear to have a bare or non-renormalized frequency. In general, if the damping kernel is of order  $[n/d]$  then we will need to determine  $d$  additional initial conditions beyond the first two.

Next we factor out the damping kernel's denominator and represent the ho-

mogeneous Langevin equation as a polynomial

$$[(1 + s \Lambda^{-1}) (s^2 \mathbf{M} + \mathbf{C}) + 2s \gamma_0] \hat{\mathbf{X}}(s) = (1 + s \Lambda^{-1}) (s \mathbf{M} \mathbf{X}_0 + \mathbf{P}_0), \quad (3.156)$$

which is equivalent to the third-order differential equation

$$\left\{ \Lambda^{-1} \mathbf{M} \frac{d^3}{dt^3} + \mathbf{M} \frac{d^2}{dt^2} + (2\gamma_0 + \Lambda^{-1} \mathbf{C}) \frac{d}{dt} + \mathbf{C} \right\} \mathbf{X}(t) = \mathbf{0}, \quad (3.157)$$

with constant coefficients. Given that we have made no approximations, solutions to this equation with proper initial conditions are immune to any issues of runaway solutions or causality violation often associated with higher-order differential equations. This is a key point that we will resurrect in discussion of the Abraham-Lorentz force. The environment induces nonlocal dynamics upon the system, and conversion into a higher-order ODE representation does not give license to choose arbitrary initial conditions for the open system.

Because we have a linear system of ordinary differential equations, we can solve this system with a matrix exponential analogous to the case of local damping.

Here it is natural to consider the extension of phase space into “forces” such that

$$\frac{d}{dt} \begin{bmatrix} \mathbf{X} \\ \mathbf{P} \\ \mathbf{F} \end{bmatrix} = \begin{bmatrix} \mathbf{0} & \mathbf{M}^{-1} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{1} \\ -\Lambda \mathbf{C} & (-2\Lambda \gamma_0 - \mathbf{C}) \mathbf{M}^{-1} & -\Lambda \end{bmatrix} \begin{bmatrix} \mathbf{X} \\ \mathbf{P} \\ \mathbf{F} \end{bmatrix}, \quad (3.158)$$

and let us denote this effective time-translation generator as  $\mathcal{F}$  so that we can

express our solutions

$$\begin{bmatrix} \mathbf{X}(t) \\ \mathbf{P}(t) \\ \mathbf{F}(t) \end{bmatrix} = e^{t\mathcal{F}} \begin{bmatrix} \mathbf{X}_0 \\ \mathbf{P}_0 \\ \mathbf{F}_0 \end{bmatrix}, \quad (3.159)$$

where the initial condition  $\mathbf{F}_0$  was previously determined from the initial-value theorem in Eq. (3.155). Given our initial value constraints we can then express our solutions

$$\begin{bmatrix} \mathbf{X}(t) \\ \mathbf{P}(t) \\ \mathbf{F}(t) \end{bmatrix} = e^{t\mathcal{F}} \begin{bmatrix} \mathbf{1} & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \mathbf{1} & \mathbf{0} \\ -2\mathbf{\Lambda}\gamma_0 - \mathbf{C} & \mathbf{0} & \mathbf{0} \end{bmatrix} \begin{bmatrix} \mathbf{X}_0 \\ \mathbf{P}_0 \\ \mathbf{0} \end{bmatrix}, \quad (3.160)$$

and let us denote this initial-value constraint matrix as  $\mathbf{T}$ ; it maps the two initial conditions  $(\mathbf{X}_0, \mathbf{P}_0, \mathbf{0}, \dots)$  of the nonlocal equation into the larger number of initial conditions required by the effective local equation  $(\mathbf{X}_0, \mathbf{P}_0, \mathbf{F}_0, \dots)$ . Given this representation we can now identify the  $2 \times 2$  leading principal minor of  $e^{t\mathcal{F}} \mathbf{T}$  with  $\Phi(t)$ , as this matrix would map  $(\mathbf{X}_0, \mathbf{P}_0)$  into  $(\mathbf{X}(t), \mathbf{P}(t))$ . In this particular case  $\mathbf{F}_0$  has no  $\mathbf{P}_0$  dependence and the second column of  $e^{t\mathcal{F}}$  directly yields  $(\mathbf{G}(t), \mathbf{M}\dot{\mathbf{G}}(t), \mathbf{M}\ddot{\mathbf{G}}(t))$ .

### 3.4.5.3 Full-Time Thermal Covariance

Here we simplify calculation of the full-time evolution of the thermal covariance, given by Eq. (3.100) and with the stationary initial-time propagator. First we express the Hermitian noise kernel in the Fourier domain as

$$\mathbf{N}(\tau, \tau') = \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \cos(\omega[\tau - \tau']) \tilde{\mathbf{N}}(\omega), \quad (3.161)$$

and then we apply the angle addition formula

$$\cos(\omega[\tau - \tau']) = \cos(\omega\tau) \cos(\omega\tau') + \sin(\omega\tau) \sin(\omega\tau'), \quad (3.162)$$

to obtain the full-time covariance formulas

$$\begin{aligned} \sigma_T(t_1, t_2) &= \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega [\Phi(t_1) * \cos(\omega t_1)] \tilde{\mathbf{N}}(\omega) [\Phi(t_2) * \cos(\omega t_2)]^T \\ &\quad + \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega [\Phi(t_1) * \sin(\omega t_1)] \tilde{\mathbf{N}}(\omega) [\Phi(t_2) * \sin(\omega t_2)]^T . \end{aligned} \quad (3.163)$$

The propagator convolutions are trivial and so, other than their inverse Laplace transform, there is only one nontrivial integral remaining, that over frequencies. It is also worth applying the block-matrix expressions of Sec. 3.4.5 to compare the position-position and momentum-momentum covariance expressions. The momentum covariance can be manipulated into

$$\begin{aligned} \sigma_T^{\text{PP}}(t_1, t_2) &= \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \omega^2 \mathbf{M} [\mathbf{G}(t_1) * \cos(\omega t_1)] \tilde{\boldsymbol{\nu}}(\omega) [\mathbf{G}(t_2) * \cos(\omega t_2)]^T \mathbf{M} \\ &\quad + \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \omega^2 \mathbf{M} [\mathbf{G}(t_1) * \sin(\omega t_1)] \tilde{\boldsymbol{\nu}}(\omega) [\mathbf{G}(t_2) * \sin(\omega t_2)]^T \mathbf{M} \\ &\quad + \mathbf{M} \mathbf{G}(t_1) \boldsymbol{\nu}(0) \mathbf{G}^T(t_2) \mathbf{M} . \end{aligned} \quad (3.164)$$

which, aside from the transient boundary-value term, takes the same form as the position covariance, except with an effectively higher-order coupling due to the additional factor of  $\omega^2$ . Therefore the momentum covariance will contain the dominant contribution to any potential ultraviolet sensitivity of the thermal covariance, whereas the position covariance will contain the dominant contribution to any possible infrared sensitivity.

In order to compute the evolution of the single-time thermal covariance, especially when calculating it numerically, it is often convenient to use the following alternative expressions

$$\dot{\sigma}_T(t) = \Phi(t) (\mathbf{N} * \Phi^T)(t) + (\mathbf{N} * \Phi)(t) \Phi^T(t) , \quad (3.165)$$

where the convolution of the propagator with the noise kernel should be performed before the frequency integral of the noise kernel. This will typically result in expressions more amenable to numerics since one can avoid increasingly oscillatory integrands.

For meromorphic damping kernels the frequency integral can be evaluated by contour integration (and the residue theorem) using the FDR (3.68) and rational expansion of the hyperbolic cotangent

$$\coth\left(\frac{\omega}{2T}\right) = \frac{2T}{\omega} + \frac{2}{\pi} \sum_{k=1}^{\infty} \frac{\frac{\omega}{2\pi T}}{k^2 + \left(\frac{\omega}{2\pi T}\right)^2}. \quad (3.166)$$

One should then be left with a sum of terms rational in the Laplace domain, which can be contracted into digamma or harmonic-number functions [respectively  $\psi(z)$  or  $H(z)$ ], which are asymptotically logarithmic. When transforming back to the time domain, the residues of the hyperbolic cotangent additionally give rise to products of rational functions of  $k$  with  $e^{-2\pi Ttk}$ . These terms contain all effects which decay at temperature-dependent rates and can be expressed in terms of Lerch transcendent functions,  $\Phi(z, 1, e^{-2\pi Tt})$ , which are useful for numerical calculations but not particularly insightful.

#### 3.4.5.4 Late-Time Thermal Covariance

Here we present the derivation of the general single-integral representation of the late-time thermal covariance. We start with the full-time, exact expression

$$\sigma_T(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \int_0^t d\tau_1 \int_0^t d\tau_2 \Phi(\tau_1) \tilde{\mathbf{N}}(\omega) e^{+i\omega(\tau_1 - \tau_2)} \Phi^T(\tau_2), \quad (3.167)$$

where we have made the simple change of variables  $\tau_i \rightarrow t - \tau_i$  for  $i = 1, 2$ . Introducing the additional change of variables  $\bar{\tau} = \tau_1 + \tau_2$ , the result can be rewritten as

$$\boldsymbol{\sigma}_T(t) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \int_0^t d\tau_2 \int_{\tau_2}^{\tau_2+t} d\bar{\tau} \boldsymbol{\Phi}(\bar{\tau}-\tau_2) \tilde{\mathbf{N}}(\omega) e^{+i\omega(\bar{\tau}-2\tau_2)} \boldsymbol{\Phi}^T(\tau_2). \quad (3.168)$$

The double time integration can then be split into two parts:

$$\int_0^t d\tau_2 \int_{\tau_2}^{\tau_2+t} d\bar{\tau} = \int_0^t d\tau_2 \int_{\tau_2}^t d\bar{\tau} + \int_0^t d\tau_2 \int_t^{\tau_2+t} d\bar{\tau}. \quad (3.169)$$

At sufficiently late times the contribution from the second integration domain can be neglected and we can approximate the whole integral as follows:

$$\int_0^t d\tau_2 \int_{\tau_2}^{\tau_2+t} d\bar{\tau} \approx \int_0^t d\tau_2 \int_{\tau_2}^t d\bar{\tau} = \int_0^t d\bar{\tau} \int_0^{\bar{\tau}} d\tau_2, \quad (3.170)$$

The next step is to manipulate the result into the form of a Laplace convolution:

$$\boldsymbol{\sigma}_T(t) \approx \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \int_0^t d\tau \left[ e^{-i\omega\tau} \boldsymbol{\Phi}(\tau) \tilde{\mathbf{N}}(\omega) \right] * \left[ e^{+i\omega\tau} \boldsymbol{\Phi}(\tau) \right]^T, \quad (3.171)$$

where we relabeled  $\bar{\tau}$  to  $\tau$ . Using the property of frequency shifting in the Laplace domain, i.e.  $\mathcal{L}\{e^{\lambda t} f(t)\} = \hat{f}(s-\lambda)$ , we obtain

$$\hat{\boldsymbol{\sigma}}_T(s) \approx \frac{1}{s} \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \boldsymbol{\Phi}(s+i\omega) \tilde{\mathbf{N}}(\omega) \boldsymbol{\Phi}^T(s-i\omega). \quad (3.172)$$

Incidentally, this full-time relation is exact for local damping. Application of the final-value theorem then immediately reveals the exact (even for nonlocal damping) late-time covariance

$$\boldsymbol{\sigma}_T(\infty) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \boldsymbol{\Phi}(+i\omega) \tilde{\mathbf{N}}(\omega) \boldsymbol{\Phi}^T(-i\omega). \quad (3.173)$$

Moreover, there is a simplification to this expression which is extremely useful for multiple system modes. The expression can be further simplified from a quadratic form into a linear form. First we express the noise kernel in terms of the damping kernel via the FDR (3.68). Next we relate the Fourier transform of the damping kernel to its Laplace transform by first noting

$$\tilde{\gamma}(\omega) = \int_{-\infty}^{+\infty} dt e^{-i\omega t} \gamma(t), \quad (3.174)$$

$$= \int_0^{\infty} dt e^{-i\omega t} \gamma(t) + \int_0^{\infty} dt e^{+i\omega t} \gamma(-t). \quad (3.175)$$

Then we use the fact that the damping kernel is doubly-Hermitian (see Sec. 2.5.2) to obtain

$$\tilde{\gamma}(\omega) = \int_0^{\infty} dt e^{-i\omega t} \gamma(t) + \int_0^{\infty} dt e^{+i\omega t} \gamma^T(t). \quad (3.176)$$

Assuming no singularities in  $\hat{\gamma}(s)$  for  $\text{Re}[s] > 0$ , which should be the case as the damping kernel is a somewhat localized distribution in time, then we may analytically continue the above relation to obtain

$$\tilde{\gamma}(\omega) = \lim_{s \rightarrow 0} [\hat{\gamma}(s+i\omega) + \hat{\gamma}^T(s-i\omega)]. \quad (3.177)$$

Now we expand  $\text{Re}[\hat{\gamma}(i\omega)]$  pairing  $\hat{\gamma}(+i\omega)$  with  $\hat{\Phi}(+i\omega)$  and  $\hat{\gamma}^T(-i\omega)$  with  $\hat{\Phi}^T(-i\omega)$ . One can then apply the Langevin equation evaluated at imaginary frequencies to rid all direct references to the damping kernel, save those intrinsic to the propagator.

Finally we are left with the expression

$$\sigma_T(\infty) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \frac{\tilde{\kappa}(\omega)}{\omega} \text{Im} \begin{bmatrix} \hat{\mathbf{G}}(-i\omega) & \mathbf{0} \\ \mathbf{0} & \omega^2 \mathbf{M} \hat{\mathbf{G}}(-i\omega) \mathbf{M} \end{bmatrix}, \quad (3.178)$$

which avoids all phase-space matrix multiplication.

### 3.4.6 Late-Time Stationary Dynamics

We now focus our attention on the dynamics generated by the stationary limit of the master equation, assuming that one exists. For an Ohmic spectrum with a large cutoff the homogeneous coefficients  $\mathcal{H}$  will reach their asymptotic value within the cutoff timescale, whereas the diffusion  $\mathbf{D}$  within the typical system timescales (although certain terms contributing to the diffusion coefficients will decay at a temperature-dependent rate whenever this is faster); see [64] for a detailed analysis of all these questions. In the weak-coupling regime this leaves the majority of the system evolution within this late-time regime wherein the master equation is effectively stationary. However, the existence of such a regime is not guaranteed in general. For instance, in the sub-Ohmic case the evolution can be persistently nonlocal and the effectively local late-time regime discussed here need not exist, as will be shown in Sec. 3.8. This non-Markovian phenomena was also noticed classically by Adelman [2].

#### 3.4.6.1 Late-Time Propagator

If the late-time stationary limit of the master equation exists, the late-time homogeneous generator  $\mathcal{H}(\infty)$  will take a form similar to the local limit (3.84), though with some modification of the system spring constant  $\mathbf{C}$ . The late-time homogeneous propagation is then given by

$$\lim_{t > \tau \gg 0} \Phi(t, \tau) = e^{-(t-\tau)\mathcal{H}(\infty)} \Phi(\tau), \quad (3.179)$$

which can be thought of as arising from the single-time propagator

$$\Phi_{\infty}(t) = e^{-t\mathcal{H}(\infty)}, \quad (3.180)$$

though it cannot be used to calculate response to forces, even in the late-time regime. The propagator  $\Phi_{\infty}(t)$  is also not equivalent to the late-time limit of the full-time propagator  $\Phi(t)$ , though they should share the same asymptotic dynamics. Specifically if one can take the asymptotic expansion

$$\Phi(t) = \phi_{\infty}(t) + \delta\phi(t), \quad (3.181)$$

where  $\phi_{\infty}(t)$  contains the asymptotic limiting behavior and  $\delta\phi(t)$  contains the early time corrections, which decay faster at late times, then  $\phi_{\infty}(t)$  should be generated by  $\mathcal{H}(\infty)$ , so that

$$\dot{\phi}_{\infty}(t) = -\mathcal{H}(\infty) \phi_{\infty}(t). \quad (3.182)$$

This can be rigorously justified if  $\hat{\gamma}(s)$  and, thus,  $\hat{\Phi}(s)$  are rational, which implies that the time dependence of  $\Phi(t)$  corresponds to damped oscillations with various timescales, all of which can be calculated from the characteristic equation (3.149). A specific example of this will be given in Sec. 3.7.1 for regulated Ohmic coupling. On the other hand, the sub-Ohmic coupling that will be studied in Sec. 3.8 provides a pertinent counter-example [in that case  $\Phi(t)$  decays as a negative power-law rather than exponentially] which shows that this situation does not necessarily exist when the damping kernel is not meromorphic.

### 3.4.6.2 Late-Time Diffusion and Covariance

Given late-time master equation coefficients which have all taken their asymptotic values, one can show that the evolution of the covariance in that regime is simply given by

$$\boldsymbol{\sigma}(t) = \boldsymbol{\sigma}_T^\infty + \boldsymbol{\Phi}_\infty(t-\tau) [\boldsymbol{\sigma}(\tau) - \boldsymbol{\sigma}_T^\infty] \boldsymbol{\Phi}_\infty^\text{T}(t-\tau), \quad (3.183)$$

which is a solution of Eq. (3.120) as long as one assumes  $\boldsymbol{\mathcal{H}}(t)$  and  $\mathbf{D}(t)$  to be time-independent after some time  $\tau$  in the late-time regime. Note that we have assumed that the master equation coefficients reached their asymptotic values much faster than the relaxation time (as illustrated in [64] with the example of the Ohmic coupling, this may be the case for high temperature, but not necessarily so for zero temperature).

The asymptotic value of the late-time thermal covariance  $\boldsymbol{\sigma}_T^\infty$  has been reduced to a single integral in 3.4.5.4. From this single integral formulation, it is actually easier to obtain first  $\boldsymbol{\sigma}_T^\infty$ , and then obtain the late-time diffusion coefficients using the Lyapunov equation (3.123):

$$\boldsymbol{\mathcal{H}}_\infty \boldsymbol{\sigma}_T^\infty + \boldsymbol{\sigma}_T^\infty \boldsymbol{\mathcal{H}}_\infty^\text{T} = 2 \mathbf{D}_\infty. \quad (3.184)$$

Additional insight can be gathered from its block-matrix decomposition (3.121)-(3.122). The **XX**-block of the Lyapunov equation yields

$$\mathbf{0} = \boldsymbol{\sigma}_{\text{XP}} \mathbf{M}^{-1} + \mathbf{M}^{-1} \boldsymbol{\sigma}_{\text{PX}}, \quad (3.185)$$

which constrains the late-time  $\boldsymbol{\sigma}_{\text{XP}}$  correlations to vanish. The **PP**-block of the

Lyapunov equation then yields

$$\mathbf{D}_{\mathbf{PP}} = \gamma_L \mathbf{M}^{-1} \boldsymbol{\sigma}_{\mathbf{PP}} + \boldsymbol{\sigma}_{\mathbf{PP}} \mathbf{M}^{-1} \gamma_L, \quad (3.186)$$

which dictates that the momentum covariance, along with its high-frequency sensitivities (Sec. 3.4.5.3), is entirely determined from the regular diffusion, which must therefore also contain these high-frequency sensitivities. Finally the  $\mathbf{XP}$ -block of the Lyapunov equation yields

$$\mathbf{D}_{\mathbf{XP}} = \boldsymbol{\sigma}_{\mathbf{XX}} \mathbf{C}_L - \mathbf{M}^{-1} \boldsymbol{\sigma}_{\mathbf{PP}}. \quad (3.187)$$

Only the momentum covariance contains the highest frequency sensitivities, therefore it must follow that here the anomalous diffusion coefficients act as “anti-diffusion” coefficients in keeping the position covariance free of such sensitivities. On the other hand, only the position covariance can contain the lowest frequency sensitivities and these must, therefore, be entirely contained in the anomalous diffusion coefficient if they exist.

## 3.5 Analysis of Solutions

### 3.5.1 Decoherence of a Quantum Superposition

In this section we will illustrate how one can get a useful qualitative picture of the phenomenon of environment-induced decoherence from the our solutions. In order to do that we will consider a quantum superposition,  $|\psi\rangle \propto |\psi_+\rangle + |\psi_-\rangle$ , of a pair of states  $|\psi_\pm\rangle$  which correspond to a pair of Gaussian wavefunctions in position space separated by a distance  $2\delta_X$ ) and with the appropriate normalization constant.

Specifically, we have

$$\psi_{\pm}(X) = \psi_0(X \mp \delta_X), \quad (3.188)$$

$$\psi_0(X) = \sqrt{N(0, \sigma_0^{XX}; X)}. \quad (3.189)$$

where  $N(\mu, \sigma^2; X)$  is a normalized Gaussian distribution for the variable  $x$  with mean  $\mu$  and variance  $\sigma^2$ , and  $\psi_0(X)$  is a reference Gaussian state centered at the origin. We calculate the resultant Wigner function to be

$$W(\mathbf{Z}) \propto W_+(\mathbf{Z}) + W_-(\mathbf{Z}) + 2 \cos(2\delta_X P) W_0(\mathbf{Z}), \quad (3.190)$$

where  $W_+$ ,  $W_-$  and  $W_0$  are respectively the Wigner functions of the states  $|\psi_+\rangle$ ,  $|\psi_-\rangle$  and  $|\psi_0\rangle$ . This Wigner function, plotted in Fig. 3.1, exhibits oscillations of size  $1/\delta_X$  along the  $P$  direction. These oscillations are closely connected to the coherence of the quantum superposition (and the existence of non-diagonal terms in the density matrix) and are absent in the Wigner function for the incoherent mixture  $W(\mathbf{Z}) = (1/2)[W_+(\mathbf{Z}) + W_-(\mathbf{Z})]$ .

In this context the decoherence effect due to the interaction with the environment corresponds to the washing-out of the oscillations in the reduced Wigner function as it evolves according to the master equation. This can be seen rather simply from the QBM solutions given by Eq. (3.113). Taking into account that the inverse Fourier transform corresponds to a convolution of the homogeneously evolving initial state and a Gaussian function with the thermal covariance  $\boldsymbol{\sigma}_T(t)$  as its covariance matrix, the Wigner function can then be expressed as

$$W[\mathbf{Z}, t] = \int d\mathbf{Z}' \frac{N[\mathbf{0}, \boldsymbol{\sigma}_T(t); \mathbf{Z} - \mathbf{Z}']}{\det[\boldsymbol{\Phi}(t)]} W[\boldsymbol{\Phi}^{-1}(t) \mathbf{Z}', 0], \quad (3.191)$$

where the thermal Gaussian acts as a Gaussian smearing function which starts as a delta function at the initial time and broadens with the passage of time until it eventually reaches its asymptotic thermal-equilibrium value. Therefore, several aspects will be at play. On the one hand, the initial state evolves as a phase-space distribution with trajectories corresponding to the homogeneous solutions of the Langevin equation. On the other hand, by diagonalizing  $\sigma_T(t)$  at each instant of time one gets the principal directions (phase-space axes) and the widths (uncertainties) of the Gaussian smearing function, which will average out any details of those sizes along the corresponding directions. When  $\sigma_T(t)$  along the direction of the interference oscillations of the Wigner function becomes comparable to their wavelength, they get washed out and the Wigner function becomes equivalent to that of the completely incoherent mixture. The time it takes for this to happen is known as the decoherence time  $t_{\text{dec}}$ .

Knowledge of the qualitative behavior of  $\sigma_T(t)$ , combined with the fact that the phase-space distribution  $\det[\Phi(t)]^{-1} W[\Phi^{-1}(t) \mathbf{Z}', 0]$  is rotating with the characteristic oscillation frequency and shrinking with the characteristic relaxation time is all that one needs to understand how different initial states decohere as time goes by. In particular, if the decoherence time-scale, given by  $t_{\text{dec}}$ , is much shorter than the characteristic oscillation period and the relaxation time (but sufficiently longer than  $1/\Lambda$ ), one can approximate the phase-space distribution by the initial reduced Wigner function (after any possible initial kick). For instance, for an Ohmic environment in the high-temperature regime one can, under those circumstances, approximately take  $\sigma_T^{PP}(t) \sim D_{PP}^\infty t$  with  $D_{PP}^\infty \sim 2\gamma_0 T$  and from the condi-

tion  $\sqrt{\sigma_{PP}(t)} \sim 1/\delta_X$  obtain an estimated decoherence time  $t_{\text{dec}} \sim 1/(2\gamma_0 T \delta_X^2)$ , in agreement with the standard result for this situation [163, 121]. On the other hand, if  $\gamma_0$  or  $\delta_X$  are very small  $t_{\text{dec}}$  can become comparable or larger than the dynamical timescales  $1/\Omega$  or  $1/\gamma$ , and the previous estimate can no longer be applied because one needs to take into account the evolution of  $\sigma_T(t)$ , which is then less simple (it will roughly oscillate with frequency  $\Omega$  around a central value which increases with a characteristic timescale  $1/\gamma$  until it approaches the asymptotic thermal value), as well as the rotation and shrinking of the initial Wigner function under the homogeneous evolution. Note also that if we had considered an initial superposition of Gaussian states peaked at the same location but with different momenta, which corresponds to a Wigner function along the position rather than momentum direction, the decoherence time would typically be much longer, since  $\sigma_T^{XX}(t)$  vanishes at the initial time and grows with a characteristic timescale of order  $1/\Omega$ . In that case, the rotation of the Wigner function becomes important since the oscillations can then be averaged out due to the larger values of  $\sigma_T^{PP}(t)$ .

The zero-temperature regime for an Ohmic environment is also qualitatively different. There is a substantial contribution to  $\sigma_T^{PP}(t)$  from a jolt of the diffusion coefficient  $D_{PP}$  for times of order  $1/\Lambda$ . However, this is actually regarded as an unphysical consequence of having considered a completely uncorrelated initial state for the system plus environment, and this kind of highly cutoff sensitive features at early times of order  $1/\Lambda$  should disappear if one considers a finite (cutoff independent) preparation time for the initial state of the system coupled to the environment (see Sec. 4.3). For sufficiently weak coupling  $\delta_X$  or  $t_{\text{dec}}$  can become comparable or larger

than the relaxation time more easily than at high temperatures since the components  $\sigma_T(t)$  are much smaller in this case. For example, the asymptotic thermal value of  $\sigma_{PP}$  is of order  $M\Omega$  (for weak coupling), much smaller than the high-temperature results, which is of order  $MT$ . In such situations, the main effect of considering a sufficiently long time is through the shrinking of  $\det[\Phi(t)]^{-1} W[\Phi^{-1}(t) \mathbf{Z}', 0]$  and the size of its oscillations.

We have focused in this subsection on describing the qualitative features of the environment-induced decoherence of an initial coherent superposition that can be easily inferred from our general result for the evolution of the reduced Wigner function. A much more quantitative study is possible by using the exact analytical results for the diffusion coefficients and, especially,  $\sigma_T(t)$ , which will be presented in Sec. 3.6. We expect agreement with the numerical results obtained in Ref. [121] for local damping, whereas significant deviations may appear when the nonlocal effects of dissipation are important (such as in the sub-Ohmic case) since previously obtained master equations are not valid in those regimes.

### 3.5.2 Initial-Time Pathologies

In this section we will thoroughly explore the initial-time pathologies which are due to our factorized initial state of the system and environment. In QBM these pathologies can be cleanly delineated in both cause and effect into two categories: the *slip*, which is associated with the damping kernel and produce a homogeneous *kick* to the initial state of the system, and the *jolts*, which are associated with the noise

kernel and produce rapid diffusion at the initial time. More generally, for nonlinear systems, all such pathologies will be labeled jolts. Preparation of correlated initial states will be considered in Sec. 4.3.

### 3.5.2.1 Initial Slip and Kick

Let us consider, most simply, the case of constant system coefficients and a stationary damping kernel which we will eventually take to be local. Full solutions were given in Sec. 3.4.5.

$$\mathbf{M}\ddot{\mathbf{X}}(t) + 2(\boldsymbol{\gamma} * \dot{\mathbf{X}})(t) + \mathbf{C}\mathbf{X}(t) + 2\boldsymbol{\gamma}(t)\mathbf{X}_0 = \boldsymbol{\xi}(t). \quad (3.192)$$

Note that the slip term can be moved to the right-hand side and reidentified as a transient driving force.

$$\mathbf{F}_\gamma(t) = -2\boldsymbol{\gamma}(t)\mathbf{X}_0, \quad (3.193)$$

whose contribution to the solution is simply given by adding a term  $\mathbf{G}(t) * \mathbf{F}_\gamma(t)$ , which reduces to  $\mathbf{G}(t)[-2\boldsymbol{\gamma}_0\mathbf{X}_0]$  in the limit of local damping. The full solution (including slip) is given by

$$\mathbf{X}(t) = \dot{\mathbf{G}}(t)\mathbf{M}\mathbf{X}_0 + \mathbf{G}(t)\mathbf{P}_0 + (\mathbf{G} * \boldsymbol{\xi})(t). \quad (3.194)$$

Whereas without the local slip, the solution to the ODE would be

$$\mathbf{X}(t) = \dot{\mathbf{G}}(t)\mathbf{M}\mathbf{X}_0 + \mathbf{G}(t)[\mathbf{P}_0 + 2\boldsymbol{\gamma}_0\mathbf{X}_0] + (\mathbf{G} * \boldsymbol{\xi})(t). \quad (3.195)$$

Therefore it can be seen that the slip produces the rapid kick to the homogeneous solutions of the form

$$\mathbf{P}_0 \rightarrow \mathbf{P}_0 - 2\boldsymbol{\gamma}_0\mathbf{X}_0, \quad (3.196)$$

which occur within the U.V. cutoff timescale.

Moreover, the effect of any such kick can easily be accounted for by simply distinguishing between the “bare” initial state before the kick and the “renormalized” state immediately after the kick. Following our solutions, one can easily see that this initial kick translates into a distortion of the Wigner distribution from the bare initial state to a shifted one

$$W_{\text{bare}}(\mathbf{X}, \mathbf{P}) \rightarrow W_{\text{ren}}(\mathbf{X}, \mathbf{P}) = W_{\text{bare}}(\mathbf{X}, \mathbf{P} - 2\gamma_0 \mathbf{X}). \quad (3.197)$$

In the density-matrix formalism, it is a straightforward calculation to show that this transformation is unitary.

$$\rho_{\text{bare}}(\mathbf{X}, \mathbf{Y}) \rightarrow \rho_{\text{ren}}(\mathbf{X}, \mathbf{Y}) = e^{+i\mathbf{X}^T \gamma_0 \mathbf{X}} \rho_{\text{bare}}(\mathbf{X}, \mathbf{Y}) e^{-i\mathbf{Y}^T \gamma_0 \mathbf{Y}}. \quad (3.198)$$

As such, pure states are mapped to pure states. Moreover, the Hamiltonian generator for this unitary transformation  $-\mathbf{X}^T \gamma_0 \mathbf{X}$  is dynamically linear, and therefore there will be a number of *kinematic moment invariants* [53]. In particular, we will single out the uncertainty function. Note that the phase-space transformation in Eq. (3.197) has a Jacobian matrix  $\mathbf{T}$  with determinant equal to one:

$$\mathbf{T} = \begin{bmatrix} \mathbf{1} & \mathbf{0} \\ -2\gamma_0 & \mathbf{1} \end{bmatrix}, \quad \det \mathbf{T} = 1. \quad (3.199)$$

Therefore, it is simple to calculate renormalized expectation values in terms of bare expectation values and vice versa:

$$\langle A(\mathbf{X}, \mathbf{P}) \rangle_{\substack{\text{ren} \\ \text{or} \\ \text{bare}}} = \iint d\mathbf{X} d\mathbf{P} A(\mathbf{X}, \mathbf{P}) W_{\substack{\text{ren} \\ \text{or} \\ \text{bare}}}(\mathbf{X}, \mathbf{P}), \quad (3.200)$$

$$\langle A(\mathbf{X}, \mathbf{P}) \rangle_{\text{ren}} = \langle A(\mathbf{X}, \mathbf{P} - 2\gamma_0 \mathbf{X}) \rangle_{\text{bare}}. \quad (3.201)$$

We can easily see that the normalization, linear entropy and state overlap are all unchanged by the kick. We can also check explicitly that the Heisenberg uncertainty relation is preserved as follows. First, we start with the covariance matrix  $\boldsymbol{\sigma}$  for  $\mathbf{X}$  and  $\mathbf{P}$  corresponding to the Wigner distribution, which transforms in the following way under linear phase-space transformations:

$$\boldsymbol{\sigma} \rightarrow \mathbf{T} \boldsymbol{\sigma} \mathbf{T}^T. \quad (3.202)$$

Hence, from Eq. (3.199) we have

$$\det \boldsymbol{\sigma}_{\text{bare}} = \det \boldsymbol{\sigma}_{\text{ren}}. \quad (3.203)$$

Finally, one takes into account that

$$(\det \boldsymbol{\sigma}) \geq \left(\frac{\hbar}{2}\right)^{2N}, \quad (3.204)$$

corresponds to the formulation in terms of the  $2N$ -dimensional Wigner representation of the generalized Heisenberg uncertainty relation due to Schrödinger [132, 43]:

$$\langle \Delta \hat{X}^2 \rangle \langle \Delta \hat{P}^2 \rangle - \left\langle \frac{1}{2} \{ \Delta \hat{X}, \Delta \hat{P} \} \right\rangle^2 \geq \frac{\hbar^2}{4}, \quad (3.205)$$

for the position and momentum operators, where  $\{\hat{A}, \hat{B}\} \equiv \hat{A}\hat{B} + \hat{B}\hat{A}$  is the anti-commutator.

In summary, the new Wigner function that results from the transformation defined by Eq. (3.197) always corresponds to a physical density matrix since the transformation preserves the normalization and the real-valuedness of the Wigner function (implying the normalization and hermiticity of the density matrix) as well as the positivity of the associated density matrix. Therefore, if one is interested in

analyzing the evolution of a certain state of the system better correlated with the environment, one can simply take such a state as  $W_{\text{ren}}(\mathbf{X}, \mathbf{P})$  and study its evolution for  $t \gg \Lambda^{-1}$  by considering the Langevin equation without the term that gives rise to the initial kick. However, given any  $W_{\text{ren}}(\mathbf{X}, \mathbf{P})$  it is always possible to follow in detail the evolution during the initial time by inverting the kick transformation to obtain the corresponding initial Wigner function before the interaction was switched on and using the full Langevin equation. In general this approach can be regarded simply as a formal procedure to generate a better correlated initial state, but the explicit construction involving unitary evolution for the whole system at all times guarantees that the result is well defined (i.e. the exact solutions of the master equation obtained in this way preserve the positivity of the density matrix).

### 3.5.2.2 Initial Jolts

Early studies by Unruh and Zurek [147] as well as HPZ [84] already revealed that at low temperatures the normal diffusion coefficient  $D_{PP}(t)$  of an Ohmic environment exhibited a strong cutoff ( $\Lambda$ ) sensitivity for very early times of order  $1/\gamma_0$ . As shown in the next section and [64], in the large cutoff limit where the use of the local propagator is a good approximation one can obtain relatively simple analytic results. They confirm that for zero temperature the normal diffusion coefficient, which vanishes at the initial time, exhibits an initial jolt with an amplitude of order  $\Lambda$  peaked around a time of order  $1/\Lambda$  and then decays roughly like  $1/t$  (for times much earlier than  $1/\Omega$  and  $1/\gamma_0$ ).

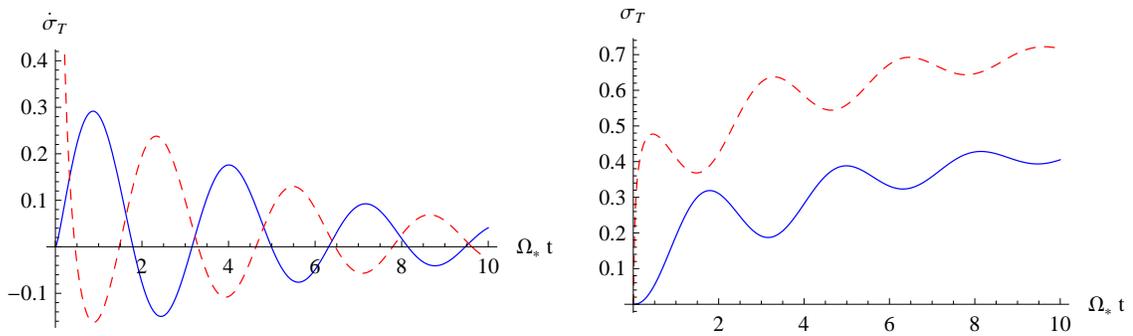


Figure 3.2: Exact thermal covariance dynamics for  $\cdot$  **normalized position uncertainty**  $M\Omega_*\sigma_{XX}^T(t)$  and  $\cdots$  **normalized momentum uncertainty**  $\frac{\sigma_{PP}^T(t)}{M\Omega_*}$  in the highly non-Markovian regime with  $T = \gamma_* = \frac{\Omega_*}{10}$ ,  $\Lambda_* = 100\Omega_*$ .

Alternatively, one can obtain the exact analytic results for finite cutoff by computing  $\dot{\sigma}_T(t)$  using Eq. (3.165), as explained in Sec. 3.4.5.3. The resulting expressions are rather lengthy and not particularly insightful, and will not be reported here, but they have been employed to plot some examples of exact results for the diagonal components of  $\dot{\sigma}_T(t)$  and  $\sigma_T(t)$  in Figs. 3.2 and 3.3. From the different components of the thermal covariance and its time derivative one can obtain the diffusion coefficients using Eq. (3.120), and in particular one can see from Fig. 3.3 the presence of the jolt mentioned above.

It is important to emphasize that such kind of behavior, as well as an associated rapid growth of  $\sigma_{PP}(t)$  and a slower growth of  $\sigma_{XX}(t)$  (which eventually decays exponentially within the relaxation time-scale  $1/\gamma_0$ ) until they both reach values which depend logarithmically on  $\Lambda$  for large values of  $\Lambda$ , is a consequence of having started with a completely uncorrelated initial stated.

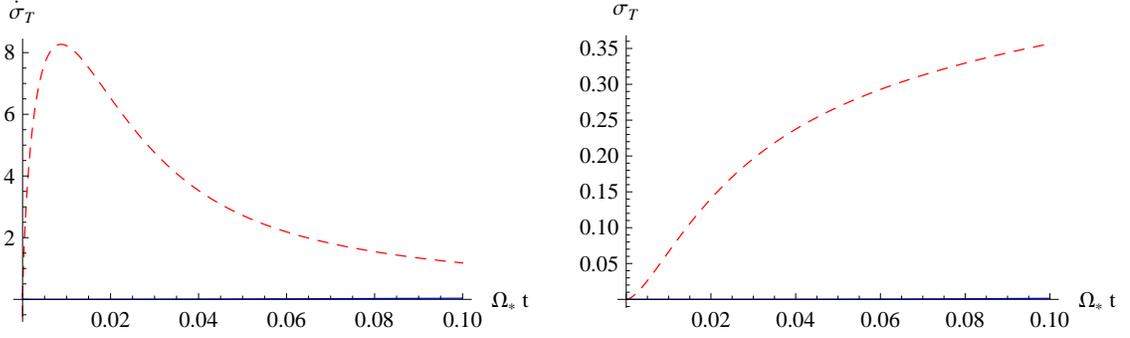


Figure 3.3: Same plot as in the previous figure, but with a much larger time resolution, which reveals the presence of the initial jolt in  $\dot{\sigma}_{PP}(t)$  peaked around  $t \sim 1/\Lambda_*$ , while  $\sigma_{XX}(t)$  and  $\dot{\sigma}_{XX}(t)$  remain essentially zero at those timescales.

Finally, we would mention that this dynamical discrepancy between the position and momentum covariance is a generic feature of QBM with position-position coupling to the environment. From Sec. 3.4.5.3 it was shown that the momentum covariance contains the high-frequency sensitivities in the full-time regime, regardless of the spectral-density function or damping kernel. This high-frequency sensitivity is the ultimate cause of this behavior.

### 3.6 Analysis of Spectral Densities

In this section we analyze exact solutions of QBM given a single oscillator and a specific archetypal spectral density: sub-Ohmic, Ohmic, and supra-Ohmic. Ohmic coupling (without regulation) is defined

$$\tilde{\gamma}(\omega) = 2\gamma_0, \quad (3.206)$$

as it results in the simple damping term  $\gamma_0$  analogous to an Ohmic resistor in an LRC circuit. In this sense our environment is analogous to the microscopic degrees of freedom in the resistor which impede current and dissipate energy. In the circuit analogy, one can also relate local damping to an infinite transmission line (parallel or coaxial) with constant impedance, which may continually and forever store energy due to its infinite length.

Local damping, or equivalently Ohmic coupling, is the most ubiquitous damping considered in the literature and perhaps also found in nature. As we will show in Chapter 6, local damping results from the linear coupling of a non-relativistic point particle to a scalar or electromagnetic field. Quantum mechanically, Ohmic coupling requires a finite cutoff-frequency regulator  $\chi(\omega/\Lambda) : [0, \infty) \rightarrow [1, 0)$  to obtain finite diffusion. Classical fluctuations are less frequency sensitive despite the fact that they are larger (see Sec. 2.5.2.3) and thus they do not necessarily require as much U.V. regulation. Depending on the context, this cutoff may be physically related to the non-vanishing size of the particle or the necessity of a field-theoretic description of the particle.

It has been standard in the QBM literature to consider the polynomial class of damping kernels (or equivalently spectral densities)

$$\tilde{\gamma}(\omega) \propto \omega^n, \tag{3.207}$$

also with additional regularization, where the  $n < 0$  case is sub-Ohmic and the  $n > 0$  case is supra-Ohmic. For  $n \leq -1$  one requires I.R. cutoff regulation whereas for  $n \geq 0$  one requires U.V. cutoff regulation.

Finally we would mention that it is conventional in 1-oscillator QBM to incorporate the system mass into the definition of the damping kernel so that  $\gamma_0$  becomes the local damping rate. For a multipartite system of differing masses, or for other quantum systems without any notion of mass, this not done for practical reasons.

## 3.7 Ohmic Coupling with Finite Cutoff

### 3.7.1 The Nonlocal Propagator

The arguably simplest example of Ohmic dissipation with finite cutoff that one can construct corresponds to the following damping kernel:

$$\hat{\gamma}(s) = \frac{\gamma_0}{1 + \frac{s}{\Lambda}}. \quad (3.208)$$

This damping kernel is constant at frequencies much smaller than the cutoff, but vanishes in the high frequency limit. In the Fourier domain we have

$$\tilde{\gamma}(\omega) = \frac{2\gamma_0}{1 + \left(\frac{\omega}{\Lambda}\right)^2}. \quad (3.209)$$

Note that in the limit of small  $\Lambda$  the damping kernel corresponds to an  $n = -2$  sub-Ohmic coupling and  $\Lambda$  becomes an I.R. cutoff. This will become relevant in understanding the limit of large coupling  $\gamma_0$ .

Calculating the Green function amounts to factoring a cubic polynomial. Specifically, one needs to factor  $(s^2 + \Omega^2)(s + \Lambda) + 2\gamma_0\Lambda s$  in the denominator of the Green function  $\hat{G}(s)$ . For the underdamped system the effect of a large finite cutoff is to

shift the system relaxation and oscillation timescales slightly:

$$\gamma_\star = \gamma_0 \left[ 1 + 2\frac{\gamma_0}{\Lambda} + \mathcal{O}\left(\frac{1}{\Lambda^2}\right) \right], \quad (3.210)$$

$$\Omega_\star^2 = \frac{\Lambda}{\Lambda - 2\gamma_\star} \Omega^2. \quad (3.211)$$

and to add an additional relaxation timescale comparable to the cutoff:

$$\Lambda_\star = \Lambda - 2\gamma_\star. \quad (3.212)$$

If we parametrize everything in terms of these phenomenological frequencies, the Green function for the fully nonlocal damping kernel can always be expressed as

$$\hat{G}(s) = \frac{1}{M} \frac{s + \Lambda}{(s + \Lambda_\star)(s^2 + 2\gamma_\star s + \Omega_\star^2)}, \quad (3.213)$$

without the need to explicitly factor a cubic polynomial, while the original parameters are given by

$$\gamma_0 = \frac{\Lambda_\star^2 + 2\gamma_\star \Lambda_\star + \Omega_\star^2}{(\Lambda_\star + 2\gamma_\star)^2} \gamma_\star, \quad (3.214)$$

$$\Omega^2 = \frac{\Lambda_\star}{\Lambda_\star + 2\gamma_\star} \Omega_\star^2, \quad (3.215)$$

$$\Lambda = \Lambda_\star + 2\gamma_\star. \quad (3.216)$$

then we never have to actually factor the cubic polynomial.

After using partial fraction decomposition in Eq. (3.213), one can easily transform back to the time domain and obtain the exact propagator for the nonlocal case:

$$G(t) = \frac{\Lambda_\star^2 + \Omega_\star^2}{(\Lambda_\star - \gamma_\star)^2 + \tilde{\Omega}_\star^2} \left[ G_\infty(t) - \frac{2\gamma_\star}{\Lambda_\star^2 + \Omega_\star^2} \left( \dot{G}_\infty(t) - \frac{e^{-\Lambda_\star t}}{M} \right) \right], \quad (3.217)$$

where  $G_\infty(t)$  is the late-time local propagator introduced in Sec. 3.4.6.1. As long as  $\Lambda_\star > \gamma_\star$  the term proportional to  $e^{-\Lambda_\star t}$  can be neglected at sufficiently late times, when the terms involving  $G_\infty(t)$  dominate. The term proportional to  $\dot{G}_\infty(t)$  simply causes a phase shift and the late-time master equation coefficients are, therefore,

$$\gamma_L(\infty) = \gamma_\star \quad , \quad \Omega_L(\infty) = \Omega_\star. \quad (3.218)$$

In the high cutoff limit one recovers the usual coefficients  $\gamma_0$  and  $\Omega$ . Furthermore, in that limit one can approximate  $G(t)$  by  $G_\infty(t)$  since the extra terms are suppressed by inverse powers of  $\Lambda^2$ . For  $G(t)$  this is true even at arbitrarily early times of order  $\Lambda^{-1}$ : although the exponential factor is not suppressed, the prefactor  $1/\Lambda_\star^2$  is sufficient to suppress its contribution to  $G(t)$ . This is not true, however, for  $\ddot{G}(t)$  (or higher-order derivatives), which also appears in  $\Phi(t)$ .  $\ddot{G}(t)$  does not contribute to the thermal covariance, but whether it contributes to its time derivative  $\dot{\sigma}_T(t)$  as well as to the diffusion coefficients, which are related to  $\dot{\sigma}_T(t)$  through Eq. (3.120), is a bit more subtle. More straightforward expressions, such as the definition of the thermal covariance, reference  $\ddot{G}(t)$  under integration, however the effects of time integration can offset this. It is more useful to turn to Eq. (3.164), which manifestly shows that there is no  $\ddot{G}(t)$  dependence in  $\dot{\sigma}_T(t)$ . Thus, the final conclusion is that we can use the approximate local propagator  $G_\infty(t)$  to calculate the diffusion coefficients at arbitrary times in the large cutoff limit. Comparison of the results evaluated using the exact expressions and plotted in Sec. 3.5.2.2 and the approximate results for the large cutoff limit also support this conclusion.

We close this subsection with a brief discussion of the possible dissipative

regimes when considering finite values of the cutoff in our damping kernel, since the presence of this new scale can give rise to a richer set of possibilities. For our rational cutoff function we have three different dissipative regimes corresponding to the three shaded regions in Fig. 3.4. The boundary between different regions corresponds to the values of the parameters for which a pair of roots of the denominator of  $\hat{G}(s)$  degenerate and change character, i.e. they change from a complex conjugate pair to two real ones. Atop the diagram where  $\Lambda \gg \Omega$ , lies the regime of local damping, whereas along the bottom of the diagram where  $\Lambda \ll \Omega$ , lies an effectively sub-Ohmic regime as  $\Lambda$  becomes an I.R. cutoff.

The white shaded vertical stripe to the left lies completely in the weak coupling regime and constitutes the *underdamped* regime. This regime is as described previously with slowly decaying oscillations and a cutoff-dependent decay rate. The grey shaded middle region denotes the *overdamped* regime. This regime is also analogous to that of the simple and overdamped harmonic oscillator but with an additional cutoff-dependent decay rate. The black shaded region to the right denotes a new nonlocal *strong-coupling* regime that emerges for a sufficiently strong coupling (such that  $\gamma_0$  is large compared to the cutoff). Specifically, as derived from Eqs. (3.214)-(3.216), the relevant scales for this regime in the limit of very strong

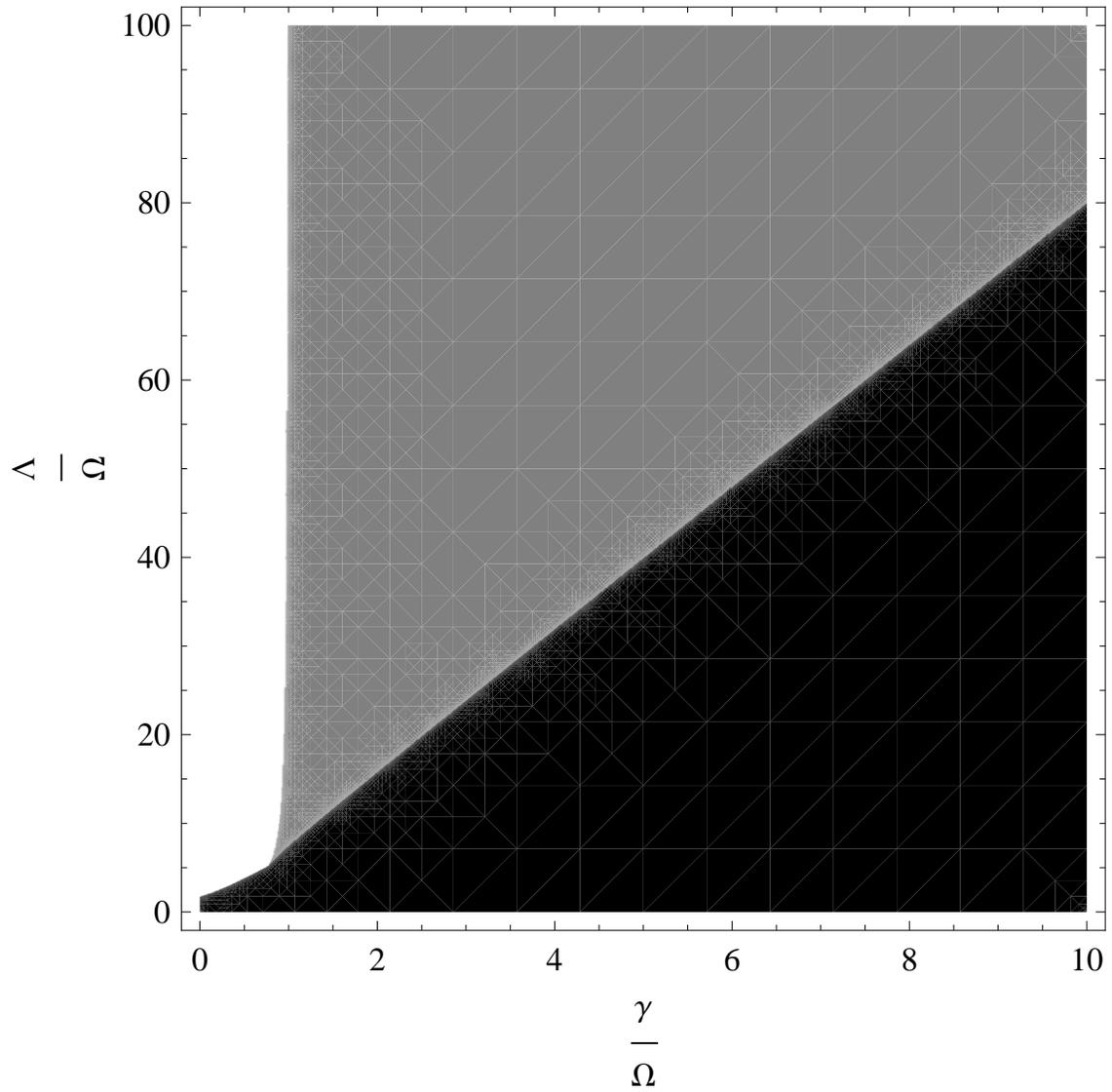


Figure 3.4: Dissipative phases for Ohmic damping with finite rational cutoff. From left to right they are *underdamped* in white, *overdamped* in grey, and *strong coupling* in black.

coupling are

$$\Lambda_\star = \frac{\Omega^2}{2\gamma_0} - \frac{\Omega^4}{4\Lambda\gamma_0^2} + \mathcal{O}\left(\frac{1}{\gamma_0^3}\right), \quad (3.219)$$

$$\gamma_\star = \frac{\Lambda}{2} - \frac{\Omega^2}{4\gamma_0} + \mathcal{O}\left(\frac{1}{\gamma_0^2}\right), \quad (3.220)$$

$$\Omega_\star = 2\Lambda\gamma_0 + \Omega^2 + \mathcal{O}\left(\frac{1}{\gamma_0}\right). \quad (3.221)$$

Hence, we can see that one has moderately damped, rapid oscillations plus an additional slow decay rate.

### 3.7.2 Late-Time Covariance for Finite Cutoff

In Sec. 3.7.1 the late-time dissipation and renormalized frequency coefficients were directly inferred from the nonlocal propagator to be  $\gamma_\star$  and  $\Omega_\star$ , the result of factoring a cubic polynomial in the nonlocal Green function. These coefficients are entirely non-perturbative in both coupling and cutoff and completely determine the late-time propagator. The remaining part of the solution pertains to the emergence of the thermal covariance, whose late-time dynamics can be described as in Sec. 3.4.6.2, given the late-time propagator and the late-time thermal covariance. The late-time thermal covariance can also be related to the late-time diffusion coefficients through the Lyapunov equation, Eq. (3.123), but the thermal covariance is an easier quantity to compute. If interested in the diffusion coefficients, one can then obtain them straightforwardly using Eq. (3.123).

For our damping kernel the simplified integrals derived in 3.4.5.4 are contour integrals and can be evaluated via the residue theorem after using the rational expansion of the hyperbolic cotangent, Eq. (3.166). The result for the late-time,

but non-perturbative thermal covariance obtained in this way is

$$\sigma_T^{XX} = \frac{T}{M\Omega^2} + \frac{1}{2M\tilde{\Omega}_*} \text{Im}[\mathcal{R}], \quad (3.222)$$

$$\sigma_T^{PP} = MT + \frac{M\tilde{\Omega}_*}{2} \text{Im} \left[ \left( 1 - i \frac{\gamma_*}{\tilde{\Omega}_*} \right)^2 \mathcal{R} \right], \quad (3.223)$$

$$\mathcal{R} \equiv \frac{2}{\pi} \frac{\Lambda_* + \gamma_* - i\tilde{\Omega}_*}{\Lambda_* - \gamma_* - i\tilde{\Omega}_*} \left\{ \text{H} \left( \frac{\gamma_* + i\tilde{\Omega}_*}{2\pi T} \right) - \text{H} \left( \frac{\Lambda_*}{2\pi T} \right) \right\}, \quad (3.224)$$

where we assumed that  $\tilde{\Omega}_* = \sqrt{\Omega_*^2 - \gamma_*^2}$  is real and  $\text{H}[z]$  denotes the harmonic number function. If one expands those expressions, and the expressions below, under the assumption that  $\tilde{\Omega}_*$  is real, e.g. using  $\text{Im}[z] = (z - \bar{z})/(2i)$ , then one will have the more general expressions which will apply even in the overdamped regime.

The harmonic number function is asymptotically logarithmic and yet  $\text{H}(0) = 0$ . While the logarithmic sensitivity appears in both uncertainties, it is suppressed in the position uncertainty by inverse powers of the cutoff. For the momentum uncertainty, the logarithmic sensitivity appears already to first order in  $\gamma_0$  (which is itself quadratic in the system-environment coupling constant) and is otherwise unsuppressed. This behavior had already been noticed for Gaussian wave-packets in the Ohmic environment [79, 147], and as we have discussed in Sec. 3.4.6.2, the position uncertainty will be free of the highest cutoff sensitivities for *any* spectral density.

At **high temperature** all of the harmonic number functions vanish, leaving only the first terms in Eqs. (3.222)-(3.223), which are proportional to temperature:

$$\sigma_T^{XX} = \frac{T}{M\Omega^2} + \mathcal{O}(T^0), \quad (3.225)$$

$$\sigma_T^{PP} = MT + \mathcal{O}(T^0). \quad (3.226)$$

This corresponds to the high-temperature result of classical statistical mechanics. It is interesting that this can happen for a finite cutoff and, therefore, outside the strict Markovian limit. The reason for this classical result is that with the system and environment linear, if the FDR is also classical (from the high-temperature limit, see Sec. 2.5.2.3) then the Fokker-Plank equation must be entirely classical.

At **zero temperature** the first terms in Eqs. (3.222)-(3.223) vanish and all of the harmonic number functions can be equivalently evaluated as logarithms, so that  $\mathcal{R}$  is simplified as follows:

$$\mathrm{H}\left(\frac{\gamma_\star + i\tilde{\Omega}_\star}{2\pi T}\right) - \mathrm{H}\left(\frac{\Lambda_\star}{2\pi T}\right) = i \cos^{-1}\left(\frac{\gamma_\star}{\Omega_\star}\right) - \log\left(\frac{\Lambda_\star}{\Omega_\star}\right) + \mathcal{O}(T). \quad (3.227)$$

This generalizes the results of Unruh and Zurek [147], who explored the zero temperature regime in the limit of local dissipation.

Finally, in the **weak coupling** limit these expressions correctly reproduce the free thermal state:

$$\sigma_T^{XX} = \frac{1}{2M\Omega} \coth\left(\frac{\Omega}{2T}\right) + \mathcal{O}(\gamma_0), \quad (3.228)$$

$$\sigma_T^{PP} = \frac{M\Omega}{2} \coth\left(\frac{\Omega}{2T}\right) + \mathcal{O}(\gamma_0). \quad (3.229)$$

One can also see that at weak coupling the uncertainty function agrees with the weak coupling approximation for moderate values of the cutoff scale, as shown in Fig. 3.5. Had one naively tried to have finite diffusion in the limit  $\Lambda \rightarrow \infty$ , subtracting by hand the  $\log(\Lambda/\Omega)$  term, one would find a violation of the Heisenberg uncertainty principle at low temperature and strong coupling (see Fig. 3.6), which renders the theory unphysical. Of course this does not happen with the unsubtracted theory, as

$$\gamma_0 = \frac{\Omega}{100}$$

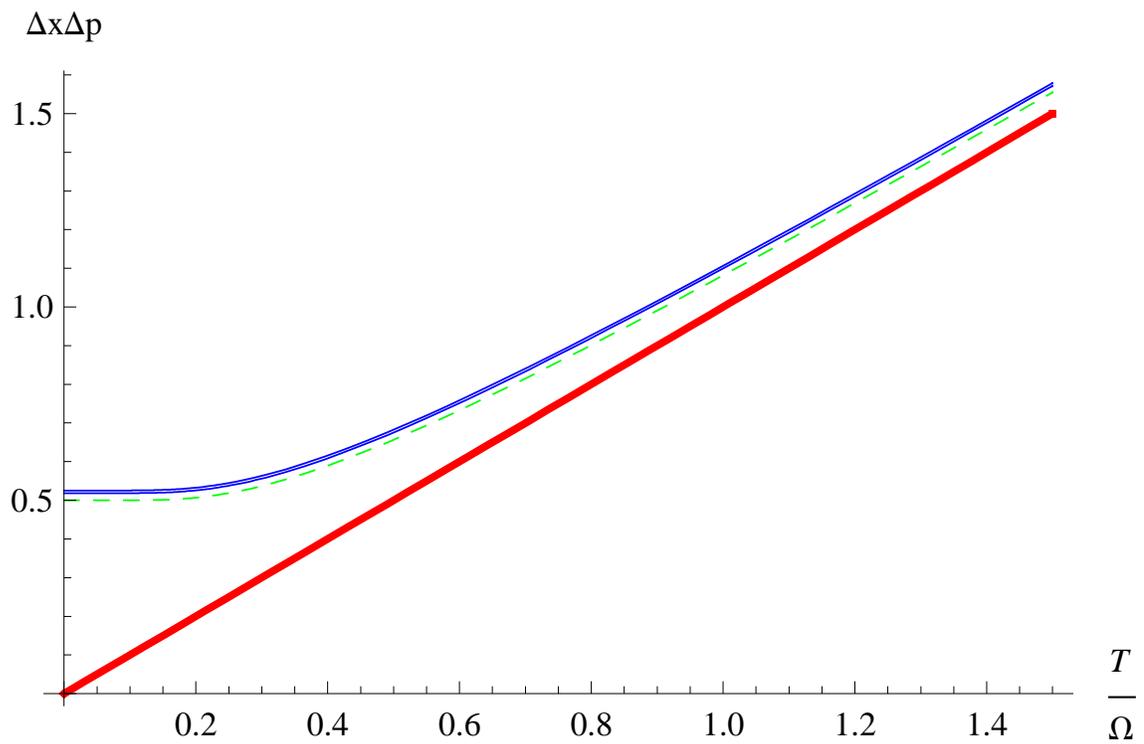


Figure 3.5: Late time  $\Delta X \Delta P$  for **• high temperature, classical statistical mechanics**, **••• weak coupling approximation  $\frac{1}{2} \coth \frac{\Omega}{2T}$  · HPZ at  $\Lambda = 10^3 \Omega$  and  $\Lambda = 10^4 \Omega$ .**

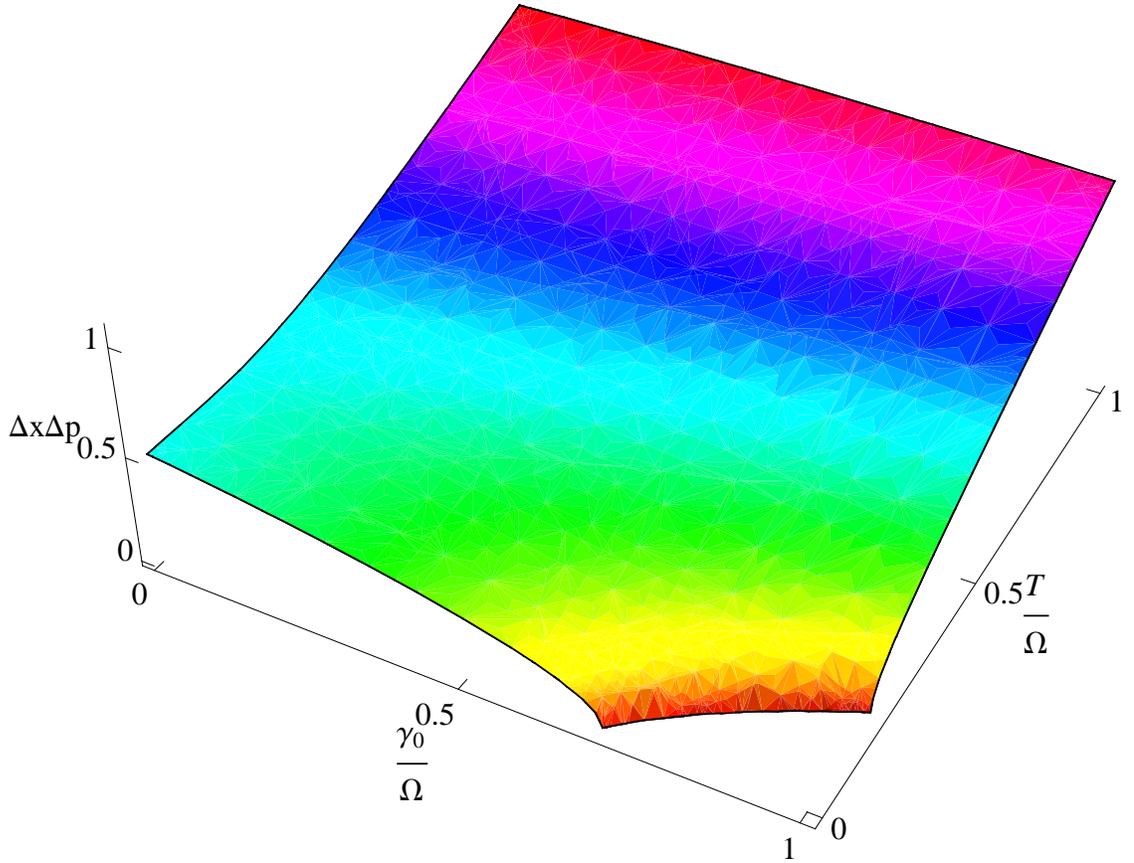


Figure 3.6: Late time  $\Delta X \Delta P$  for the unphysical, subtracted theory.

seen in Fig. 3.7. It is, thus, clear that the logarithmic dependence on the ultraviolet cutoff that appears in the diffusion is a physically important parameter and not something that can be subtracted away.

Finally, given that our results are *nonperturbative*, it is also interesting to point out what happens in the highly nonlocal **strong coupling** regime mentioned Sec. 3.7.1. The late-time thermal covariance for this case essentially corresponds to

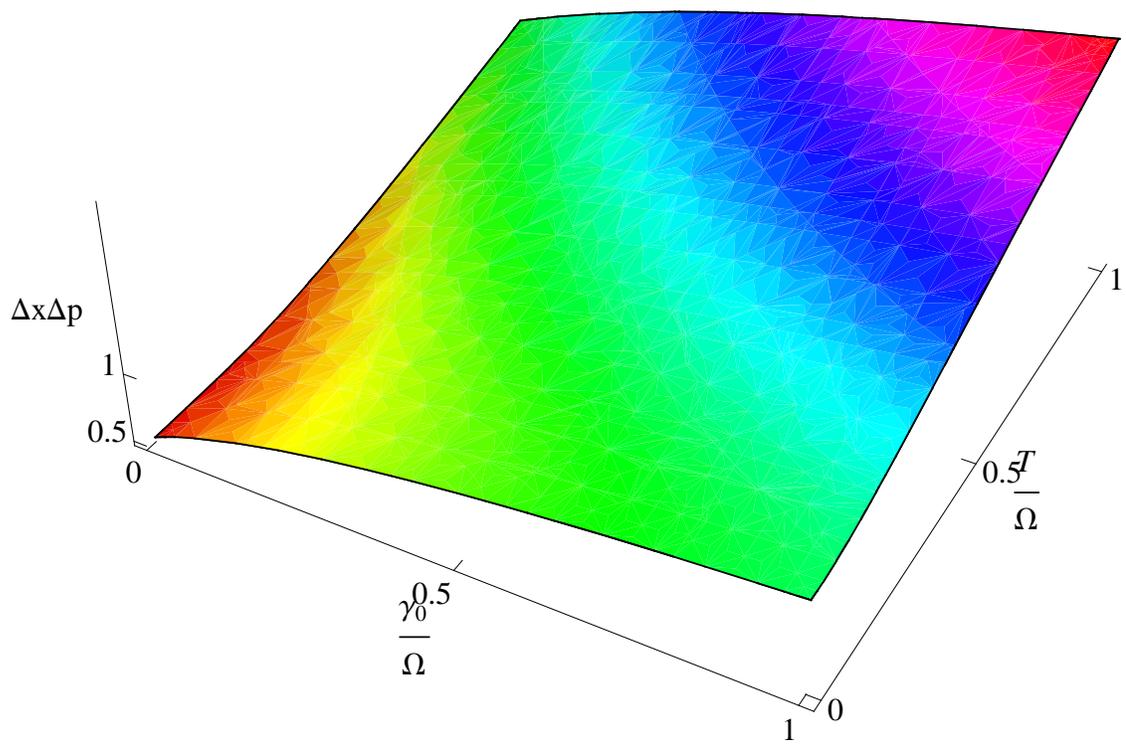


Figure 3.7: Late time  $\Delta X \Delta P$  for the  $\Lambda = 10^3 \Omega$  theory.

taking the large  $\Omega_*$  limit limit of Eqs. (3.222)-(3.223):

$$\sigma_T^{XX} = \frac{T}{M\Omega^2} + \frac{1}{2M\Omega_*} + \mathcal{O}\left(\frac{1}{\Omega_*^2}\right), \quad (3.230)$$

$$\sigma_T^{PP} = \frac{M\Omega_*}{2} + \mathcal{O}(\Omega_*^0). \quad (3.231)$$

For this model of strong coupling to the environment, and yet finite cutoff, the Brownian particle will become strongly localized in position at late time and sufficiently low temperatures. And although the particle is localized in position, the uncertainty principle is not violated but at most minimized in the zero temperature limit.

### 3.7.3 Notes on the Diffusion Coefficients

Applying our late-time covariance formulas in Sec. 3.7.2 to the Lyapunov equation (3.123) immediately determines the late-time diffusion coefficients. Expanding these expressions perturbatively in  $\gamma_0$  we get

$$D_{PP}(\infty) = M \gamma_0 \Omega \coth\left(\frac{\Omega}{2T}\right) + \mathcal{O}(\gamma_0^2), \quad (3.232)$$

$$D_{XP}(\infty) = \frac{2}{\pi} \gamma_0 \operatorname{Re} \left[ \operatorname{H}\left(\frac{\Lambda}{2\pi T}\right) - \operatorname{H}\left(\frac{i\Omega}{2\pi T}\right) \right] + \mathcal{O}(\gamma_0^2). \quad (3.233)$$

In comparison to the weak coupling master equation of Caldeira *et al.* [25], the normal diffusion coefficient is the same to lowest order in the coupling, but the anomalous diffusion coefficient is *completely absent* there. The largest contribution (in the weak coupling regime) to the anomalous diffusion coefficient comes from the cutoff and it does not vanish at finite temperature. This logarithmic sensitivity does not enter into the normal diffusion coefficient until second order, but in the

anomalous diffusion coefficient it is only proportional to one power of the coupling constant, which is the order to which the master equation of Caldeira *et al.* [25] should be valid. In this weak-coupling perturbative expansion, both diffusion coefficients are of order  $\gamma_0$  plus higher-order corrections, but they give contributions of different orders to the late-time thermal covariance  $\sigma_T^\infty$ , Sec. 3.4.6.2. Whereas  $D_{PP}^\infty$  gives contributions of order 1 because it appears multiplied by a factor  $1/\gamma_0$ ,  $D_{XP}^\infty$  gives contributions of order  $\gamma_0$ . That is why the correct thermalization in the weak-coupling limit was obtained in Ref. [25] despite having completely neglected the anomalous diffusion coefficient. The non-equilibrium dynamics are, in fact, incorrect in this regard.

### 3.7.4 Full-Time Diffusion at High Cutoff

In this section we calculate the diffusion coefficients for the Ohmic case using the local damping for the propagator, which is a valid approximation in the high cutoff regime, as discussed in Sec. 3.7.1. The big advantage of using local damping is that the diffusion coefficients in Eq. (3.120) experience a considerable number of simplifications and ultimately reduce to a single time integral. Furthermore, the Laplace transforms of the corresponding equations for the diffusion coefficients exhibit a rather simple form if one takes the following steps. First, one writes the cosine of the noise kernel in exponential form; next, manipulates the time integral until one has a Laplace convolution; and then uses frequency shifting in the Laplace

domain, i.e.  $e^{\lambda t} f(t) \rightarrow \hat{f}(s-\lambda)$ . After some algebraic manipulations one finally gets

$$\hat{D}_{xp}(s) = -\frac{1}{s} \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \tilde{\nu}(\omega) \operatorname{Re} \left[ \hat{G}(s+i\omega) \right], \quad (3.234)$$

$$\hat{D}_{pp}(s) = +\frac{1}{s} \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \tilde{\nu}(\omega) \operatorname{Re} \left[ \hat{G}(s+i\omega) \right]. \quad (3.235)$$

Our Green function is rational in the Laplace domain, as is our damping kernel in the Fourier domain, given by Eq. (3.209). Together with the rational expansion of the hyperbolic cotangent in Eq. (3.166), this implies that the frequency integrals over  $\omega$  in the above diffusion coefficients become sums over  $k$  of trivial contour integrals in the Laplace domain. Still in the Laplace domain, these sums can be identified as harmonic number functions (or, equivalently, digamma functions): <sup>1</sup>

$$\hat{D}_{xp}(s) = -\frac{2\gamma_0 T}{\Lambda s} \mathcal{F}_s + \frac{\gamma_0}{s} \operatorname{Im}[\mathcal{I}_s], \quad (3.236)$$

$$\hat{D}_{pp}(s) = \frac{2\gamma_0 T}{s} \left(1 + \frac{s}{\Lambda}\right) \mathcal{F}_s + \frac{\gamma_0}{s} \operatorname{Im} \left[ \left(\gamma_0 + i\tilde{\Omega}\right) \mathcal{I}_s \right], \quad (3.237)$$

in terms of the dimensionless quantities  $\mathcal{F}_s$  and  $\mathcal{I}_s$  defined

$$\mathcal{F}_s \equiv \left[ \left(1 + \frac{\gamma_s}{\Lambda}\right)^2 + \left(\frac{\tilde{\Omega}}{\Lambda}\right)^2 \right]^{-1}, \quad (3.238)$$

$$\mathcal{I}_s \equiv \frac{2}{\pi} \frac{i + \frac{\gamma_s}{\tilde{\Omega}}}{1 - \left(\frac{\gamma_s + i\tilde{\Omega}}{\Lambda}\right)^2} \left\{ \operatorname{H}\left(\frac{\Lambda}{2\pi T}\right) - \operatorname{H}\left(\frac{\gamma_s + i\tilde{\Omega}}{2\pi T}\right) \right\}, \quad (3.239)$$

and where  $\gamma_s = \gamma_0 + s$ . Note that by making use of the final-value theorem, we only need to discard the overall  $1/s$  factor and replace  $\gamma_s$  with  $\gamma_0$  in Eqs. (3.236)-

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<sup>1</sup>Many of the expressions derived throughout this paper assume underdamping, i.e.  $\gamma_0 < \Omega$  with  $\tilde{\Omega} = \sqrt{\Omega^2 - \gamma_0^2}$ . They can be used for the overdamping regime by making the following analytical continuation:  $\tilde{\Omega} \rightarrow i\tilde{\gamma}$  with  $\tilde{\gamma} = \sqrt{\gamma_0^2 - \Omega^2}$  real. Therefore, Eqs. (3.236)-(3.237) can be applied to the overdamping case if the Im and Re terms are first expanded assuming that  $\tilde{\Omega}$  is real, e.g. using  $\operatorname{Im}[z] = (z - \bar{z})/(2i)$ , and then the analytical continuation  $\tilde{\Omega} \rightarrow i\tilde{\gamma}$  is made.

(3.237) to obtain the late-time asymptotic values  $D_{xp}(\infty)$  and  $D_{pp}(\infty)$ . The  $H(z)$  functions are harmonic number function, which is asymptotically logarithmic but with  $H(0) = 0$ . These terms make up, among other things, the well known  $\log(\Lambda/\Omega)$  divergence.

The diffusion coefficients can be expressed in the time domain as their asymptotic values plus damped oscillating differential operators acting on the same decay function  $DF(t)$  (although the sums over  $k$  cannot in general be identified with any simply behaved special functions):

$$D_{xp}(t) = D_{xp}(\infty) - M\gamma_0 \left\{ \dot{G}(t) + G(t) \left( 2\gamma_0 - \frac{d}{dt} \right) \right\} DF(t), \quad (3.240)$$

$$D_{pp}(t) = D_{pp}(\infty) - M\gamma_0 \left\{ \dot{G}(t) \left( \gamma_0 + \frac{d}{dt} \right) + G(t) \Omega^2 \right\} DF(t), \quad (3.241)$$

with the thermal decay function

$$DF(t) = -\frac{\cot\left(\frac{\Lambda}{2T}\right) e^{-\Lambda t}}{\left(1 + \frac{\gamma_0}{\Lambda}\right)^2 + \left(\frac{\tilde{\Omega}}{\Lambda}\right)^2} + \frac{2}{\pi} TS(t), \quad (3.242)$$

$$TS(t) = \sum_{k=1}^{\infty} \frac{\left(\frac{\Lambda}{2\pi T}\right)^2}{\left(\frac{\Lambda}{2\pi T}\right)^2 - k^2} \frac{k e^{-2\pi T k t}}{\left(k + \frac{\gamma_0}{2\pi T}\right)^2 + \left(\frac{\tilde{\Omega}}{2\pi T}\right)^2}. \quad (3.243)$$

For numerical evaluation purposes, it is useful to express this thermal sum in terms of Lerch transcendent functions:

$$TS(t) = \operatorname{Re} \left[ \frac{1 - i\frac{\gamma_0}{\tilde{\Omega}}}{1 - \left(\frac{\gamma_0 + i\tilde{\Omega}}{\Lambda}\right)^2} \Phi_1 \left( \frac{\gamma_0 + i\tilde{\Omega}}{2\pi T}; 2\pi T t \right) \right] - \operatorname{Sy}_{\Lambda} \left[ \frac{\Phi_1 \left( \frac{\Lambda}{2\pi T}; 2\pi T t \right)}{\left(1 - \frac{\gamma_0}{\Lambda}\right)^2 + \left(\frac{\tilde{\Omega}}{\Lambda}\right)^2} \right], \quad (3.244)$$

with the definitions of  $\Phi_1(z; \lambda)$ , which is related to the more canonical *Lerch tran-*

scendent function by  $\Phi_1(z; \lambda) = \Phi(e^{-\lambda}, 1, z) - 1/z$ , and of the symmetric part being

$$\Phi_1(z; \lambda) = \sum_{k=1}^{\infty} \frac{e^{-\lambda k}}{k+z}, \quad (3.245)$$

$$\text{Sy}_z[f(z)] = \frac{f(+z) + f(-z)}{2}. \quad (3.246)$$

The decay function is such that at the initial time it causes cancellation with the asymptotic values and the diffusion coefficients vanish. In this (asymptotic) high temperature perspective, the decay function contains two terms. The first decays at a cutoff dependent rate and can be expressed in closed form. The second decays with primarily temperature dependent rates and cannot be expressed in closed form with intuitive functions. It contains the initial time cancellation of the  $\log(\Lambda/\Omega)$  divergence. Although well convergent at moderate times, the sum's contribution to the regular diffusion coefficient is very slow to converge at the initial time, even for moderate temperatures; see Fig. 3.9.

While our expressions (3.236)-(3.237) can easily give us the zero temperature diffusion coefficients at asymptotically late time, they cannot easily give us the corresponding moderate time behavior in closed form. Moreover, the zero temperature limit of  $\coth(\omega/2T) \rightarrow \text{sign}(\omega)$  means that our diffusion coefficient integrals cannot be cast as closed contour integrals. Nevertheless, the frequency integrals can be performed and the results expressed in terms of exponential integrals with predictable time scales. At zero temperature (and in the high cutoff limit) we find the decay function to take the following form:

$$\lim_{T \rightarrow 0} \text{DF}(t) = \frac{2}{\pi} \frac{d}{dt} \left\{ \text{Re} \left[ \frac{\text{E}_1\left(\left[\gamma_0 + i\tilde{\Omega}\right]t\right)}{i\tilde{\Omega} e^{-(\gamma_0 + i\tilde{\Omega})t}} \right] - \text{Sy}_\Lambda \left[ \frac{\text{E}_1(\Lambda t)}{\Lambda e^{-\Lambda t}} \right] \right\}, \quad (3.247)$$

where  $E_1(z)$  is the exponential integral, which behaves like  $e^{-z}/z$  for large  $z$ . It should be noted that unlike the asymptotic limits of the diffusion coefficients, the full time behavior is highly sensitive to the form of the cutoff regulator at low temperature. For our smooth regulator, we find relatively smoothly evolving diffusion coefficients (similar to the result in Ref. [84] at  $T = 10\Omega$ ) all the way down to zero temperature. In contrast, a sharp cutoff of the form  $I(\omega) \propto \theta(\omega - \Lambda)$  would produce the same average behavior, but with a slowly decaying envelope modulating with considerable oscillations at the cutoff frequency.

Analogous functions appear when we approximate the thermal sum in (3.243) [together with the first term on the right-hand side of (3.242), which cancels any spurious poles at  $\Lambda = 2\pi Tk$ ] as an integral with a comparably soft cutoff:

$$\sum_{k=1}^{\infty} \frac{\left(\frac{\Lambda}{2\pi T}\right)^2}{\left(\frac{\Lambda}{2\pi T}\right)^2 - k^2} f(k) \approx \int_{k_i}^{\infty} dk \frac{\left(\frac{\Lambda}{2\pi T}\right)^2}{\left(\frac{\Lambda}{2\pi T}\right)^2 + k^2} f(k), \quad (3.248)$$

where  $k_i \approx 1$ . Still in the high cutoff limit, we find this qualitative approximation of the decay function to be

$$\text{DF}(t) \approx \frac{2}{\pi} \frac{d}{dt} \left\{ \text{Re} \left[ \frac{E_1\left(\left[2\pi Tk_i + \gamma_0 + i\tilde{\Omega}\right]t\right)}{i\tilde{\Omega} e^{-(\gamma_0 + i\tilde{\Omega})t}} \right] - \text{Sy}_{\Lambda} \left[ \frac{E_1\left([2\pi Tk_i + i\Lambda]t\right)}{i\Lambda e^{-i\Lambda t}} \right] \right\}, \quad (3.249)$$

where we have discarded all finite terms at the initial time which decay at cutoff rates, as our approximation ultimately ruins the behavior of  $\text{DF}(t)$  there. Thus, when using this approximate decay function, the time-dependent, decaying part of the diffusion coefficients must be “clamped” at the initial time. At moderate times, our approximation reveals the exact same form of exponential integral behavior as in the zero temperature limit, and the two functions are compared in Fig. 3.8. But

$$\gamma_0 = \frac{\Omega}{10} \quad T=0$$

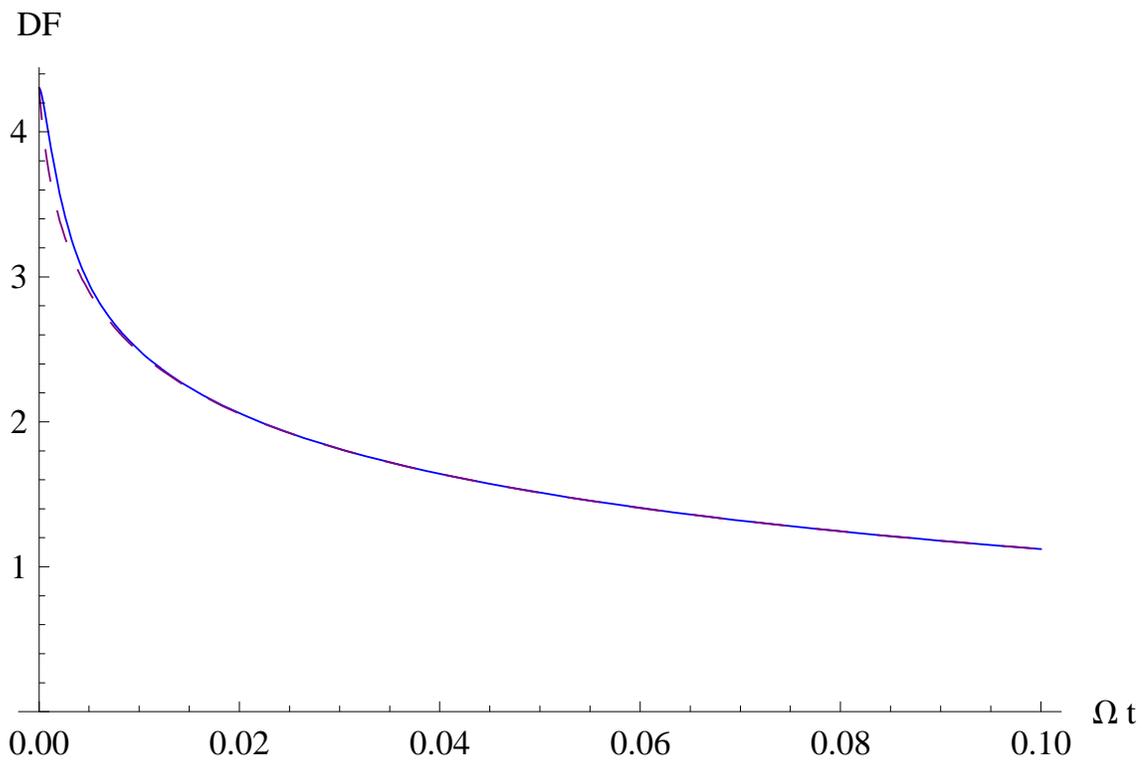


Figure 3.8: Zero temperature decay functions for  $\cdot$  zero temperature,  $\cdots$  qualitative approximation at  $\Lambda = 10^3 \Omega$ . The slopes differ near the initial time (within the cutoff time scale).

$$\gamma = \frac{\Omega}{10} \quad T = 10\Omega$$

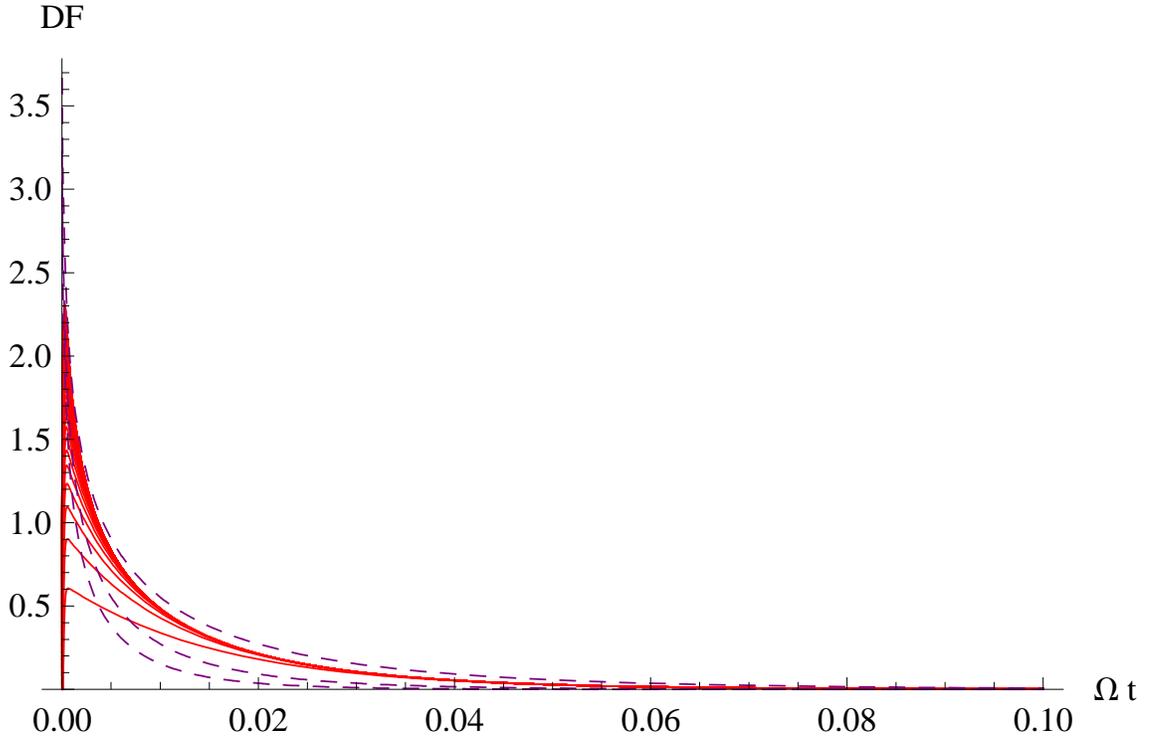


Figure 3.9: Moderate temperature decay functions for  $\cdot$  a sequence of the first 50 high temperature sums,  $\cdots$  qualitative approximations for  $k_i = \frac{1}{2}, 1, 1\frac{1}{2}$  at  $\Lambda = 10^5\Omega$ . The high temperature sums are very slow to converge at the initial time.

the temperature enters in such a way that the exponential decay inherent in  $E_1$  is not balanced out with a  $e^{-2\pi T k_i t}$  factor. Therefore, temperature is an inherently stronger relaxation scale here [although there are additional  $e^{-\gamma_0 t}$  factors from  $G(t)$  functions in the full diffusion coefficients].

### 3.8 Sub-Ohmic Coupling with no Cutoff

As an example where the nonlocal effects of dissipation are important, we will consider one of the most common and well-behaved sub-Ohmic spectral densities,  $\tilde{\gamma}(\omega) \propto \omega^{-1/2}$ , which requires neither a U.V. nor an I.R. cutoff in the final results (although one still needs to renormalize the frequency introducing a logarithmically divergent bare counterterm). Our formulas will take a simpler form if we express our damping kernel in terms of a phenomenological damping coefficient  $\gamma_*$  as follows:

$$\tilde{\gamma}(\omega) = 2\gamma_*\sqrt{\frac{\omega_*}{|\omega|}}, \quad (3.250)$$

$$\omega_*^2 \equiv \Omega^2 + \gamma_*^2. \quad (3.251)$$

It is then a straightforward calculation to find the propagator

$$\hat{G}(s) = \frac{\frac{1}{M}}{s^2 + 2\gamma_*\sqrt{2\omega_*}s + \Omega^2}, \quad (3.252)$$

which is amenable to partial fraction decomposition in  $\sqrt{s}$  since  $s$  is strictly positive. As we have defined our nonlinear coupling strength in anticipation of this polynomial, the roots of the quartic denominator  $r_k : k \in \{1, 2, 3, 4\}$  can be shown to be the conjugate pairs

$$r_{1,2} = \frac{1}{\sqrt{2}} \left( +\sqrt{\omega_*} \pm i\sqrt{\omega_* + 2\gamma_*} \right), \quad (3.253)$$

$$r_{3,4} = \frac{1}{\sqrt{2}} \left( -\sqrt{\omega_*} \pm i\sqrt{\omega_* - 2\gamma_*} \right). \quad (3.254)$$

After partial fraction decomposition, we may cast our propagator in the form

$$\hat{G}(s) = \sum_{k=1}^4 \frac{A_k}{M} \frac{1}{\sqrt{s - r_k}}, \quad (3.255)$$

$$A_j = \prod_{\substack{k=1 \\ k \neq j}}^4 \frac{1}{r_j - r_k}, \quad (3.256)$$

with inverse Laplace transform

$$G(t) = \sum_{k=1}^4 \frac{A_k}{M} r_k e^{r_k^2 t} \operatorname{erfc}(-r_k \sqrt{t}), \quad (3.257)$$

where  $\operatorname{erfc}(z)$  is the cumulative error function of the normal distribution. There are additional terms from the individual Laplace transforms, like  $t^{-1/2}$ , but they vanish in the sum. Using the asymptotic expansion of  $\operatorname{erfc}(z)$  in order to expand the Green function in Eq. (3.257) at late times, we obtain terms of the form

$$z e^{z^2} \operatorname{erfc}(z) = \frac{1}{\sqrt{\pi}} \sum_{k=0}^{\infty} (-1)^k \frac{(2k)!}{k!} \frac{1}{(2z)^{2k}} + \begin{cases} 0 & \operatorname{Re}[z] \geq 0 \\ 2z e^{z^2} & \operatorname{Re}[z] \leq 0 \end{cases}, \quad (3.258)$$

which we can use to expand the Green function in Eq. (3.257). After grouping all the contributions together, we will find exponential terms with characteristic frequencies  $f = -\gamma_\star \pm i\sqrt{\omega_\star^2 + 2\gamma_\star\omega_\star}$ , which are actually the solutions to the characteristic rate equation (3.149) with smallest *negative* real part. These are the only terms that one would have considered if the local propagator  $G_\infty(t)$  within the late-time approximation of Sec. 3.4.6 had been employed. In addition, and more importantly are the power-law decay terms which admit no local representation.

This sub-Ohmic model provides a perfect example showing when effectively local treatments, such as that in Sec. 3.4.6, will fail. At first the local contribution will dominate and the master equation coefficients will appear to trend towards  $\gamma_L \approx \gamma_\star$

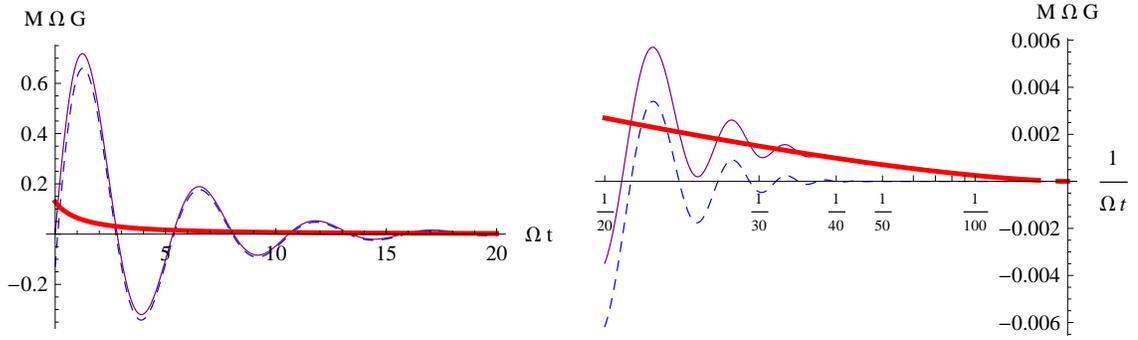


Figure 3.10: Asymptotic expansion of sub-Ohmic  $\cdot$  propagator  $G(t)$  into  $\cdots$  the local contribution and  $\bullet$  the nonlocal contribution for  $\gamma_\star = \frac{\Omega}{4}$ . The local contribution is initially more significant, but the nonlocal contribution dominates eventually.

and  $\Omega_L \approx \omega_\star + \gamma_\star$ . However, as shown in Fig. 3.10, the nonlocal contribution (the power-law terms) will eventually dominate the more swiftly decaying local contribution (the exponential terms) and a correct treatment of the nonlocal dynamics will be required. In fact, as the nonlocal contribution becomes comparable to the local contribution, the master equation coefficients will become periodically divergent, as can be seen in Fig. 3.11. This is related to the fact that  $\det[\Phi(t)]$  vanishes and changes sign at those times. The underlying homogeneous evolution is well behaved and strictly dissipative (the damping kernel is positive definite), but the localizing perspective of the master equation becomes divergently unnatural. Any errors, numeric or analytic, can be catastrophic in the master equation perspective. In this respect, the subtleties missed in previous derivations of the master equation, as pointed out in Sec. 3.4.2.1 and Ref. [64] and which are relevant whenever nonlocal effects are important, will likely give rise to substantial discrepancies in this case.

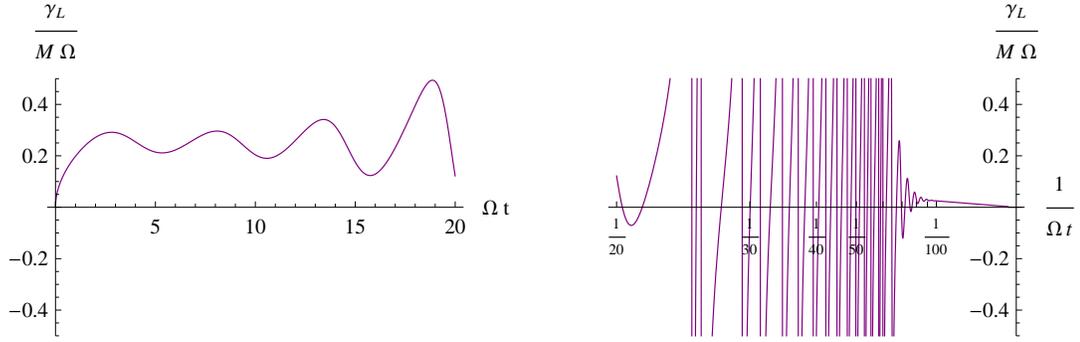


Figure 3.11: Exact time-local damping coefficient for  $\gamma_* = \frac{\Omega}{4}$ . The transition between local and nonlocal domination is highly erratic.

The full-time evolution is rather complicated, but the late-time limit is very manageable. From Eq. (3.173) we can express the late-time thermal position uncertainty as

$$\sigma_T^{XX}(\infty) = \frac{1}{\pi} \int_0^\infty \tilde{\gamma}(\omega) \coth\left(\frac{\omega}{2\pi T}\right) |\hat{G}(i\omega)|^2 2\sqrt{\omega} d\sqrt{\omega}, \quad (3.259)$$

where we have used the relation  $d\omega = 2\sqrt{\omega} d\sqrt{\omega}$ . The integrand is amenable to partial fraction decomposition, after a rational expansion of the hyperbolic cotangent with Eq. (3.166), and can therefore be integrated without resorting to numerics. Additionally, and in contrast to the Ohmic case, the integrand is even in  $\sqrt{\omega}$  for all temperatures, including zero, and contour integration techniques are more generally applicable.

Strictly speaking we cannot compare exact sub-Ohmic solutions to those obtained with an incorrect master equation since the master equation will yield nonsense, but we can compare the exact nonlocal dynamics to those obtained by extracting the local dynamics and assuming it to be the dominant behavior. Obviously

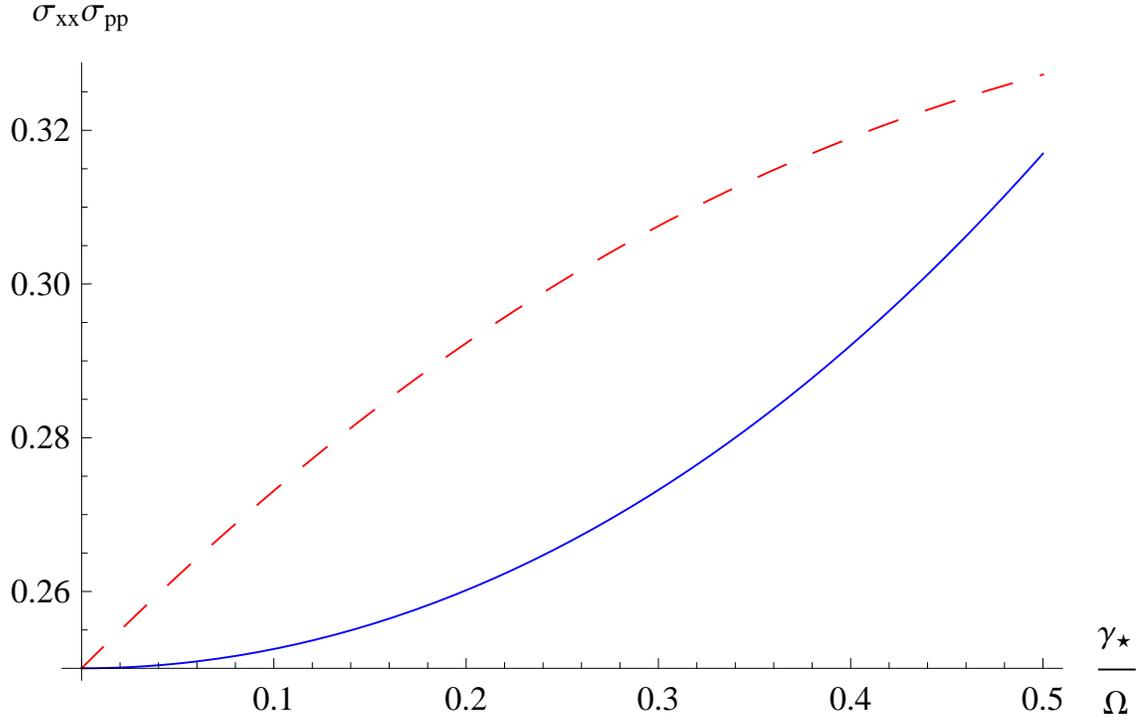


Figure 3.12: Late-time sub-Ohmic uncertainty function at zero temperature with the **· exact nonlocal solution** and **· · · fictitious effectively local solution**. In the limit of vanishing dissipation, one has the minimal uncertainty ground state (zero temperature thermal state) in each case.

the effectively local approximation is incorrect, but it should be good to zeroth order in the coupling and one might naively expect that it might also behave reasonably for finite coupling strength. However, in Fig. 3.12 we compare the late-time uncertainty functions and show there to be sharp disagreement to the first two orders in the coupling constant squared (the slope and the curvature of the curves on the plot).

### 3.8.1 Supra-Ohmic with Finite Cutoff

The conventional wisdom has been to consider supra-Ohmic damping kernels of the form

$$\tilde{\gamma}_n(\omega) = 2 \gamma_n \left(\frac{\omega}{\Lambda}\right)^n \chi\left(\frac{\omega}{\Lambda}\right), \quad (3.260)$$

where  $\chi : [0, \infty) \rightarrow [1, 0)$  denotes the cutoff regulator. Without a cutoff regulator, all supra-Ohmic couplings have greater than logarithmic high frequency divergence in the diffusion and thermal covariance integrals (see Sec. 3.4.5.3). Even when regulated, the mere potential for divergence therefore corresponds to cutoff sensitivity from the high frequency portion of noise integrals, which is balanced by the extra inverse powers of the cutoff in the pre-factor of the above spectral density.

Here we will restrict our investigation to the following damping kernel

$$\tilde{\gamma}(\omega) = \frac{2 \gamma_2 \left(\frac{\omega}{\Lambda}\right)^2}{\left(1 + \left(\frac{\omega}{\Lambda}\right)^2\right)^2}, \quad (3.261)$$

because this example is exactly solvable. The corresponding damping kernel in Laplace space is

$$\hat{\gamma}(s) = \frac{\gamma_2}{2} \frac{\frac{s}{\Lambda}}{\left(1 + \frac{s}{\Lambda}\right)^2}. \quad (3.262)$$

Upon asymptotic expansion in  $1/\Lambda$ , might be inclined to view this damping kernel's effect in the Langevin equation as a tiny mass renormalization plus even less significant higher order terms, but the effect quite different from that, as we will see. After factoring the fourth-order polynomial, the fully nonlocal propagator can be decomposed by partial fractions into two sets of timescales. Expanding perturbatively in  $\gamma_2$ , the first set of timescales correspond to the system frequency with weak

damping

$$\gamma_\star = \gamma_2 \frac{\left(\frac{\Omega}{\Lambda}\right)^2}{\left(1 + \left(\frac{\Omega}{\Lambda}\right)^2\right)^2} + \mathcal{O}(\gamma_2^2), \quad (3.263)$$

$$\Omega_\star = \Omega \left( 1 - \frac{\gamma_2}{\Lambda} \frac{1 - \left(\frac{\Omega}{\Lambda}\right)^2}{\left(1 + \left(\frac{\Omega}{\Lambda}\right)^2\right)^2} + \mathcal{O}(\gamma_2^2) \right), \quad (3.264)$$

while the second set of timescales correspond to quickly decaying nonlocal contributions associated with the cutoff scale:

$$\gamma_\Lambda = \Lambda - \gamma_\star, \quad (3.265)$$

$$\Omega_\Lambda = \frac{\Omega}{\Omega_\star} \Lambda. \quad (3.266)$$

The situation is analogous to Ohmic case with a finite cutoff except that the nonlocal part of the propagator is also oscillating at the rate  $\tilde{\Omega}_\Lambda \approx \sqrt{\gamma_2 \Lambda}$ , for weak coupling and high cutoff.

This form of damping kernel was constructed only with well-behaved high frequency contributions in mind. Nevertheless, as shown in Fig. 3.13, we find the conventional form of spectral density or damping kernel to be inadequate. There is clearly some cutoff sensitivity in the thermal covariance which is remedied by introducing an additional power of cutoff suppression. E.g. the conventional form of spectral density is not well behaved, but the substitution

$$\gamma_2 \rightarrow \frac{\Omega}{\Lambda} \gamma_2, \quad (3.267)$$

is well behaved.

An explanation only emerges after a more thorough examination of the contour integrals. The high-frequency regime,  $\omega \gg \Lambda$ , is already rendered well behaved

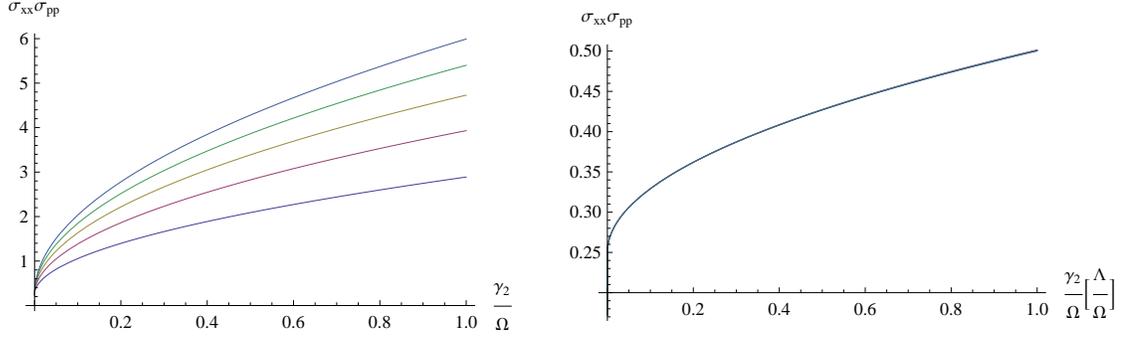


Figure 3.13: Late-time supra-Ohmic uncertainty function at zero temperature for cutoffs between  $100\Omega$  and  $500\Omega$ . The left plot is with a conventional coupling scale, while the right plot has decreased the coupling strength by an extra power of the cutoff.

by the conventional cutoff-dependent prefactor. The near-resonance regime,  $\omega \approx \Omega$ , which produces the weak coupling limit, also appears to be well behaved. There is only one remaining suspect and it proves to be the culprit. The previously unaccounted for cutoff sensitivity arises here from the nonlocal timescales of the propagator, i.e. the  $\omega \approx \Lambda$  regime. This is quite surprising as unlike sub-Ohmic coupling, supra-Ohmic coupling does yield a well-behaved local representation for its late-time dynamics. But residues of the contour integral which correspond to the nonlocal timescales reveal the correct dominant behavior  $\sigma_{PP} \approx \frac{1}{2}M\tilde{\Omega}_\Lambda = \frac{1}{2}M\sqrt{\gamma_2\Lambda}$ , for weak coupling and high cutoff. Therefore the conventional, linear coupling  $\gamma_2$  must be suppressed by an additional factor of the cutoff, else the momentum covariance will be plagued by a  $\sqrt{\Lambda}$  sensitivity.

### 3.9 Conclusions

Quantum Brownian motion of an oscillator coupled to a thermal reservoir of quantum oscillators has been the canonical model for the study of open quantum systems where one can use it to investigate all the environmental effects on an open quantum system it interacts with, even of macroscopic scale, such as quantum dissipation, diffusion, decoherence and entanglement. It also provides important information on quantum measurement, such as noise, fluctuations, correlations, uncertainty relation and standard quantum limit in mesoscopic systems. Many experiments have been carried out for testing these processes. An exact master equation was reported some years ago [84] governing the reduced density matrix of an open quantum system coupled to a general environment of arbitrary spectral density and temperature. Subsequently there have also been claims of exact solutions [69] (and also, previous to the master equation [77]). We have found many previous derivations to be correct for local damping, but containing errors or omissions for nonlocal damping; in their place we have presented the most complete and correct derivation of the QBM master equation and solutions to date. In this work we report on solutions to this equation for a fairly general set of physical conditions and a generalization of the QBM master equation to a system with an arbitrary number of oscillators. Most of the previous results required one to solve integro-differential equations numerically, whereas for time-constant parameters and stationary kernels we have reduced everything to quadrature, which can be further simplified in many cases using contour integration techniques. We expect these results to be useful in

realistic settings for the analysis of many problems which can be described by this model. In Chapter 6 we apply these results to the consideration of multiple local oscillators in a common field.

As a complete generalization of the QBM master equation we have included the influence of external forces and not limited our setup to one system oscillator. These results may be useful for the study of low-temperature measurements of driven oscillators, which are relevant for experiments with nanomechanical resonators [119, 102]. They also play a crucial role in future schemes for the detection of gravitational waves with high-intensity laser interferometers, where the radiation pressure effects on the cavity mirrors are important [95, 24]. Furthermore, simple linear models may allow for the preparation of non-classical states in mesoscopic objects through state swapping (via phase-space rotations) [149].

More specifically, we have found a compact expression for the general solution of this master equation, showing that at late times it tends to a Gaussian state entirely characterized by its asymptotic covariance matrix. For meromorphic damping kernels, and many others, the result for this late-time covariance matrix can be evaluated as a simple contour integral. As an example we provide explicit exact nonperturbative results for an Ohmic environment with a finite cutoff which are valid for an arbitrarily strong coupling. At sufficiently low temperatures and strong coupling this equilibrium state becomes highly squeezed and the system becomes extremely localized in position space, a phenomenon with potentially interesting applications in the realm of mesoscopic systems.

The general solution of the master equation involves the matrix propagator of

a linear integro-differential equation. We have been able to solve these equations exactly for several Ohmic, sub-Ohmic and supra-Ohmic environments with a finite cutoff and studied the evolution of the system for finite times. This is achieved using Laplace transforms and eventually transforming back to time domain. From such exact (and simple) solutions for the propagator one gains highly valuable information. For instance, one can justify that using the local propagator is a valid approximation for the Ohmic environment in the large cutoff limit. This approximation leads to great simplifications and we are then able to provide relatively simple analytic expressions for the diffusion coefficients of the master equation at all times. Similarly, our exact solutions for the propagator in specific examples of sub-Ohmic and super-Ohmic environments reveal a dominant contribution from nonlocal damping effects. In the first case it is a consequence of long-time correlations, due to the low-frequency modes of the environment, that become important at late times. In contrast, the source of nonlocality in the supra-Ohmic case is the UV regulator function, and it gives rise to a marked cutoff sensitivity of the momentum covariance which had not been noticed so far. On the other hand, it should be pointed out that although the results for the exact propagator of the integro-differential equation are rather simple, some of the general expressions for the solutions of the master equation are rather lengthy and have not been reported here. They have, nevertheless, been employed to evaluate and plot the exact time evolution of the thermal covariance for an Ohmic environment with a finite cutoff in Sec. [3.5.2.2](#).

It is important to discuss the cutoff sensitivity of the late-time covariance and diffusion coefficients for an Ohmic environment in the weak coupling regime.

While  $\sigma_{XX}^\infty$  is finite in the infinite cutoff limit,  $\sigma_{PP}^\infty$  depends logarithmically on  $\Lambda$  for large  $\Lambda$  already at order  $\gamma_0$  [Eq. (3.223)]. This means that it is absolutely necessary to consider a finite cutoff. The kind of divergences that appear otherwise cannot be dealt with by renormalizing the frequency or other bare parameters of the theory. In fact, as shown in Sec. 3.7.2, subtracting the divergent term would lead to inconsistencies (violation of Heisenberg’s uncertainty principle). Furthermore, from the late-time thermal covariance one can immediately obtain the late-time diffusion coefficients as well (see the discussion at the end of Sec. 3.7.3). One finds then that both the normal and anomalous diffusion coefficients are logarithmically sensitive to large cutoffs. However, while this dependence appears in  $D_{XP}$  [Eq. (3.233)] at order  $\gamma_0$ , in  $D_{PP}$  it only appears at order  $\gamma_0^2$ , and it had been missed in previous analytic studies which treated  $\gamma_0$  perturbatively to lowest order.

We would also like to stress the following point. When studying an Ohmic environment with a finite but large cutoff, it can be a good approximation to consider local damping (infinite cutoff limit for the damping kernel) while keeping the cutoff finite in the noise kernel. This has already been discussed above and justifies calculations like those of Ref. [147] up to corrections suppressed by inverse powers of the cutoff. However, the opposite is not true: it is essential to keep a finite cutoff in the noise kernel to avoid the divergences discussed in the previous paragraph. This is precisely the origin of the divergences and pathological behavior found in Ref. [69], where a finite cutoff was employed in the damping kernel but not in the noise kernel. Instead one should use the same spectral function everywhere, which means having a finite cutoff in both kernels, and everything would be well defined

then. Note that these divergences would appear in the momentum covariance even at asymptotically late times, as discussed in the previous paragraph. There is a different kind of sensitivity to large values of the cutoff that is due to having started with a uncorrelated state for the system and the environment. This gives rise to a jolt in the normal diffusion coefficient at early times of order  $1/\Lambda$  with an amplitude proportional to  $\Lambda$ , as well as a logarithmic dependence on the cutoff of  $\sigma_{XX}$  (and  $\sigma_{PP}$ ) that decays exponentially with the relaxation time-scale  $1/\Gamma$ . They would not be present if one had started with an appropriately correlated initial state, and then prepared the system in a finite time (not suddenly).

## Chapter 4

### General Theoretical Results

#### 4.1 Overview

In this chapter we discuss some more pointed results of our perturbative formalism. In Sec. 4.2 we more carefully consider the solutions of perturbative master equations. We demonstrate that, contrary to intuition, full-time solutions of order- $2n$  accuracy require an order- $(2n+2)$  master equation. Furthermore, we show how this accuracy reduction arises in a specific example for which exact solutions are available. This result has a wide-ranging impact on the validity of coupling (or friction) sensitive results derived from second-order convolutionless, Nakajima-Zwanzig, Redfield, and Born-Markov master equations.

In Sec. 4.3 we discuss the dynamical construction of properly correlated initial states. The dependence of the dynamics of open quantum systems upon initial correlations between the system and environment is an utterly important yet poorly understood subject. For technical convenience most prior studies assume factorizable initial states where the system and its environments are uncorrelated, but these conditions are not very realistic and give rise to peculiar behaviors. One distinct feature is the rapid build up or a sudden jolt of physical quantities immediately after the system is brought in contact with its environments. The ultimate cause of this is an initial imbalance between system-environment correlations and coupling. In this

section we demonstrate explicitly how to avoid these unphysical behaviors by proper adjustments of correlations and/or the coupling, for setups of both theoretical and experimental interest. We provide simple analytical results in terms of quantities that appear in linear (as opposed to affine) master equations derived for factorized initial states.

In Sec. 4.4 we provide an in-depth and thorough treatment of the validity of the rotating-wave approximation (RWA) in an open quantum system. We find that when it is introduced after tracing out the environment, all timescales of the open system are correctly reproduced, but the details of the quantum state may not be. The RWA made before the trace is more problematic: it results in incorrect values for environmentally-induced shifts to system frequencies, and the resulting theory has no Markovian limit.

In Sec. 4.5 section we expand upon the equilibrium states of open systems. Attention is given to the equilibrium state of the open system, where one only has the free Boltzmann distribution  $e^{-\beta\mathbf{H}}$  for vanishing system-environment interaction (though this may also happen in other, very specific approximations [73]). We explicitly show that the master equation, Schrödinger equation, and Boltzmann state for the full system + environment + interaction are all consistent at second order.

In Sec. 4.6, we discuss our previously mentioned *fluctuation-dissipation inequality*. The *fluctuation-dissipation relation* is usually formulated for a system interacting with a heat bath at finite temperature in the context of linear response theory, where only small deviations from the mean are considered. We have shown

that for an open quantum system interacting with a non-equilibrium environment, where temperature is no longer a valid notion, a fluctuation-dissipation inequality exists. Clearly stated, quantum fluctuations are bounded below by quantum dissipation, whereas classically the fluctuations can be made to vanish. The lower bound of this inequality is exactly satisfied by (zero-temperature) quantum noise and is in accord with the Heisenberg uncertainty principle, both in its microscopic origins and its influence upon systems. Moreover, it is shown that the non-equilibrium fluctuation-dissipation relation determines the non-equilibrium uncertainty relation of linear systems in the weak-damping limit.

Finally in Sec. 4.7, we discuss our previously mentioned measure of *decoherence strength*. It is known that one can characterize the decoherence strength of a Markovian environments by the product of their temperature and induced damping and order the decoherence strength of multiple environments by this quantity. For non-Markovian environments in the weak coupling regime we show that there exists a natural, albeit partial ordering of decoherence strengths via a perturbative treatment. This measure can be applied to both low-temperature and non-equilibrium environments.

## 4.2 The Accuracy of Perturbative Master Equations

Here we more carefully consider the accuracy of solutions provided by perturbative master equations, as briefly mentioned in Sec. 2.2.2.1. For simplicity we will primarily consider perturbative master equations where the Liouvillian  $\mathcal{L}(t)$  is time

independent at zeroth order and asymptotically constant for late times. We will primarily assume that the perturbative expansion of  $\mathcal{L}(t)$  is in even powers of the coupling, as would arise from a Gaussian influence. The expansion of  $\mathcal{L}(t)$  will then take the form

$$\mathcal{L}(t) = \sum_{n=0}^{\infty} \mathcal{L}_{[2n]}(t), \quad (4.1)$$

$$\mathcal{L}_{[0]}(t) \rho \equiv [-i \mathbf{H}, \rho], \quad (4.2)$$

where  $\mathcal{L}_{[2n]}(t) = \mathcal{O}(g^{2n})$  and to zeroth order the system is driven in a unitary manner by its Hamiltonian  $\mathbf{H}$ .

One might easily assume that solving the second-order master equation defined by the Liouvillian  $\mathcal{L}_{[0]} + \mathcal{L}_{[2]}$  would yield a solution that would match the exact solution to the exact master equation up to second order, having error terms of order  $\mathcal{O}(g^4)$ ; however we will show that in general they will differ by second-order terms, so that one can only say they are in perturbative agreement at zeroth order. One very significant implication of these facts is for positivity. Not being exact, nor generally of Lindblad form, a perturbative master equation is not guaranteed to yield a dynamical map with exact complete positivity. Solutions can and should be completely positive to the relevant perturbative order, and as we show in this work that order is not what one might naively expect. Solutions to the second-order master equation can violate positivity by an amount that is  $\mathcal{O}(g^2)$ . We show that to find solutions good to second-order, canonical perturbation theory generally demands the fourth-order Liouvillian.

### 4.2.1 Indeterminacy of Solutions

Before determining what the appropriate level of accuracy is for the solutions of perturbative master equations, we will first demonstrate that there is issue with the naive expectation of order- $2n$  accuracy. This argument is a generalization of one found in Ref. [118], where the discrepancy was noticed for the second-order equilibrium state. Let  $\rho_{(2n)}(t)$  be any solution which satisfies the master equation (and is supposedly accurate) to order  $2n$ , then

$$\frac{d}{dt}\rho_{(2n)}(t) = \mathcal{L}(t)\rho_{(2n)}(t) + \mathcal{O}(c^{2n+2}). \quad (4.3)$$

Furthermore consider the order- $2n$  state

$$\rho'_{(2n)}(t) \equiv \rho_{(2n)}(t) + \delta\rho_{[2n]}(t), \quad (4.4)$$

where  $\delta\rho_{[2n]}$  is an order- $2n$  traceless and diagonal (in the energy basis) perturbation for which

$$\delta\rho_{[2n]}(0) = 0, \quad (4.5)$$

$$\frac{d}{dt}\delta\rho_{[2n]}(t) = \mathcal{O}(c^{2n+2}), \quad (4.6)$$

so that both  $\rho_{(2n)}(t)$  and  $\rho'_{(2n)}(t)$  share the same initial conditions, and the discrepancy  $\delta\rho_{[2n]}(t)$  grows slowly with the perturbation as to also satisfy

$$\frac{d}{dt}\rho'_{(2n)}(t) = \mathcal{L}(t)\rho'_{(2n)}(t) + \mathcal{O}(c^{2n+2}). \quad (4.7)$$

given that  $\mathcal{L}_0\delta\rho_{[2n]}(t) = 0$  by construction. This demonstrates that, for non-perturbative durations of time, there is an order  $2n$  ambiguity in the stationary (e.g. diagonal) entries of all solutions if one only compares terms up to order  $2n$ .

This proof also applies to time-nonlocal master equations, replacing perturbative contributions to the Liouvillian with corresponding memory-kernel operators. Next we will proceed to our main proofs where we show how this issue arises, that this is the full extent of the problem, and precisely how it can be remedied.

## 4.2.2 Late-time accuracy

It is clear that if Eq. (4.1) is well defined then for sufficiently short times an order- $2n$  master equation (in which the sum in Eq. (4.1) only includes terms up to order  $2n$ ) can produce a solution that is also accurate to order  $2n$ . We find that for longer spans of time, and in particular the late-time regime wherein the master equation assumes its stationary limit, solutions to the order- $2n$  master equation are only accurate to order  $2n-2$ . The reason is an ultimately mundane but slightly subtle result of degenerate perturbation theory. In this section we will address the late-time stationary dynamics, and then in following sections we will address the full-time dynamics, including the crossover from consistent accuracy to loss of accuracy.

Assuming we have the perturbative expansion of a stationary master equation (i.e., an expansion of  $\mathcal{L}$ ), we then seek perturbative solutions obtained by applying canonical perturbation theory of the eigenvalue problem (see Sec. 2.2.2.1)

$$\mathcal{L} \mathbf{o} = f \mathbf{o}, \tag{4.8}$$

where  $\mathbf{o}$  is a Hilbert-space eigen-operator and  $f$  is its corresponding eigen-value. We

already know the zeroth-order solutions

$$\mathcal{L}_{[0]} |\omega_i\rangle\langle\omega_j| = -\imath\omega_{ij} |\omega_i\rangle\langle\omega_j| , \quad (4.9)$$

where  $\mathbf{H}|\omega\rangle = \omega|\omega\rangle$  and  $\omega_{ij} = \omega_i - \omega_j$  denote the (free) energy basis of the system. In the appropriate regime of validity, exact solutions to the perturbative master equation should agree with the perturbative solutions to the exact master equation up to the appropriate order. Note that perturbation theory with master equations is always degenerate perturbation theory as  $\omega_{ii} = \omega_{jj} = 0$ . This inevitably-degenerate subspace corresponds to the space of operators that are diagonal in the energy basis of the free system. For simplicity let us assume no other degeneracy in the spectrum of the free Liouvillian (though the possibility of extra degeneracy or near degeneracy arising from resonance can be suitably dealt with).

Perturbation theory tells us that the second-order corrections to all eigenvalues and eigenoperators of  $\mathcal{L}$  outside the degenerate subspace (off-diagonal operators) can be computed using only the second-order master equation (4.81)-(4.82):

$$f_{ij}^{[2]} = \langle\omega_i| \mathcal{L}_{[2]} \{|\omega_i\rangle\langle\omega_j|\} |\omega_j\rangle , \quad (4.10)$$

$$\langle\omega_{i'}| \mathbf{o}_{ij}^{[2]} |\omega_{j'}\rangle = \frac{\langle\omega_{i'}| \mathcal{L}_{[2]} \{|\omega_i\rangle\langle\omega_j|\} |\omega_{j'}\rangle}{-\imath(\omega_{ij} - \omega_{i'j'})} . \quad (4.11)$$

As is usual in degenerate perturbation theory, to compute corrections to eigenoperators from the degenerate subspace, which all satisfy  $\mathcal{L}_{[0]} \mathbf{o}^{[0]} = \mathbf{0}$ , we must diagonalize  $\mathcal{L}$  in the degenerate subspace. This is equivalent to finding the correct linear combination of eigen-operators which branch under perturbation. The

associated characteristic equation can be written

$$\mathbf{W} \vec{\mathbf{o}} = f \vec{\mathbf{o}}, \quad (4.12)$$

$$[[\vec{\mathbf{o}}]]_i \equiv \langle \omega_i | \mathbf{o} | \omega_i \rangle, \quad (4.13)$$

where  $\vec{\mathbf{o}}$  denotes the degenerate-subspace projection of  $\mathbf{o}$  represented as a vector, i.e. diagonal entries of the eigen-operator while in the free energy basis, and  $\mathbf{W}$  defines the Pauli master equation

$$[[\mathbf{W}]]_{ij} = \langle \omega_i | \mathcal{L} \{ |\omega_j\rangle\langle\omega_j| \} | \omega_i \rangle, \quad (4.14)$$

which is the degenerate-subspace projection of  $\mathcal{L}$  represented as a matrix, i.e. master-equation super-operators which map diagonal entries to diagonal entries. Therefore Eq. (4.12) must be solved for with  $\mathbf{W}_{[2]}$  exactly, and then the further effects of  $\mathbf{W}_{[4]}$ ,  $\mathbf{W}_{[6]}$ , etc., can be incorporated via canonical perturbation theory. [Note that this is slightly more complicated than the usual canonical perturbation in the Schrödinger equation where one knows the Hamiltonian perturbation exactly.] The eigenvalues obtained in diagonalizing  $\mathbf{W}_{[2]}$  give the second-order corrections  $f^{[2]}$  to the eigenvalues of  $\mathcal{L}$  and the correct zeroth-order eigenoperators  $\mathbf{o}^{[0]}$  for the degenerate subspace. Degenerate perturbation theory tells us that, in order to calculate each  $\vec{\mathbf{o}}_i^{[2]}$  for the degenerate subspace, one actually requires  $\mathbf{W}_{[4]}$  from the fourth-order master equation; it will contribute the second-order correction

$$\sum_{j \neq i} \frac{\left( \vec{\mathbf{o}}_j^{[0]} \right)^* \mathbf{W}_{[4]} \left( \vec{\mathbf{o}}_i^{[0]} \right)}{f_i^{[2]} - f_j^{[2]}} \vec{\mathbf{o}}_j^{[0]}, \quad (4.15)$$

where  $\vec{\mathbf{o}}_i^*$  is the left eigen-vector of  $\mathbf{W}$  such that  $\vec{\mathbf{o}}_i^* \mathbf{W} = \vec{\mathbf{o}}_i^* f_i$  and  $\vec{\mathbf{o}}_j^* \vec{\mathbf{o}}_i = \delta_{ij}$ . Such corrections would be fourth order in a non-degenerate problem, but because

the free Liouvillian is always degenerate, they become second order as the relevant lowest-order nonvanishing eigenvalue splitting is always second order here. Without this information from the fourth-order master equation, one cannot generate the complete second-order solution.

Finally note that this requirement must extend even to exact solutions of the perturbative master equation. A perturbative solution to the second-order master equation will be equivalent to solving the full master equation perturbatively and then artificially setting  $\mathcal{L}_{[4]}$  and all higher-order contributions to the Liouvillian to vanish. From this and the preceding perturbative analysis we know that the second-order perturbative solutions to the exact and second-order master equations must differ by a term that is  $\mathcal{O}(g^2)$ . Since the exact solutions to each given master equation must differ from the corresponding second-order perturbative solutions by terms of  $\mathcal{O}(g^4)$ , we can conclude from our analysis that even the exact solution to the second-order master equation differs from the exact solution to the full master equation by a term of  $\mathcal{O}(g^2)$ . In the final section we use the example of quantum Brownian motion, where an exact solution is available, to show that the second-order corrections arising from the fourth-order Liouvillian are indeed present.

More generally, the same argument tells us that while the short-time accuracy of an order- $2n$  master equation can also be order  $2n$ , the long-time accuracy can only be order  $2n-2$ . To obtain order- $2n$  solutions one requires not only the order- $2n$  master equation but in addition the order- $(2n+2)$  Pauli master equation. In particular, the second-order master equation after taking the *rotating-wave approximation* [22] will contain just enough terms to generate solutions which are accurate

to zeroth-order (see Sec. 4.4). The full second-order master equation improves upon this but not enough to generate the full second-order solutions.

Among the information missing due to the second-order errors of the solution to the second-order master equation are important contributions to the asymptotic state of the system. When coupled to a thermal reservoir the system must asymptote to  $\rho \propto e^{-\beta \mathbf{H}}$  for vanishing system-environment coupling (though this may also happen in other, very specific approximations [73]). One often desires to find the additional environmentally induced system-system correlations (and possibly entanglement) provided by perturbative corrections, but these will not be given correctly by directly finding the steady state of the second-order master equation. However, at least for zero-temperature noise, it is still possible to easily construct via other methods the order- $2n$  corrections using only order- $2n$  master equation coefficients and limits thereof (see Sec. 4.5).

Another important characteristic that is mangled by the second-order master equation is positivity, as was mentioned in the introduction. The second-order inaccuracies that arise from using the second-order master equation imply that the diagonal elements of the density matrix in the (free) energy basis are off by second-order terms. This can lead to second-order violations of positivity. In fact, this is almost guaranteed at low temperature, where any off-diagonal perturbation to the ground state will immediately cause second-order positivity violation, given that the necessary inequality

$$\rho_{ii} \rho_{jj} \geq \rho_{ij} \rho_{ji}, \quad (4.16)$$

cannot be satisfied with the left-hand side vanishing at zeroth-order and not perturbed to the correct second-order values.

### 4.2.3 Full-time accuracy

In analyzing the full-time accuracy of time-dependent master equations, first we will show that the short-time solutions are accurate to the order of the master equation, and then we will show that longer-time solutions display accuracy loss. The timescale for this transition is determined by the frequency perturbations, e.g.  $1/f_{[2]}$ .

To analyze the short-time behavior we rotate to the interaction picture defined

$$\underline{\rho}(t) \equiv \mathcal{G}_0^{-1}(t) \rho(t), \quad (4.17)$$

$$\mathcal{G}_0(t) \rho \equiv e^{-i\mathbf{H}t} \rho e^{+i\mathbf{H}t}, \quad (4.18)$$

wherein the master equation is now given by

$$\frac{d}{dt} \underline{\rho}(t) = \underline{\delta\mathcal{L}}(t) \underline{\rho}(t), \quad (4.19)$$

$$\underline{\delta\mathcal{L}}(t) \equiv \mathcal{G}_0^{-1}(t) \delta\mathcal{L}(t) \mathcal{G}_0(t), \quad (4.20)$$

$$\delta\mathcal{L}(t) \equiv \mathcal{L}(t) - \mathcal{L}_0, \quad (4.21)$$

and so the interaction-picture dynamics are strictly perturbative. Short-time solutions can be obtained from the Neumann series

$$\underline{\rho}(t) = \underline{\mathcal{G}}(t) \rho(0), \quad (4.22)$$

$$\underline{\mathcal{G}}(t) = \mathbf{1} + \int_0^t d\tau \underline{\delta\mathcal{L}}(\tau) + \int_0^t d\tau \int_0^\tau d\tau' \underline{\delta\mathcal{L}}(\tau) \underline{\delta\mathcal{L}}(\tau') + \dots, \quad (4.23)$$

where the order- $2n$  solution is fully determined by  $\mathcal{L}_{[2n]}(t)$ . However, such solutions are inherently secular in time. If  $f_{[2]}$  denotes the second-order frequency perturbations, e.g. dissipation and diffusion rates, then the above solutions (at second order) are only good for times  $t \ll 1/f_{[2]}$ . This is the regime wherein perturbative master equations are ensured to provide matching accuracy in their solutions.

For longer spans of time, one must resort to time-ordered integration for solutions. For weak coupling the master equation can asymptote to its stationary value within timescales much shorter than  $1/f_{[2]}$ , and so one can apply the stationary master equation and our corresponding proof of accuracy loss. More generally one may consider the behavior of the time-dependent eigen-value equation

$$\mathcal{L}(t) \mathbf{o}(t) = f(t) \mathbf{o}(t), \quad (4.24)$$

so that the time-translation generator may be given by its spectral decomposition

$$\mathcal{L}(t) = \sum_k f_k(t) \mathbf{o}_k(t) \mathbf{o}_k^*(t). \quad (4.25)$$

Again, the order- $2n$  master equation can only determine the perturbatively-stationary eigen-operators  $\mathbf{o}(t)$  to within order  $2n-2$ . Given that the time-dependent basis of the time-translation generator cannot be determined to second order, neither can the solutions.

One might be concerned with how the proof of short-time accuracy is compatible with this proof of full-time accuracy loss. In fact, the short-accuracy occurs within a span of time  $0 < t \ll 1/f_{[2]}$ , which is not sufficient enough to accumulate full-order contributions from the perturbation. Therefore the regime of short-time accuracy is a rather trivial result.

#### 4.2.4 Time Non-Local Accuracy

Corresponding to the time-local master equation is the time-nonlocal master equation

$$\frac{d}{dt}\boldsymbol{\rho}(t) = \int_0^t d\tau \boldsymbol{\mathcal{K}}(t-\tau) \boldsymbol{\rho}(\tau), \quad (4.26)$$

as considered in Sec. 2.3. The nonlocal kernel  $\boldsymbol{\mathcal{K}}(t)$  also has a perturbative expansion with zeroth-order dynamics given by

$$\boldsymbol{\mathcal{K}}_{[0]}(t) = 2\delta(t) \boldsymbol{\mathcal{L}}_{[0]}. \quad (4.27)$$

which is time-local and unitary. Solutions are most easily calculated in the Laplace domain wherein one has the kernel

$$\hat{\boldsymbol{\mathcal{K}}}(s) = \int_0^\infty dt e^{-ts} \boldsymbol{\mathcal{K}}(t), \quad (4.28)$$

$$\hat{\boldsymbol{\mathcal{K}}}_{[0]}(s) = \boldsymbol{\mathcal{L}}_{[0]}. \quad (4.29)$$

As was mentioned in Sec. 2.3, perturbative solutions can then be acquired by solving the nonlocal eigen-value equation [63]

$$\hat{\boldsymbol{\mathcal{K}}}(s) \hat{\boldsymbol{o}}(s) = \hat{k}(s) \hat{\boldsymbol{o}}(s), \quad (4.30)$$

where from Eq. (4.29) the nonlocal eigen-system must be a perturbation of the free system-energy eigen-system, and therefore our proof of accuracy loss will carry over. The order- $2n$  master equation can only determine the perturbatively-stationary eigen-operators  $\hat{\boldsymbol{o}}(s)$  to within order  $2n-2$ .

### 4.2.5 Example: QBM

As an example of an exactly-solvable open system, let us consider the master equation of an oscillator bilinearly coupled (position-position) to an environment of oscillators initially in a thermal state [84]:

$$\frac{d}{dt}\boldsymbol{\rho} = [-\imath \mathbf{H}_L, \boldsymbol{\rho}] - \imath \Gamma [\mathbf{X}, \{\mathbf{P}, \boldsymbol{\rho}\}] - MD_{PP} [\mathbf{X}, [\mathbf{X}, \boldsymbol{\rho}]] - D_{XP} [\mathbf{X}, [\mathbf{P}, \boldsymbol{\rho}]] , \quad (4.31)$$

where  $\mathbf{H}_L$  is the system Hamiltonian but with frequency  $\Omega_L$ ,  $\Gamma$  is the dissipation coefficient,  $D_{PP}$  and  $D_{XP}$  are the regular and anomalous diffusion coefficients. This master equation describes the dynamics of damped nano-mechanical resonators at low temperature, among other physical systems.

In Chapter 3 we have provided exact solutions with full time dependence, and it is from there that we take the following results. Let us consider Ohmic coupling to the bath with damping kernel  $\gamma(t) = 2\gamma_0\delta_\Lambda(t)$ , where  $\delta_\Lambda(t)$  is a representation of the delta function in the high-frequency cutoff limit  $\Lambda \rightarrow \infty$ . [The damping kernel, and thus  $\gamma_0$ , is second order in the system-environment interaction  $g$ .] The homogeneous coefficients quickly asymptote to  $\Omega_L = \Omega$  and  $\Gamma = \gamma_0$  within the cutoff timescale, whereas the diffusion coefficients asymptote to

$$D_{xp} = +\gamma_0 \text{Im}[\mathcal{I}_0] , \quad (4.32)$$

$$D_{PP} = 2\gamma_0 T + \gamma_0 \text{Im} \left[ \left( \gamma_0 + \imath \tilde{\Omega} \right) \mathcal{I}_0 \right] , \quad (4.33)$$

$$\mathcal{I}_0 \equiv \frac{2}{\pi} \left( \imath + \frac{\gamma_0}{\tilde{\Omega}} \right) \left\{ \text{H} \left( \frac{\Lambda}{2\pi T} \right) - \text{H} \left( \frac{\gamma_0 + \imath \tilde{\Omega}}{2\pi T} \right) \right\} , \quad (4.34)$$

$$\tilde{\Omega} \equiv \sqrt{\Omega^2 - \gamma_0^2} , \quad (4.35)$$

mostly within the system timescale, but also hastened by temperature. In all coef-

ficients we have neglected terms of order  $\mathcal{O}(1/\Lambda)$ . Here  $H$  is the harmonic number function, which is asymptotically logarithmic and yet  $H(0) = 0$ . Therefore both diffusion coefficients contain logarithmic cutoff sensitivities, though the sensitivity is present in the anomalous diffusion coefficient at second order, whereas it does not appear in the regular diffusion coefficient until fourth order.

In the stationary limit, the system relaxes into a Gaussian state with phase-space covariance

$$\boldsymbol{\sigma}_T = \begin{bmatrix} \frac{1}{M\Omega_R^2} \left( \frac{1}{2\Gamma} D_{PP} - D_{XP} \right) & 0 \\ 0 & \frac{M}{2\Gamma} D_{PP} \end{bmatrix}. \quad (4.36)$$

One can see that for a second-order master equation, the contribution from the regular diffusion  $D_{PP}/\Gamma$  starts at zeroth order, while the contribution from anomalous diffusion  $D_{XP}$  starts at second order. The full second-order contribution from the regular diffusion actually requires the fourth-order coefficients.

In the exact calculation, or in any consistent perturbative calculation, the logarithmic cutoff sensitivities present in the diffusion coefficients actually cancel in the position uncertainty. In this sense the anomalous diffusion coefficient acts as an anti-diffusion coefficient and this behavior will also occur for supra-Ohmic couplings. If one were to naively apply the second-order diffusion coefficients, and solve the master equation exactly, then one would obtain a mixed-order result and the logarithmic cutoff sensitivities would not precisely cancel. The position uncertainty would contain a second-order *negative*  $\log(\Lambda)$  contribution. For sufficiently large cutoff frequencies, the Heisenberg uncertainty principle would be violated. For even larger frequencies, the covariance would become negative. In any case the

second-order master equation would produce a (supposedly) second-order position uncertainty which is an underestimation of the true second-order uncertainty.

### 4.3 Dynamically Generated System-Environment Correlations

The most common choice global initial conditions is to assume factorized initial states for the system and environment<sup>1</sup>

$$\rho_C(0) = \rho_S(0) \otimes \rho_E(0), \quad (4.37)$$

$$\rho_E(0) = \frac{1}{Z_E(\beta)} e^{-\beta \mathbf{H}_E}. \quad (4.38)$$

where  $Z_E(\beta)$  denotes the partition function of the free (noninteracting) environment and  $T = 1/\beta$  is the temperature of the environment, which acts here as a thermal reservoir.

When considering environments with a large number of high-frequency modes and characterized by a UV frequency cutoff  $\Lambda$ , such a factorized initial state (chosen for mathematical simplicity) unfortunately engenders unphysical behavior such as a sudden jolt in physical quantities near the initial time (this was analyzed in some detail in Ref. [84]) or spurious cutoff sensitivity of certain system correlators (see Ref. [86] and references therein). This kind of initial conditions assumes that an uncorrelated system and environment are instantaneously coupled with non-vanishing strength. The pathological behavior arises because the factorized initial state con-

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<sup>1</sup>Even after resolving issues of renormalization, the system will typically be displaced by a finite amount of the order of the induced damping within a very short time of the order of the inverse UV cutoff of the environment.

tains a number of highly excited energy states of the full Hamiltonian (including the interacting Hamiltonian), even when the initial reduced states of the system and environment are not highly excited in the free theory, and it is a reflection of the high-frequency modes of the environment quickly becoming correlated with the system within a time of order  $1/\Lambda$ .

The next most common choice of initial state (see Ref. [77]) has been to consider system deformations or measurements of the global equilibrium state of the combined system ‘C’, with density matrix

$$\rho_{\text{C}}(0) = \sum_n \mathbf{O}'_n \frac{1}{Z_{\text{C}}(\beta)} e^{-\beta \mathbf{H}_{\text{C}}} \mathbf{O}_n, \quad (4.39)$$

where the  $\mathbf{O}$  and  $\mathbf{O}'$  operators are restricted to act on the system. However, this method still gives rise to jolts for sufficiently general deformations or measurements [134], which can be understood as a consequence of altering the state of the system instantaneously [12].

To cure or tame these drastic effects, especially in the context of linear systems, the following procedure has been suggested: a) force the system by a constant amount, b) wait for it to relax into the displaced equilibrium state, and then c) release the force [77]. Alternatively and in order to generate interesting coherent superposition states for the system, one can start with the equilibrium state of the combined system and act on the system, but for a non-vanishing time [12]. Essentially we view the problem as an imbalance between initial correlations and initial coupling strength; the imbalance can be countered on either side. We also believe that the most natural resolution should be a dynamical preparation which

relies upon equilibration [77, 21] followed by an additional preparation of the system for a finite time [12]. Our key contribution is showing that this can be achieved while still taking advantage of the simpler analytical results obtained when deriving the master equation for a factorized initial state, without the need to introduce inhomogeneous terms and an affine master equation [21].

From this perspective of the second-order master equation (see Chapter 2), the mathematical cause of the initial jolts becomes clear. For constant Hamiltonians and an initially stationary environment, the second-order operator (which determines the master equation) obeys the relation

$$\frac{d}{dt}(\mathbf{A}_{nm} \diamond \mathbf{L}_m)(t) = \alpha_{nm}(t) \{ \mathcal{G}_0(t) \mathbf{L}_m \}, \quad (4.40)$$

which can be extremely large near the initial time when considering an environment with a sufficient amount of high frequency modes (such as low-temperature ohmic and supra-ohmic environments) since  $\alpha(t)$  is typically a very localized distribution in those cases. For a finite but large cutoff  $\Lambda$ , the correlation function becomes of order  $\Lambda$  for a time of order  $1/\Lambda$ .

The traditional (mathematical) approach to handling factorized initial states has been to consider the initial state to be almost factorized, but with some small amount of correlation consistent with the interaction strength. The projective operator methods essentially consider representing the initial state as

$$\rho_C(0) = \rho_S(0) \otimes \rho_E(0) + \delta\rho_C(0), \quad (4.41)$$

with  $\delta\rho_C(0)$  perturbative in the interaction. Following through with a very analogous

derivation to Sec. 2.2.1, the master equation appears to change from linear to affine.

$$\frac{d}{dt}\underline{\rho}(t) = \underline{\mathcal{L}}(t)\underline{\rho}(t) + \underline{\mathcal{J}}(t), \quad (4.42)$$

$$\underline{\mathcal{J}}(t) \equiv \langle \underline{\mathcal{L}}_I(t) \underline{\delta\rho}_C(t) \rangle_E. \quad (4.43)$$

This is the (time-localized) Nakajima-Zwanzig generalized master equation. The linear dependence upon the reduced density matrix remains unchanged, but the traceless correlation matrix  $\underline{\delta\rho}_C$  introduces a probability conserving current  $\underline{\mathcal{J}}(t)$  which is dependent upon the initial system-environment correlations.  $\underline{\mathcal{J}}(t)$  is often referred to as an inhomogeneity and calculated perturbatively via Neumann series for  $\underline{\mathcal{G}}_C(t)$ , as we did to obtain  $\underline{\mathcal{L}}(t)$ . Eq. (4.42) is then presented as the “master equation” for initially entangled states.

The formal solution to the generalized master equation can be directly related to the homogeneous solutions for factorized initial states:

$$\rho(t) = \mathcal{G}(t)\rho(0) + \int_0^t d\tau \mathcal{G}(t, \tau) \mathcal{J}(\tau). \quad (4.44)$$

One can see that the contribution from initial correlations is always higher order in the interaction. Moreover, if the initially factorized system would relax into some asymptotic state, the current contribution should vanish in the late-time limit, regardless of coupling strength. At least in the weak-coupling regime, the current contribution is most relevant near the initial time when, given proper correlations, it can compensate for jolts that would occur in the homogeneous coefficients due to factorized initial conditions [21]. With exception to the jolting, the dissipative corrections to the master equation would relax much more quickly than the rate at

which their effects would be felt by the system. So at least in the weak-coupling limit, the asymptotic master equation coefficients can well approximate the full-time coefficients when the jolting has been properly dealt with.

There is some mathematical displeasure with this combined perturbative and affine approach. While the reduced density matrix has the correct linear dependence upon  $\delta\rho_C$ , the induced current  $\mathcal{J}(t)$  is left evolving in a secular manner from its perturbative expansion. This was not sufficient for the reduced density matrix and, strictly speaking, it is not entirely sufficient here. The current requires a dynamical equation or exact expression, otherwise the “master equation” is likely only suitable for short times, though well chosen correlations can still allow convergence. Therefore, with the added computational difficulty and potential for secularity in mind, it is an added luxury that we are able to avoid the affine formalism when generating proper correlations.

### 4.3.1 Coupling Switch-On

One method for balancing the initial coupling between the system and environment with their initial lack of correlation, is to turn on the coupling slowly with a time-dependent interaction such as

$$\mathbf{H}_I = \theta_s(t) \sum_n \mathbf{L}_n \otimes \mathbf{I}_n, \quad (4.45)$$

where  $\theta_s(t) : [0, \infty) \rightarrow [0, 1)$  is a smooth switch-on function with a characteristic timescale  $\tau_s$ , which vanishes at the initial time and becomes (effectively) one for times longer than  $\tau_s$ . To some extent, we considered this for linear systems in

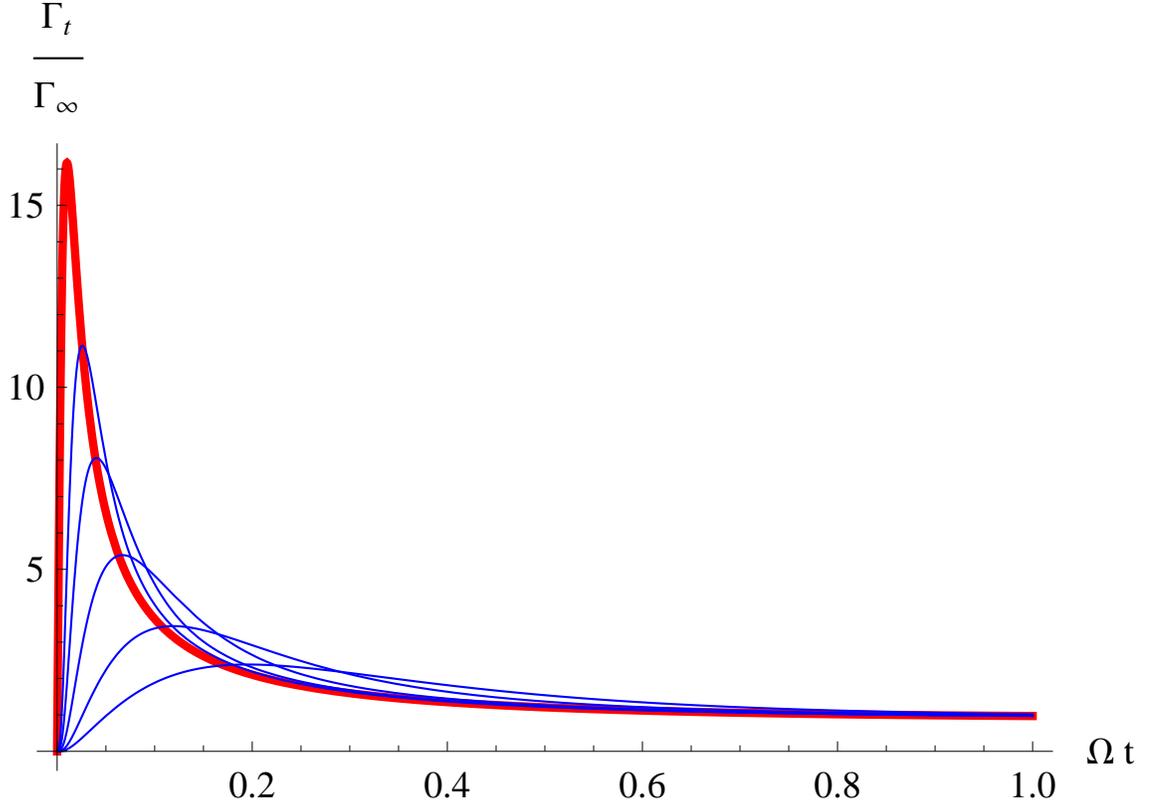


Figure 4.1: Zero-temperature, ohmic decay rate for the **• instantaneously coupled** and **· gradually coupled** initial states of a two-level system with exponential cutoff frequency  $\Lambda = 100\Omega$ . In this case the switch-on function is exponential,  $\theta_s(t) = 1 - e^{-t/\tau_s}$ , and the switch-on times  $\tau_s$  are chosen to take the values  $1/\Lambda, 2/\Lambda, 4/\Lambda, 8/\Lambda, 16/\Lambda$ .

Ref. [64].

Such a time-dependent interaction is equivalent to employing the second-order operator

$$(\mathbf{A}_{nm} \diamond \mathbf{L}_m)(t) = \theta_s(t) \int_0^t d\tau \theta_s(t-\tau) \alpha_{nm}(\tau) \{\mathcal{G}_0(\tau) \mathbf{L}_m\}, \quad (4.46)$$

for otherwise constant couplings and Hamiltonians. Therefore, any initial jolt due to the localized nature of  $\alpha(t)$  will be suppressed by  $\theta_s(t)$  as long as  $\tau_s \gg 1/\Lambda$ . As can be seen in Fig. 4.1, the cutoff-frequency jolts are essentially replaced by jolts of

frequency  $\min[\Lambda, 1/\tau_s]$  and amplitude proportional to the same value. This approach provides a useful way of generating initial system-environment correlations when  $\tau_s$  is much larger than  $1/\Lambda$  but smaller than any other relevant timescales (such as the system frequencies). Furthermore, even if a mild jolt is still present, the important point is that it is cutoff insensitive (for fixed  $\tau_s$  and sufficiently large  $\Lambda$ ).

### 4.3.2 Dynamically Prepared Initial States

Alternatively, in order to balance the initial correlations with an initially non-vanishing interaction strength, we will consider here initial states with suitable correlations to the environment. Such states will be obtained via an auxiliary construction which involves evolving an initially uncorrelated state for a sufficiently long time (a similar procedure was used in Ref. [127, 130] within the context of semiclassical gravity). The system-environment correlations are then dynamically generated through the environmental interaction itself. Our first examples of *equilibrium preparation* will be the simplest mathematically, while the final examples of *non-equilibrium preparation* will be closer to actual laboratory experiments.

In all cases we will take the system and environment to be uncorrelated not at  $t = 0$  but in the infinite past.

$$\rho_{\text{C}}(-\infty) = \rho_{\text{S}}(-\infty) \otimes \rho_{\text{E}}(-\infty), \quad (4.47)$$

for some (possibly unimportant) system state  $\rho_{\text{S}}(-\infty)$  and thermal  $\rho_{\text{E}}(-\infty)$ . We

then define the system Hamiltonian piecewise in time

$$\mathbf{H}_S(t) = \begin{cases} \mathbf{H}_+(t) & 0 < t \\ \mathbf{H}_- & t < 0 \end{cases}, \quad (4.48)$$

such that in past the system is allowed to equilibrate with the environment for an infinite time, which determines the correlated initial state at  $t = 0$ . The second-order master equation is then determined by

$$(\mathbf{A}_{nm} \diamond \mathbf{L}_m)(t) = \int_{-\infty}^t d\tau \alpha_{nm}(t, \tau) \{ \mathcal{G}_S(t, \tau) \mathbf{L}_m(\tau) \}. \quad (4.49)$$

To analyze the coefficients associated with the initially-correlated state, we will reduce them to a sum of coefficients for the auxiliary initially-uncorrelated state involving various time ranges. First, we split the integration into two parts

$$\int_{-\infty}^t d\tau = \int_0^t d\tau + \int_{-\infty}^0 d\tau, \quad (4.50)$$

with the first integral depending only upon  $\mathcal{G}_+(t, \tau)$  and corresponding to the uncorrelated coefficients. Inserting the product  $\mathcal{G}_-(0, t) \mathcal{G}_-(t, 0)$ , which equals the identity, the second integral can be written as

$$\mathcal{M}(t) \int_{-\infty}^0 d\tau \alpha_{nm}(t, \tau) \{ \mathcal{G}_-(t, \tau) \mathbf{L}_m(\tau) \}, \quad (4.51)$$

given the operator

$$\mathcal{M}(t) \equiv \mathcal{G}_+(t, 0) \mathcal{G}_-(0, t). \quad (4.52)$$

The integral in Eq. (4.51) is then broken up into two parts

$$\int_{-\infty}^0 d\tau = \int_{-\infty}^t d\tau - \int_0^t d\tau, \quad (4.53)$$

corresponding to the asymptotic and finite-time coefficients for an initially uncorrelated system driven by the time-independent preparation Hamiltonian  $\mathbf{H}_-$ . Finally, our correlated coefficients can be expressed in terms of the uncorrelated coefficients as

$$\underbrace{(\mathbf{A}_{nm} \diamond \mathbf{L}_m)(t)}_{\text{correlated}} = \underbrace{(\mathbf{A}_{nm} \diamond \mathbf{L}_m)_+(t)}_{\text{uncorrelated}} - \mathcal{M}(t) \left\{ \underbrace{(\mathbf{A}_{nm} \diamond \mathbf{L}_m)_-(t)}_{\text{jolt suppression}} - \underbrace{(\mathbf{A}_{nm} \diamond \mathbf{L}_m)_-(\infty)}_{\text{preparation eraser}} \right\}, \quad (4.54)$$

where the subscripted  $(\mathbf{A} \diamond \mathbf{L})_{\pm}$  coefficients are defined as

$$(\mathbf{A}_{nm} \diamond \mathbf{L}_m)_{\pm}(t) \equiv \int_0^t d\tau \alpha_{nm}(t, \tau) \{ \mathcal{G}_{\pm}(t, \tau) \mathbf{L}_m(\tau) \}. \quad (4.55)$$

If the system frequencies are always small as compared to the cutoff, we can inspect the early-time behavior (and jolts) by letting  $\mathcal{G}_{\pm}(t) \approx \mathbf{1}$ . Then one can see that the first two terms of Eq. (4.54) will precisely cancel in the early-time regime. Therefore, the correlated initial states are jolt-free given sufficiently small system frequencies as compared to the cutoff:  $\Omega \ll \Lambda$ . The final term in turn is such that in the late-time limit it precisely cancels the second term and erases all memory of  $\mathbf{H}_-$ . Finally note that, quite trivially, if we choose  $\mathbf{H}_+(t) = \mathbf{H}_-$ , then the first two terms cancel and we recover the equilibrium coefficients at any finite time.

#### 4.3.2.1 Equilibrium Preparation

To prepare an initial state in this approach, we choose the past Hamiltonian  $\mathbf{H}_-$  such that its dynamics along with the environment interaction relaxes our system

to the desired initial state:

$$\lim_{t \rightarrow \infty} e^{t\mathcal{L}_-(\infty)} \rho_0 = \rho_0, \quad (4.56)$$

$$\mathcal{L}_-(\infty) \rho_0 = \mathbf{0}, \quad (4.57)$$

where  $\mathcal{L}_-(\infty)$  is the stationary limit of the Liouvillian for a system with the past Hamiltonian as well as the coupling to the environment.

Our target state  $\rho_0$  will only be specified to zeroth order in the system-environment interaction. This is because for sufficiently long times (and in particular for the asymptotic equilibrium state) the diagonal elements of the reduced density matrix in the energy basis cannot be determined beyond zeroth order anyway when using the second-order perturbative master equation (see Sec. 4.2). Due to unavoidable degeneracy present in all open-system dynamics, one actually requires components of the fourth-order master equation to calculate the full second-order solutions. The second-order master equation provides for all second-order dynamical quantities, such as frequency shifts, dissipation, diffusion and decoherence rates. We are concerned here with the induced jolts, which are dynamical quantities, and so this subtle point does not raise any additional problems for us.

For  $\mathbf{L}_n$  all commuting with each other, one can force a general environment into  $\ell$ -state *preparation via decoherence*. If the past Hamiltonian is deactivated, or more generally taken to commute with  $\mathbf{L}_n$ , then since all system operators commute with each other, the master equation and its solutions will trivially result in a system which decoheres in the  $\ell$ -basis associated with the  $\mathbf{L}_n$ . Thus, coefficients prepared in this manner are consistent with any initial state which is a completely incoherent

mixture of  $\ell$ -states. [Note that if  $\rho_S(-\infty)$  corresponds to a pure eigenstate of the set  $\{\mathbf{L}_n\}$ , this procedure simply adjusts the state of the environment, while system and environment remain unentangled.]

A finite-temperature environment allows mixed state *preparation by equilibration*. Essentially one chooses the past Hamiltonian so that its thermal state (or some other steady state) is the desired initial state. For a positive-temperature environment, at zeroth order one can prepare a (sufficiently) mixed state  $\rho_0$  with the past Hamiltonian  $\mathbf{H}_- = -T \log(\rho_0)$ . However, one must be careful that past system frequencies are small as compared to the high frequency jolts, otherwise this preparation will fail to remedy jolting. One can work out that the *adiabatic preparation* regime is given by

$$\frac{p_{\max}}{p_{\min}} \ll e^{\beta \Lambda}, \quad (4.58)$$

where  $\Lambda$  is the jolt frequency and  $p$  are the initial state probabilities of preparation energy levels connected by  $\mathbf{L}_n$ . (Clearly, for this method to work there can only be a finite number of such energy levels.)

To prepare an initially pure state via equilibration at the order that we are working, one requires a zero-temperature environment for *preparation by freezing*. Then one can choose any  $\mathbf{H}_-$  with ground state  $\rho_0$ . It is important to emphasize that the reduced density matrix of the system corresponding to the ground state of the combined system will not be a pure state in general due to the entanglement between the system and the environment: the free ground state of the system is a pure state, but the reduced density matrix of the open system is in general a mixed

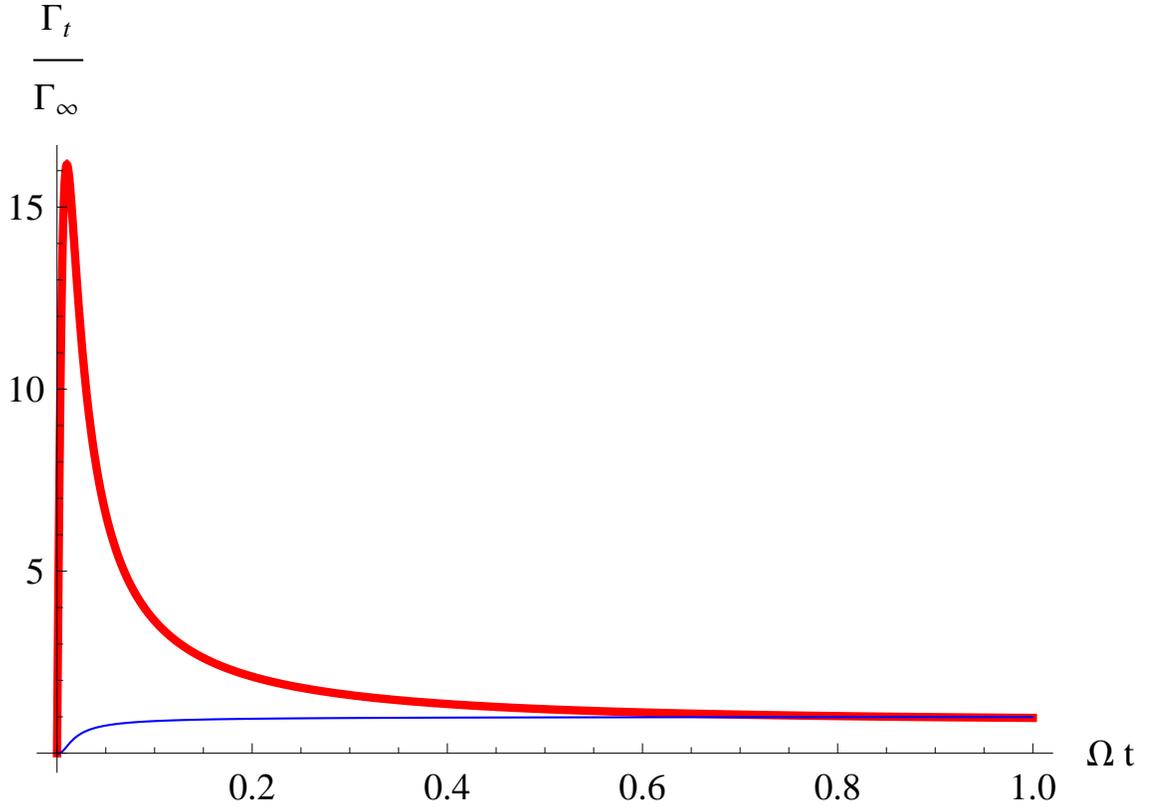


Figure 4.2: Zero-temperature, ohmic decay rate for the • unprepared and • prepared initial states of a two-level system with exponential cutoff frequency  $\Lambda = 100\Omega$ . In this case preparation by freezing was used to create an initially excited state.

state beyond zeroth order in the system-environment coupling. However, this point becomes irrelevant at the order that we are working since, as explained above, when using the second-order perturbative master equation to prepare the initial state by equilibration, one cannot meaningfully specify  $\rho_0$  beyond zeroth order.

### 4.3.2.2 Non-Equilibrium Preparation

In order to consider situations closer to actual laboratory experiments, here we will first allow the system to equilibrate with the environment (as described in the previous subsection) and then choose some preparation Hamiltonian  $\mathbf{H}_P(t)$ , which would (in the absence of coupling to the environment) generate the desired initial state in some finite time  $\tau_P$ . One simply applies the master-equation coefficients in Eq. (4.54) with future Hamiltonian

$$\mathbf{H}_+(t) = \begin{cases} \mathbf{H}_0(t) & \tau_P < t \\ \mathbf{H}_P(t) & t < \tau_P \end{cases}, \quad (4.59)$$

where  $\mathbf{H}_0(t)$  is the desired post-preparation Hamiltonian. All jolts will be avoided if  $1/\tau_P \ll \Lambda$ : the introduction of a non-vanishing preparation frequency serves to tame the jolts and eliminate their high-cutoff sensitivity.

A possible preparation Hamiltonian, which could model as a particular case Rabi oscillations induced by an appropriate laser field acting on a two-level system, is the following *state-flipping* Hamiltonian:

$$\mathbf{H}_P = \frac{\pi}{2\tau_P} (|\psi_0\rangle\langle 0| + |0\rangle\langle\psi_0|). \quad (4.60)$$

Assuming that one has a zero-temperature environment and that the system is already equilibrated, driving the system with this Hamiltonian for a time  $\tau_P$  provides a relatively easy way of preparing an initial pure state  $|\psi_0\rangle$ . As discussed above, the reduced density matrix of the system will actually be a mixed state in general, because of the system-environment entanglement of the equilibrium state as well as the interaction to the environment while evolving the combined system during this

additional finite preparation time. In fact, the preparation time  $\tau_{\text{P}}$  cannot be too long if we want the state of the system to be more or less close to  $|\psi_0\rangle$ .

Now let us consider a system which is initially equilibrated, without making any assumption as to the temperature of the environment. We couple the system to an ancillary and analog system (equivalent Hilbert spaces) that is already prepared in the desired initial state. The system of interest and ancilla are temporarily coupled in such a way that they swap states, for instance by means of the following block-matrix *state-swapping* Hamiltonian:

$$\mathbf{H}_{\text{P}} = \frac{\pi}{2\tau_{\text{P}}} \begin{bmatrix} \mathbf{0} & \mathbf{1} \\ \mathbf{1} & \mathbf{0} \end{bmatrix}. \quad (4.61)$$

In the absence of coupling to the environment this would exactly swap the system and ancilla states in a time  $\tau_{\text{P}}$ . The same remarks as for state flipping concerning the purity and accuracy of the prepared state when taking into account the coupling to the environment also apply in this case.

### 4.3.2.3 Other possibilities

Within the second-order perturbative approach, generation of equilibrium correlations in a laboratory setting can always be calculated using Eq. (4.54). One only needs to make sure that any additional state preparation does not rely upon large system energies as compared to the bath cutoff. For instance, one can consider the preparation of Ref. [12], which relies on ancillary degrees of freedom to drive the equilibrium state into a coherent superposition. In fact, one could simply apply their own time-dependent Hamiltonian to our formulas as  $\mathbf{H}_+(t)$  and obtain results

consistent with theirs.

## 4.4 The Rotating-Wave Approximation

The rotating-wave approximation (RWA) is used in many places in the study of open quantum systems, particularly in the field of quantum optics (see for example [22, 150, 32]), but the validity of the approximation is treated in depth far less often. There are actually two distinct rotating-wave approximations both in widespread use : 1) the “pre-trace” RWA, which consists of modifying the interaction Hamiltonian by dropping the so-called counter-rotating terms that are quickly oscillating in the Dirac picture; and 2) the “post-trace” RWA, which is obtained by neglecting terms in the master equation for the reduced density matrix that are quickly oscillating in the Dirac picture (see, e.g., [150, 32] and [22, 5] respectively). Agarwal has carried out a systematic study [5, 3, 4] differentiating between these two RWAs and addressed their validity for atom-field interactions and spontaneous emission processes. More recently, various authors have claimed some features of the RWA that may limit its applicability, which we will now discuss.

The most widely acknowledged problem with the pre-trace RWA seems to be that it yields incorrect frequency shifts in the atomic energy levels, so that it is not suitable for calculating environmentally induced level shifts or induced cooperative frequency shifts [5, 3, 4]. West and Lindenberg [152] found that the reduced system dynamics obtained from the pre-trace RWA do not have a Markovian limit <sup>2</sup>. Finally,

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<sup>2</sup>These authors also claim that there is no fluctuation-dissipation theorem for this model, a statement that the present authors cannot agree with.

Ford and O’Connell [68] have raised concerns that in general the total Hamiltonian obtained by the pre-trace RWA does not have a spectrum which is bounded below, and they suggest that this limits the applicability of the approximation to first-order transition amplitudes. In our view, this is more a matter of the renormalization being less trivial for the pre-trace RWA Hamiltonian.

Other authors have raised a very different sort of concern about the pre-trace RWA Hamiltonian for coupling of a localized system to a quantized field, that it may produce spurious causality violation in the calculations. Consider, for example, a two-level atom in the dipole approximation interacting with the electromagnetic field. The multipolar form of the Hamiltonian for this interaction is

$$\mathbf{H}_I = \mathbf{d} \cdot \mathbf{D}(\mathbf{X}), \quad (4.62)$$

$$\underline{\mathbf{H}}_I(t) = \sum_{\mathbf{k}, \epsilon} i\tilde{g}\sqrt{k} (\mathbf{d} \cdot \boldsymbol{\epsilon}) (e^{+i\Omega t} \boldsymbol{\sigma}_+ + e^{-i\Omega t} \boldsymbol{\sigma}_-) \left( e^{+i(\mathbf{k} \cdot \mathbf{X} - \omega_k t)} \mathbf{a}_{\mathbf{k}, s} - e^{-i(\mathbf{k} \cdot \mathbf{X} - \omega_k t)} \mathbf{a}_{\mathbf{k}, s}^\dagger \right), \quad (4.63)$$

in the Dirac picture, and in the pre-trace RWA the terms with frequency  $\Omega + \omega_k$  would be neglected. However, with these terms dropped the interaction can no longer be expressed in terms of the local field variable  $\mathbf{D}(\mathbf{X})$  [37]. Indeed, a numerical study of a three-atom problem [52] found that noncausal terms appear when the pre-trace RWA is used, unless one makes the *ad hoc* modification of extending frequency integrals to  $-\infty$ . The pre-trace RWA may then misrepresent the effects of retarded propagation in the electromagnetic field, which suggests problems with causality in the study of multipartite systems.

Moreover, the Glauber detector model [75], long used in photodetection theory

and quantum optics, uses the pre-trace RWA, and it might give rise to quantum correlations between spacelike separated events that do not represent the effects of actual entanglement. The effective status of pre-trace RWA in Glauber’s theory is debated: some authors have shown that photodetection probabilities at short times appear to violate causality [146], and modifications to Glauber’s photodetection theory have been suggested [146, 59], while others indicate that a different form of the RWA in photodetection theory can guarantee causality [115]. Our interest in this problem partly arose from finding how the imposition of pre-trace RWA affects the range of validity of results from the calculation of the entanglement dynamics of two atoms interacting with a common quantum field at large atomic separation [10].

Some form of the RWA is often invoked in the quantum optics and atomic physics literature in derivations of the Born-Markov master equation for a system weakly coupled to bosonic reservoir. In such derivations, the Born-Markov master equation requires an RWA to render it in *Lindblad form* [22], thus providing a completely-positive dynamical map (for all states at all times) as is useful to assume for many quantum information theory discussions.

With these two distinct RWAs in widespread use while some open questions remain about their limitations and fallacies, we find it useful to carry out a systematic analysis of the consistency and applicability of the RWAs in the modern language of open quantum systems. This would be fitting in view of the fact that researchers today have to tackle problems beyond those of level population and dissipation rates to deal with more subtle issues such as quantum decoherence and

entanglement dynamics and perform more demanding tasks such as quantum state tomography and engineering.

In the analysis that follows, we find that the RWA may be sufficient or insufficient depending on what information is desired about the system. For the perturbative relaxation rates either the pre-trace or post-trace RWA is sufficient. To obtain the environmentally induced shifts in system frequencies, only the post-trace RWA is sufficient. In order to get more detailed information about the evolution of the quantum state and the asymptotic steady state neither RWA is sufficient in general. We also find that the pre-trace RWA does not in general have a Markovian limit.

This section is organized as follows: In Sec. 4.4.1 we review the RWA as it applies to interactions within a closed system. In Sec. 4.4.2 we discuss the consistency and applicability of the post-trace RWA. In Sec. 4.4.3 we discuss the same for the pre-trace RWA. In the last subsection we conclude with some discussions.

#### 4.4.1 The RWA in Closed Systems

We first examine the rotating-wave approximation as ordinarily applied to a closed system consisting of several interacting subsystems. Its wave function  $|\psi\rangle$  evolves as

$$\frac{d}{dt}|\psi\rangle = -i\mathbf{H}|\psi\rangle, \quad (4.64)$$

under the total Hamiltonian  $\mathbf{H} \equiv \mathbf{H}_0 + \mathbf{H}_1$  where  $\mathbf{H}_0$  represents the sum of all uncoupled subsystems and  $\mathbf{H}_1$  represents the part from the subsystems coupling.

One seeks to solve the eigenvalue problem

$$\mathbf{H} |\omega\rangle = \omega |\omega\rangle, \quad (4.65)$$

for eigenstates  $|\omega\rangle$  with eigenvalues  $\omega$  by perturbing off the free eigensystem

$$\omega_i = \omega_i^{(0)} + \delta\omega_i^{(1)} + \dots, \quad (4.66)$$

$$|\omega_i\rangle = |\omega_i^{(0)}\rangle + |\delta\omega_i^{(1)}\rangle + \dots, \quad (4.67)$$

where  $\omega_i^{(0)}$  are the eigenvalues of the uncoupled system. The *non-degenerate* first-order corrections are then

$$\delta\omega_i^{(1)} = \langle \omega_i^{(0)} | \mathbf{H}_1 | \omega_i^{(0)} \rangle, \quad (4.68)$$

$$\langle \omega_j^{(0)} | \delta\omega_i^{(1)} \rangle = \frac{\langle \omega_j^{(0)} | \mathbf{H}_1 | \omega_i^{(0)} \rangle}{\omega_i - \omega_j} \quad (\omega_i \neq \omega_j), \quad (4.69)$$

and for the *degenerate* corrections one must find the correct linear combination of degenerate states  $|f\rangle$ , which exist solely in the degenerate subspace  $|\omega_{d_i}^{(0)}\rangle$  wherein  $\mathbf{H}_0 |\omega_{d_i}^{(0)}\rangle = \omega_d |\omega_{d_i}^{(0)}\rangle$ .  $|f\rangle$  are the eigenstates of the degenerate interaction  $\mathbf{H}_d$

$$\mathbf{H}_d |f\rangle = f |f\rangle, \quad (4.70)$$

which possesses matrix elements

$$(H_d)_{ij} = \langle \omega_{d_i}^{(0)} | \mathbf{H}_1 | \omega_{d_j}^{(0)} \rangle, \quad (4.71)$$

This kind of analysis should also extend to *nearly-degenerate* subspaces where the basis corrections in (4.69) would nearly diverge.

In the rotating-wave approximation one only considers components of the interaction  $\mathbf{H}_1$  which oscillate least rapidly in the interaction picture. If these terms

are stationary, e.g. at resonance, then they will include the correct non-degenerate first-order frequencies, (4.68), as well as the correct characteristic equation, (4.71), which determines the degenerate first-order frequencies and zeroth-order energy states. What the RWA generally neglects are the first-order basis corrections, (4.69). If the RWA terms are non-stationary, then they will include the most nearly-degenerate first-order frequencies and also neglect their first order basis corrections. Therefore the RWA has limited correspondence to perturbation theory as long as all terms that are close to resonance have been retained in the interaction Hamiltonian. If, however, the RWA is made such that there are neglected terms that are near resonance, then the correspondence fails even in the weak coupling regime (because the missing first-order basis corrections become large, and even the first-order eigenvalue corrections are inaccurate). Moreover, for subsystems with a multiplicity of timescales, there may be several near-resonance frequencies which a proper application of RWA would have to take into account.

In the rest of this paper we consider open quantum systems and divide our attention between two cases: the post- and pre- trace RWA.

#### 4.4.2 The Post-Trace RWA

The RWA is an approximation that is employed in the weak-coupling regime and places the master equation into a Lindblad form. Under only the assumption of weak coupling, the master equation is not necessarily Markovian or of Lindblad form. While the master equation can assume a Lindblad form in the limit of vanishing

coupling, for any actual finite coupling it will generally differ from Lindblad form. It is a valid question to ask precisely what this approximation is doing by comparing it to the weak coupling dynamics without the RWA. As we will show, the post-trace RWA actually lies between the zeroth-order and second-order master equations.

The master equation for the RDM of an open quantum system can be written in the form

$$\dot{\rho} = \mathcal{L}_0\{\rho\} + \delta\mathcal{L}\{\rho\}, \quad (4.72)$$

where

$$\mathcal{L}_0\{\rho\} = -i[\mathbf{H}, \rho] \quad (4.73)$$

is the term that would already be present in the unitary dynamics of the closed system and  $\delta\mathcal{L}$  represents the dissipative corrections introduced by coupling to the environment as was considered in Chapter 2. The second-order perturbative master equation is generally non-Markovian in its dynamics and not of Lindblad form. The rotating-wave approximation is often introduced, not in a purely perturbatively derived master equation, but in one derived via the Born-Markov approximation. To second order in the system-environment interaction, the Born-Markov approximation is consistent with weak coupling perturbation, even well outside of the Markovian limit. Therefore, given that the RWA will only be applicable to second order, it is of no consequence if one starts from the Born-Markov approximation or a more rigorous perturbative analysis. One only has to keep in mind that RWA has no reliance upon any kind of Markovian approximation.

It may also be useful to note that Davies has derived [45, 47] a Lindblad-

form master equation in the weak-coupling regime with the additional requirement that one take the limit where the coupling  $\lambda$  vanishes and rescale time as  $\tau = \lambda^2 t$ , effectively taking a simultaneous  $t \rightarrow \infty$  limit. Our analysis supposes only weak (but non-vanishing) coupling and can be applied even at early times, although we focus on the late-time dynamics, so in principle we are restricted to neither limit.

The post-trace RWA effectively consists of only considering the parts of the super-operator  $\delta\mathcal{L}$  which commute with the free system propagation super-operator

$$\mathbf{G}_0(t)\{\rho\} = e^{-i\mathbf{H}t} \rho e^{+i\mathbf{H}t}. \quad (4.74)$$

If we consider evaluating our master equation coefficients in the energy basis, which in pseudo-Lindblad form amounts to resolving

$$\mathcal{D}_{ki;jl} = \sum_{nm} \mathcal{D}_{nm} \langle \omega_k | \mathbf{e}_n | \omega_i \rangle \langle \omega_j | \mathbf{e}_m^\dagger | \omega_l \rangle \quad (4.75)$$

with  $|\omega_j\rangle$  representing the system energy eigenstate with frequency  $\omega_j$ , then the RWA essentially amounts to projecting out the diagonal of this Hermitian matrix, i.e. terms with  $\omega_k - \omega_i = \omega_j - \omega_l$  or equivalently  $\omega_i - \omega_j = \omega_k - \omega_l$ . For the perturbative master equation to second order in the system-environment coupling, which is assumed to be weak, these diagonal entries will settle to positive values and therefore this projection yields a master equation of the Lindblad form. We will refer to the master equation obtained this way as the RWA-Lindblad equation.

Such a Lindblad projection is only reasonable because the system-environment coupling is assumed to be weak and the projection is performed in the energy basis. As was discussed in Chapter 2, eigen-operators of the open-system Liouvillian have the form  $|\omega_j\rangle\langle\omega_k|$  plus corrections at second order in the coupling strength so that the

discrepancy introduced by the dropped terms is small for sufficiently weak coupling. The RWA-Lindblad equation is not fully equivalent to the weak coupling master equation, but it generates an evolution which is very close to that of the weak coupling master equation in a perturbative sense.

#### 4.4.2.1 Correspondence With Perturbation Theory

Assuming the system-environment coupling to be weak, one can solve an open-system master equation in a perturbative fashion as was done in Chapter 2. One seeks a solution to the eigenvalue problem

$$\mathcal{L}\mathbf{o} = f\mathbf{o}, \quad (4.76)$$

for the operators  $\mathbf{o}$  which would evolve with characteristic rate  $f$ , given the perturbative expansions

$$\mathcal{L} = \mathcal{L}_0 + \delta\mathcal{L}, \quad (4.77)$$

$$\mathbf{o}_{ij} = |\omega_i\rangle\langle\omega_j| + \delta\mathbf{o}_{ij} + \dots, \quad (4.78)$$

$$f_{ij} = -i\omega_{ij} + \delta f_{ij} + \dots, \quad (4.79)$$

where the zeroth order terms are correctly set to match the free evolution of the system and  $\omega_{ij} \equiv \omega_i - \omega_j$ . For Gaussian noise, which has very well behaved asymptotic properties, the perturbative corrections are at minimum second order in the system-environment interaction. Therefore we will speak of all lowest order perturbative corrections as being second order. The second-order constraint upon our eigenvalue

problem is then

$$\langle \omega_k | \delta \mathcal{L} \{ |\omega_i\rangle\langle\omega_j| \} | \omega_l \rangle = -\imath(\omega_{ij} - \omega_{kl}) \langle \omega_k | \delta \mathbf{o}_{ij} | \omega_l \rangle + \delta f_{ij} \delta_{ij;kl} . \quad (4.80)$$

Evaluating the components of this equation yields the non-degenerate corrections.

$$\langle \omega_k | \delta \mathbf{o}_{ij} | \omega_l \rangle = \frac{\langle \omega_k | \delta \mathcal{L} \{ |\omega_i\rangle\langle\omega_j| \} | \omega_l \rangle}{-\imath(\omega_{ij} - \omega_{kl})} \text{ where } \omega_{ij} \neq \omega_{kl} , \quad (4.81)$$

$$\delta f_{ij} = \langle \omega_i | \delta \mathcal{L} \{ |\omega_i\rangle\langle\omega_j| \} | \omega_j \rangle . \quad (4.82)$$

For  $\mathbf{o}_{ii}$  and  $f_{ii}$  the system is degenerate, and one must solve the characteristic equation (Pauli master equation)

$$\mathbf{W} \vec{\sigma} = \delta f \vec{\sigma} , \quad (4.83)$$

$$[\mathbf{W}]_{ij} = \langle \omega_i | \delta \mathcal{L} \{ |\omega_j\rangle\langle\omega_j| \} | \omega_i \rangle , \quad (4.84)$$

$$[\vec{\sigma}]_i \equiv \langle \omega_i | \mathbf{o} | \omega_i \rangle , \quad (4.85)$$

for the branching under the perturbation.

The essential point is that the perturbative corrections to the eigenvalues are entirely captured by the post-trace RWA, while the perturbative basis corrections are entirely neglected. The lack of basis perturbation can lead to discrepancies (with the non-RWA evolution) even at late times, as was seen for example by Haikka and Maniscalco in [78]. We can readily see a late-time discrepancy in the thermal state of the system. The system evolving under the RWA-Lindblad equation will relax into the free thermal state described by the Boltzmann density matrix  $\rho \propto e^{-\beta \mathbf{H}}$ . But this is generally not how most systems would actually thermalize in a Hamiltonian formulation, as we will discuss in Sec. 4.5.

This property suggests a limitation to the applicability of post-trace RWA in the study of entanglement dynamics. In a multipartite system with components interacting only through the bath, this precludes the presence of asymptotic residual entanglement. This is in contrast to the asymptotic behaviour of bipartite systems which we will discuss in Chapter 6.

We also remark that if one takes the simultaneous limits of vanishing coupling and  $t \rightarrow \infty$  on the second-order master equation such that all  $\delta ft$  remain finite following Davies [45, 47], then one will similarly obtain a master equation of Lindblad form, which is exactly the RWA-Lindblad equation. Our perturbative approach is not restricted to such a limit, of course, and the difference between the perturbative weak-coupling dynamics and the post-trace RWA dynamics will also show the difference with the limit of vanishing coupling used by Davies.

#### 4.4.2.2 RWA Fails When Perturbation Theory Fails

The second-order master equation should be valid when the second-order corrections provided by  $\delta\mathcal{L}$  are small as compared to the unperturbed dynamics generated by  $\mathcal{L}_0$ . Let us denote the strength of the dissipative corrections generically by the frequency  $\gamma_D$ , then the weak coupling condition is most stringently

$$\gamma_D \ll \min_{\omega_{ij} \neq 0} |\omega_{ij}|, \quad (4.86)$$

as  $\omega_{ij}$  corresponds to the eigenvalues of  $\mathcal{L}_0$  (not  $\omega_i$  which corresponds to the eigenvalues of the unperturbed Schrödinger equation).

The RWA-Lindblad equation does not directly correspond to the second-order

master equation, but more correctly to the second-order solutions of said master equation. For the second-order solutions to be valid the coupling must not only be weak in the above sense but also in the following sense

$$\gamma_D \ll \min_{\omega_{ij} \neq \omega_{kl}} |\omega_{ij} - \omega_{kl}| . \quad (4.87)$$

which justifies the perturbative solutions. One cannot have near degeneracy in the energy level splittings or the naive perturbative solutions, which the RWA-Lindblad equation corresponds to, will fail. Perfect degeneracy is acceptable; the RWA-Lindblad equation retains these terms in the Pauli master equation for instance. But near-degeneracy needs to be treated in a manner analogous to degeneracy; the nearly degenerate subspace should be diagonalized. With the second-order master equation this is still possible, but with the RWA-Lindblad equation these terms have been discarded and one is left with an invalid master equation. Thus, there can be situations where the weak-coupling condition is satisfied while the post-trace RWA condition is not.

This problem arises, for example, in cavity QED. A two-level atom of frequency  $\Omega$  coupled to a resonant intracavity field mode will result in an energy spectrum of the composite system that has the form of the harmonic oscillator with each level split in two by the Rabi frequency  $\sqrt{n} \Omega_{VR}$  of the dressed states,

$$E_{n,\pm} = \hbar (n \Omega \pm \sqrt{n} \Omega_{VR}) . \quad (4.88)$$

If the intracavity field is coupled to the field outside the cavity, this becomes an open quantum system. If the intracavity field is coupled weakly enough to the atom, then the system will be in the weak-coupling regime of cavity QED and the vacuum Rabi

frequency  $\Omega_{VR}$  will be small compared to  $\gamma_D$ . In this case the post-trace RWA procedure does not, strictly speaking, apply as was noted by Scala et. al. [137].

In such a case one can still do a partial RWA, neglecting terms that oscillate much faster than  $\gamma_D$  and keeping those that are slower. This still leaves the master equation in pseudo-Lindblad form; however, assuming these timescales are sufficiently slow and the spectrum of environmental noise is sufficiently flat, one may be able to make an effective Markovian approximation for the remaining pseudo-Lindblad terms (even if one might not have been valid for original master equation due to the faster system dynamics that have been ignored in the post-trace RWA) to recover a Lindblad-form master equation. Scala et. al. argued this is the case for cavity QED with a low-temperature bath [135].

#### 4.4.2.3 Application to the Two-Level Atom

Here we consider a two-level system with  $\sigma_z$  Hamiltonian and energy level splitting  $\Omega$ , bilinearly coupled to a thermal reservoir via a  $\sigma_x$  coupling. This would, for example, model a two-level atom coupled to the electromagnetic field in the dipole approximation. Denoting our reduced density matrix

$$\boldsymbol{\rho} = \begin{bmatrix} \rho_{++} & \rho_{+-} \\ \rho_{-+} & \rho_{--} \end{bmatrix}, \quad (4.89)$$

one can compute the second-order master equation (see Chapter 2) and place it into the form

$$\frac{d}{dt} \begin{bmatrix} \rho_{++} \\ \rho_{--} \end{bmatrix} = \frac{\Gamma}{\cosh\left(\frac{\Omega}{2T}\right)} \begin{bmatrix} -e^{+\frac{\Omega}{2T}} & +e^{-\frac{\Omega}{2T}} \\ +e^{+\frac{\Omega}{2T}} & -e^{-\frac{\Omega}{2T}} \end{bmatrix} \begin{bmatrix} \rho_{++} \\ \rho_{--} \end{bmatrix}, \quad (4.90)$$

$$\frac{d}{dt} \begin{bmatrix} \rho_{+-} \\ \rho_{-+} \end{bmatrix} = \begin{bmatrix} -\Gamma - \imath(\Omega - \delta\Omega) & +\Gamma + \imath\delta\Omega \\ +\Gamma - \imath\delta\Omega & -\Gamma + \imath(\Omega - \delta\Omega) \end{bmatrix} \begin{bmatrix} \rho_{+-} \\ \rho_{-+} \end{bmatrix}, \quad (4.91)$$

with decoherence rate (here also the half thermalization rate) and energy level shift

$$\Gamma \equiv \Gamma(\Omega), \quad (4.92)$$

$$\delta\Omega \equiv \frac{2}{\pi} \int_0^\infty d\varepsilon \mathcal{P} \left[ \frac{\Omega}{\varepsilon^2 - \Omega^2} \right] \Gamma(\varepsilon), \quad (4.93)$$

in terms of the phenomenological decoherence rate function  $\Gamma(\omega)$ .  $\mathcal{P}$  denotes the Cauchy principal value which regulates contained poles from contributing to the integral.

This master equation and those that follow are exact to second order, only the coefficients have been allowed to relax to their asymptotic values. The relaxation occurs quickly, within the system and bath timescales, as compared to their effect, which occurs in the coupling timescale. Therefore, when considering properly correlated initial states which do not jolt, it is safe to consider this “late-time” regime.

In terms of the microscopically derived damping kernel  $\tilde{\gamma}(\omega)$ , the anti-derivative of the dissipation kernel, the decoherence rate can be expressed

$$\Gamma(\Omega) = \tilde{\gamma}(\Omega) \Omega \coth\left(\frac{\Omega}{2T}\right). \quad (4.94)$$

Regardless of system-environment coupling, the damping kernel is effectively constant for Ohmic coupling (which occurs for a fixed and ideal dipole in the electromagnetic field), which along with high temperature is responsible for thermal white noise. For linear coupling to the collective positions of a bath of harmonic oscillators the dissipation kernel has no more temperature dependence than the system-environment coupling itself.

The post-trace RWA here amounts to neglecting the dynamical interaction between  $\rho_{+-}$  and  $\rho_{-+}$ . To second order in the coupling, the only effect of this is to neglect a perturbative amount of phase information pertaining to their damped oscillations, i.e. the perturbative change of basis. The asymptotic state works out to be exactly the same in either case. Thus under the specific conditions leading to these results the post-trace RWA can be viewed as largely acceptable and somewhat innocuous.

#### 4.4.2.4 Application to Quantum Brownian Motion

The exact Quantum Brownian Motion (QBM) master equation [84] for the reduced dynamics of an oscillator bilinearly coupled (position-position) to a thermal reservoir of harmonic oscillators can be written

$$\dot{\rho} = -i[\mathbf{H}_L, \rho] - i\Gamma[\mathbf{X}, \{\mathbf{P}, \rho\}] - M D_{PP}[\mathbf{X}, [\mathbf{X}, \rho]] - D_{XP}[\mathbf{X}, [\mathbf{P}, \rho]], \quad (4.95)$$

where  $\mathbf{H}_L$  denotes the system Hamiltonian with frequency coefficient  $\Omega_L$ ,  $\Gamma$  the dissipation coefficient,  $D_{PP}$  and  $D_{XP}$  the regular and anomalous diffusion coefficients (see Chapter 3 for details). This master equation will have a stationary limit if the

noise correlation is not excessively widespread in time, e.g. a regulated Ohmic coupling is perfectly suitable. To lowest order in the coupling, the late-time expressions for these coefficients can be determined from the weak-coupling master equation (Chapter 2) to be

$$\Gamma = \Gamma(\Omega), \quad (4.96)$$

$$\Omega_R = \Omega - \frac{2}{\pi} \int_0^\infty d\varepsilon \mathcal{P} \left[ \frac{\varepsilon^2}{\varepsilon^2 - \Omega^2} \right] \Gamma(\varepsilon), \quad (4.97)$$

$$D_{PP} = \Gamma(\Omega) \Omega \coth\left(\frac{\Omega}{2T}\right), \quad (4.98)$$

$$D_{XP} = +\frac{2}{\pi} \int_0^\infty d\varepsilon \mathcal{P} \left[ \frac{1}{\varepsilon^2 - \Omega^2} \right] \Gamma(\varepsilon) \varepsilon \coth\left(\frac{\varepsilon}{2T}\right), \quad (4.99)$$

in terms of the phenomenological dissipation function, which is proportional to the damping kernel at second order. In QBM with linear coupling to the bath, the dissipation coefficient will always have no more temperature dependence than the system-environment coupling. Ohmic coupling here will imply a constant dissipation function, though a cutoff is required for this theory. The frequency shift will have a linear cutoff sensitivity which can be renormalized, but the anomalous diffusion  $D_{XP}$  has a logarithmic cutoff sensitivity at second order.

The Fokker-Planck equation for the pseudo-distribution function (Wigner function)  $W$  in a phase space representation presents a much cleaner picture with simple solutions.

$$\frac{d}{dt} W = \{ \nabla_{\mathbf{Z}}^T \mathcal{H} \mathbf{Z} + \nabla_{\mathbf{Z}}^T \mathbf{D} \nabla_{\mathbf{Z}} \} W, \quad (4.100)$$

$$\mathbf{Z} = [X, P]^T, \quad (4.101)$$

$$\nabla_{\mathbf{Z}} = \left[ \frac{\partial}{\partial X}, \frac{\partial}{\partial P} \right]^T, \quad (4.102)$$

The matrices  $\mathcal{H}$  and  $\mathbf{D}$  are the homogeneous and diffusion coefficient matrices respectively.

$$\mathcal{H} = \begin{bmatrix} 0 & -\frac{1}{M} \\ M\Omega_{\text{R}}^2 & 2\Gamma \end{bmatrix}, \quad (4.103)$$

$$\mathbf{D} = \begin{bmatrix} 0 & -\frac{1}{2}D_{xp} \\ -\frac{1}{2}D_{xp} & M D_{pp} \end{bmatrix}. \quad (4.104)$$

Do not confuse the homogeneous generator with the Hamiltonian; they differ by some frequency renormalization and the dissipation  $\Gamma$ . For simplicity we will assume  $\Omega_{\text{L}}$  has been renormalized to simply  $\Omega$ . The diffusion matrix contains two components: the regular diffusion  $D_{PP}$  and an anomalous anti-diffusion  $D_{XP}$  which keeps the position uncertainty insensitive to high frequency.

If one expresses the QBM master equation in terms of ladder operators, the pseudo-Lindblad coefficient matrix can be calculated to be

$$\mathcal{D} = \frac{1}{\Omega} \begin{bmatrix} D_{PP} - \Gamma \Omega & D_{PP} + \imath D_{XP} \Omega \\ D_{PP} - \imath D_{XP} \Omega & D_{PP} + \Gamma \Omega \end{bmatrix}. \quad (4.105)$$

The rotating-wave approximation then constitutes projecting out the diagonal of this matrix, which will be positive definite. Transforming back into the phase space representation, the Fokker-Plank coefficients become

$$\mathcal{H}_{\text{RWA}} = \begin{bmatrix} \Gamma & -\frac{1}{M} \\ M\Omega^2 & \Gamma \end{bmatrix}, \quad (4.106)$$

$$\mathbf{D}_{\text{RWA}} = \begin{bmatrix} \frac{D_{PP}}{2M\Omega^2} & 0 \\ 0 & \frac{MD_{PP}}{2} \end{bmatrix}. \quad (4.107)$$

The anomalous diffusion coefficient vanishes entirely while the dissipation and regular diffusion coefficients are both broken in half, with the missing half reappearing as an analogous coefficient of the master equation.

The role of the homogeneous coefficients are to generate the homogeneous propagator  $e^{-t\mathcal{H}}$  or damped oscillations. The RWA homogeneous coefficients are just slightly off in both the oscillation rates and phase; the dissipation rates are entirely correct. Compare the characteristic frequencies of the two matrices

$$h = \Gamma \pm i\sqrt{\Omega^2 - \Gamma^2}, \quad (4.108)$$

$$h_{\text{RWA}} = \Gamma \pm i\Omega. \quad (4.109)$$

The diffusion coefficients are relatively more mangled given that the anomalous coefficient is entirely absent; despite what is amiss in the early calculations of the weak coupling QBM master equation, this coefficient does exist at lowest order in the system-environment coupling. The effect of diffusion is only present in the second cumulant or covariance of the Wigner function. For this stationary master equation, the evolution of the covariance is simply

$$\boldsymbol{\sigma}(t) = e^{-t\mathcal{H}} [\boldsymbol{\sigma}(0) - \boldsymbol{\sigma}(\infty)] e^{-t\mathcal{H}^T} + \boldsymbol{\sigma}(\infty), \quad (4.110)$$

where the stationary covariance is determined by the Lyapunov equation

$$\mathcal{H}\boldsymbol{\sigma}(\infty) + \boldsymbol{\sigma}(\infty)\mathcal{H}^T = 2\mathbf{D}. \quad (4.111)$$

We can easily compare the stationary covariances.

$$\boldsymbol{\sigma}(\infty) = \begin{bmatrix} \frac{1}{M\Omega^2} \left( \frac{1}{2\Gamma} D_{PP} - D_{XP} \right) & 0 \\ 0 & \frac{M}{2\Gamma} D_{PP} \end{bmatrix}, \quad (4.112)$$

$$\boldsymbol{\sigma}_{\text{RWA}}(\infty) = \begin{bmatrix} \frac{1}{M\Omega^2} \frac{1}{2\Gamma} D_{PP} & 0 \\ 0 & \frac{M}{2\Gamma} D_{PP} \end{bmatrix}. \quad (4.113)$$

Amazingly the only difference in the stationary state will come from the lack of an anomalous diffusion coefficient. This contribution will ultimately be lower order in the coupling, due to the  $\Gamma^{-1}$  prefactor before  $D_{PP}$ , and therefore its absence is acceptable perturbatively.

### 4.4.3 The Pre-Trace RWA

#### 4.4.3.1 Inconsistency of approximation

Let us consider a bilinear interaction Hamiltonian  $\mathbf{H}_I$  between a system observable  $\mathbf{L}$  and the collective environment observable  $\mathbf{l}$ .

$$\mathbf{H}_I = \mathbf{L}\mathbf{l}, \quad (4.114)$$

For each of these operators, assuming them to be completely non-stationary, there is a *gross* raising and lowering decomposition  $\mathbf{L}_{\pm}$  given by

$$\mathbf{L}_+ = \sum_{i>j} \langle \omega_i | \mathbf{L} | \omega_j \rangle | \omega_i \rangle \langle \omega_j |, \quad (4.115)$$

$$\mathbf{L}_- = \sum_{i<j} \langle \omega_i | \mathbf{L} | \omega_j \rangle | \omega_i \rangle \langle \omega_j |, \quad (4.116)$$

such that

$$\mathbf{L} = \mathbf{L}_+ + \mathbf{L}_-, \quad (4.117)$$

$$\mathbf{J} = i(\mathbf{L}_+ - \mathbf{L}_-), \quad (4.118)$$

$$\mathbf{L}_\pm^\dagger = \mathbf{L}_\mp, \quad (4.119)$$

where  $\mathbf{L}$  and  $\mathbf{J}$  will be two relevant observables. For position coupling with a harmonic oscillator the decomposition becomes

$$\mathbf{L} = \mathbf{X}, \quad (4.120)$$

$$\mathbf{L}_+ = \frac{1}{\sqrt{2M\Omega}} \mathbf{a}^\dagger, \quad (4.121)$$

$$\mathbf{L}_- = \frac{1}{\sqrt{2M\Omega}} \mathbf{a}, \quad (4.122)$$

$$\mathbf{J} = \frac{1}{M\Omega} \mathbf{P}, \quad (4.123)$$

and for  $\sigma_x$  coupling with a  $\sigma_z$  Hamiltonian (two-level system) we have

$$\mathbf{L} = \sigma_x, \quad (4.124)$$

$$\mathbf{L}_\pm = \frac{1}{2} \sigma_\pm, \quad (4.125)$$

$$\mathbf{J} = -\sigma_y. \quad (4.126)$$

Now consider coupling the system to an environment made of a large number of harmonic oscillators in their collective positions. Let us furthermore assume the system coupling is like that of the above harmonic oscillator or two-level system such that it is characterized by a single frequency  $\omega$ .

$$\mathbf{l} = \sum_k g_k \mathbf{x}_k, \quad (4.127)$$

$$\mathbf{H}_I = \sum_k \frac{g_k}{\sqrt{m_k \varepsilon_k}} \left\{ \left( \mathbf{L}_+ \mathbf{a}_k + \mathbf{L}_- \mathbf{a}_k^\dagger \right) + \left( \mathbf{L}_+ \mathbf{a}_k^\dagger + \mathbf{L}_- \mathbf{a}_k \right) \right\}, \quad (4.128)$$

where  $\mathbf{x}_k$  is the environment position operator with ladder operator  $\mathbf{a}_k$ , energy  $\varepsilon_k$  and mass  $m_k$ . In the interaction picture we have the interaction Hamiltonian

$$\begin{aligned} \mathbf{H}_I(t) = & \sum_k \frac{g_k}{\sqrt{2m_k\varepsilon_k}} \left( \mathbf{L}_+ \mathbf{a}_k e^{+i(\omega-\varepsilon_k)t} + \mathbf{L}_- \mathbf{a}_k^\dagger e^{-i(\omega-\varepsilon_k)t} \right) \\ & + \sum_k \frac{g_k}{\sqrt{2m_k\varepsilon_k}} \left( \mathbf{L}_+ \mathbf{a}_k^\dagger e^{+i(\omega+\varepsilon_k)t} + \mathbf{L}_- \mathbf{a}_k e^{-i(\omega+\varepsilon_k)t} \right). \end{aligned} \quad (4.129)$$

An often utilized pre-trace rotating-wave approximation is to neglect the second terms, conventionally referred to as counter-rotating terms, as they are deemed more rapidly oscillating than the first. However, this is only true in a mode-by-mode comparison. Keeping terms of frequency  $|\omega - 2\omega| = \omega$  while discarding terms of frequency  $|\omega + 0| = \omega$  serves no good purpose. There is no a priori sense in which this is an approximation at all, unless the only environment modes which exist are near resonance. More accurately this approximation is in the spirit of the Friedrichs approximation [71], which is the extent of the interaction which is mathematically solvable for a zero-temperature environment with arbitrary system.

A true bandwidth approximation which does what the pre-trace RWA claims would instead modify the interaction Hamiltonian of (4.129) (before tracing out the environment) by neglecting all the “rapid” terms that oscillate with a frequency outside some frequency band  $\Delta\omega$  in the interaction picture while retaining all the slower terms<sup>3</sup>. The resulting Hamiltonian would be

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<sup>3</sup>Note that this “bandwidth” Hamiltonian does *not* arise from restricting the field to some bandwidth of modes around the resonance frequency.

$$\mathbf{H}_I(t) = \sum_{\varepsilon_k=0}^{\Delta\omega+\omega} \frac{g_k}{\sqrt{2m_k\varepsilon_k}} \left( \mathbf{L}_+ \mathbf{a}_k e^{+i(\omega-\varepsilon_k)t} + \mathbf{L}_- \mathbf{a}_k^\dagger e^{-i(\omega-\varepsilon_k)t} \right) + \sum_{\varepsilon_k=0}^{\Delta\omega-\omega} \frac{g_k}{\sqrt{2m_k\varepsilon_k}} \left( \mathbf{L}_+ \mathbf{a}_k^\dagger e^{+i(\omega+\varepsilon_k)t} + \mathbf{L}_- \mathbf{a}_k e^{-i(\omega+\varepsilon_k)t} \right) \quad (4.130)$$

Note that if  $\Delta\omega < \omega$ , then the bandwidth approximated Hamiltonian would have no counter-rotating terms. Furthermore, if the environment were such that all environmental frequencies  $\varepsilon_k$  lie in a band around resonance with  $|\varepsilon_k - \omega| < \omega$ , then a bandwidth approximation using this band would be equivalent to dropping all counter-rotating terms. However, in the general case the two approximations are inequivalent, and simply dropping all counter-rotating terms is inconsistent.

It is also important to note that if the bandwidth approximation of (4.130) is performed with  $\Delta\omega$  chosen such that all near-degenerate terms are retained, then this is just the sort of RWA we discussed in Sec. 4.4.1. The only difference is that the environment is to be traced out at the end of the calculation. However, such a bandwidth approximation would render the problem more difficult to solve than simply calculating a full perturbative solution.

#### 4.4.3.2 Noise and the Markovian Limit

The Hamiltonian obtained after RWA is not generally an approximation of the full interaction Hamiltonian for reservoirs. It is nonetheless a linear Hamiltonian interaction with a thermal reservoir and will affect dissipation, decoherence, thermalization, etc. Therefore it still possesses some of the same character to the original model.

Back in terms of observables, the RWA interaction Hamiltonian takes the form

$$\mathbf{H}_I = \frac{1}{2} \mathbf{L} \sum_k g_k \mathbf{x}_k + \frac{1}{2} \mathbf{J} \sum_k g_k \frac{\mathbf{p}_k}{m_k \omega_k}, \quad (4.131)$$

and thus it describes a different but related set of system variables coupled to a different, but related set of bath variables. This results in two quantum noise sources

$$\mathbf{l}_{\text{RWA}} = \frac{1}{2} \sum_k g_k \mathbf{x}_k, \quad (4.132)$$

$$\mathbf{j}_{\text{RWA}} = \frac{1}{2} \sum_k g_k \frac{\mathbf{p}_k}{m_k \omega_k}, \quad (4.133)$$

which have not only autocorrelations in and of themselves but cross-correlations between themselves. Perhaps more clearly, if we consider the original damping kernel with one  $\mathbf{L}$ -coupled source

$$\tilde{\gamma}(\omega) = \tilde{\gamma}(\omega) \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix}, \quad (4.134)$$

then the RWA damping kernel with both  $\mathbf{L}$  and  $\mathbf{J}$ -coupled sources becomes

$$\tilde{\gamma}_{\text{RWA}}(\omega) = \frac{\tilde{\gamma}(\omega)}{4} \begin{bmatrix} 1 & -\imath \text{sign}(\omega) \\ +\imath \text{sign}(\omega) & 1 \end{bmatrix}, \quad (4.135)$$

with reference to the original damping kernel  $\tilde{\gamma}(\omega)$ . The diagonal components come from the self-correlations  $\langle \mathbf{l}(t) \mathbf{l}(\tau) \rangle_{\text{B}}$  and  $\langle \mathbf{j}(t) \mathbf{j}(\tau) \rangle_{\text{B}}$ , while the off-diagonal components come from the cross-correlations  $\langle \mathbf{l}(t) \mathbf{j}(\tau) \rangle_{\text{B}}$  and  $\langle \mathbf{j}(t) \mathbf{l}(\tau) \rangle_{\text{B}}$ .

There is a subtle pathology in the cross-correlations of these two noise sources. The RWA interaction is an example of couplings to *different* kinds of bath observables with strong cross-coupling. Such couplings do always not admit a Markovian limit. The reason for this is because in addition to high temperature, the white

noise limit also requires a local damping kernel, i.e. one constant in the Fourier domain. This is not a problem with one noise source as one can typically choose an appropriate coupling, e.g. Ohmic, such that the damping kernel will work out to be local. But with multivariate noise one must make all components of the damping tensor local, including new kinds of terms which arise from the cross-correlations. Whether or not this is possible depends in part upon any relation between the self-correlations and the cross-correlations.

For the RWA damping tensor, if we make the diagonal components local with what was Ohmic coupling, then the off-diagonal components will appear highly non-local like  $\text{sign}(\omega)$ . But if we were to choose a coupling as to make the off-diagonal components local, then the diagonal components will necessarily be highly non-local. There is no choice of coupling which can give us white noise. This problem with the white noise limit of pre-trace RWA has been noted before [152], although they further asserted that there was no fluctuation-dissipation relation. From our perspective the FDR naturally follows from Eq. (4.135) relating to a  $2 \times 2$  noise kernel, and so it exists though it is not as obvious in the context of other formalisms.

#### 4.4.3.3 Correspondence with Perturbation Theory

The perturbative correspondence between the pre-trace RWA and the original model is a bit more complicated to demonstrate. Let us start with the second-order corrections for our simple separable coupling without any sort of RWA following

Chapter 2.

$$\begin{aligned}
\langle \omega_k | \delta \mathcal{L} \{ |\omega_i\rangle\langle\omega_j| \} | \omega_l \rangle &= \langle \omega_k | \mathbf{L} | \omega_i \rangle [A(\omega_{ki}) + \bar{A}(\omega_{lj})] \langle \omega_j | \mathbf{L} | \omega_l \rangle \\
&\quad - \delta_{lj} \sum_h \langle \omega_k | \mathbf{L} | \omega_h \rangle A(\omega_{hi}) \langle \omega_h | \mathbf{L} | \omega_i \rangle \\
&\quad - \delta_{ki} \sum_h \langle \omega_j | \mathbf{L} | \omega_h \rangle \bar{A}(\omega_{hj}) \langle \omega_h | \mathbf{L} | \omega_l \rangle , \tag{4.136}
\end{aligned}$$

with late-time master equation coefficients

$$A(\omega) \equiv \frac{1}{2} \tilde{\alpha}(\omega) - \frac{i}{2\pi} \int_{-\infty}^{+\infty} d\varepsilon \mathcal{P} \left[ \frac{1}{\omega - \varepsilon} \right] \tilde{\alpha}(\varepsilon), \tag{4.137}$$

$$\alpha(t) \equiv \langle \mathbf{I}(t) \mathbf{I}(0) \rangle_{\text{B}}, \tag{4.138}$$

where  $\alpha$  is the quantum noise correlation for our stationary bath. These corrections capture all of the second-order relaxation rates, perturbative frequency shifts, and basis corrections.

As we have discussed, the post-trace RWA essentially considers taking only the diagonal entries where  $\omega_{ki} = \omega_{lj}$ , and under appropriate conditions this is sufficient to reproduce all of the perturbative frequency shifts and relaxation rates but not the basis corrections.

$$\begin{aligned}
\langle \omega_i + \omega | \delta \mathcal{L} \{ |\omega_i\rangle\langle\omega_j| \} | \omega_j + \omega \rangle &= \langle \omega_i + \omega | \mathbf{L} | \omega_i \rangle 2 \operatorname{Re}[A(\omega)] \overline{\langle \omega_j + \omega | \mathbf{L} | \omega_j \rangle} \\
&\quad - \delta_{0\omega} \sum_{\omega'} A(\omega') |\langle \omega_i + \omega' | \mathbf{L} | \omega_i \rangle|^2 \\
&\quad - \delta_{0\omega} \sum_{\omega'} \bar{A}(\omega') |\langle \omega_j + \omega' | \mathbf{L} | \omega_j \rangle|^2 . \tag{4.139}
\end{aligned}$$

One can see that the first terms, which directly correspond to the pseudo-Lindblad dissipator, are now only determined by the real part of  $A(\omega)$  or the characteristic

function of the noise correlation  $\tilde{\alpha}(\omega)$ . This function is always positive by Bochner's theorem.

The pre-trace RWA master equation has four related sets of terms because of the two correlated noise sources. But as far as these diagonal terms are concerned, which determine the perturbative timescales, one can essentially consider a master equation of the same form but with the modified coefficients

$$A_{\text{RWA}}(\omega) = \frac{1}{2}\tilde{\alpha}(\omega) - \frac{i}{2\pi} \int_{-\infty}^{+\infty} d\varepsilon \frac{1 + \text{sign}(\omega) \text{sign}(\varepsilon)}{2} \mathcal{P} \left[ \frac{1}{\omega - \varepsilon} \right] \tilde{\alpha}(\varepsilon). \quad (4.140)$$

The real part, which determines the relaxation rates, remains unchanged. But the imaginary part, which determines the energy level shifts, is very different. So while the post-trace RWA can correctly produce all of the perturbative timescales, the pre-trace RWA can only produce the relaxation rates, consistent with what has been found in earlier specific cases [5, 3].

#### 4.4.3.4 Non-Markovian Nature of the Master Equation

It is necessary to point out that, although the pre-trace RWA can often produce a master equation of Lindblad form, the coefficients are inherently non-Markovian. Even though the master equation is in a convolutionless form, the coefficients themselves contain integrals over the system's history alongside nonlocal correlations of the noise. As such, they cannot be universally applied to different systems (which would have different histories) even if one only wants the relaxation rates. This was easy to notice for the post-trace RWA as the correctly derived master equation coefficients would come out to be completely different. Here the reason is much the

same.

For instance, let us consider an oscillator system with  $\mathbf{X}$  coupling to the environment. The accuracy of the pre-trace RWA decay rates stems from a correct  $\mathbf{a}^\dagger$ ,  $\mathbf{a}$  raising and lowering operator decomposition of  $\mathbf{X}$ . This leads to a different but related model of environmental interaction with  $\mathbf{X}$  and  $\mathbf{P}$ -coupled noise. We have proven that the perturbative decay timescales will work out to be equivalent, but only by using the raising and lowering properties. If we couple this oscillator to additional degrees of freedom in some larger system, then  $\mathbf{a}^\dagger$  and  $\mathbf{a}$  are no longer ensured to be raising and lowering operators for the new energy eigenstates of the system. Once this criterion has been broken, the proof fails to apply and all coefficients of the misapplied master equation will likely be wrong. A correct pre-trace RWA interaction would have to involve a raising and lowering operator decomposition which utilizes the full Hamiltonian of the larger system.

#### 4.4.3.5 Application to the Two-Level Atom

Utilizing the second-order master equation, we find the RWA interaction Hamiltonian yields

$$\frac{d}{dt} \begin{bmatrix} \rho_{++} \\ \rho_{--} \end{bmatrix} = \frac{\Gamma}{\cosh\left(\frac{\Omega}{2T}\right)} \begin{bmatrix} -e^{+\frac{\Omega}{2T}} & +e^{-\frac{\Omega}{2T}} \\ +e^{+\frac{\Omega}{2T}} & -e^{-\frac{\Omega}{2T}} \end{bmatrix} \begin{bmatrix} \rho_{++} \\ \rho_{--} \end{bmatrix}, \quad (4.141)$$

$$\frac{d}{dt} \begin{bmatrix} \rho_{+-} \\ \rho_{-+} \end{bmatrix} = \begin{bmatrix} -\Gamma - \imath(\Omega - \delta\Omega_*) & +\Gamma + \imath\delta\Omega_* \\ +\Gamma - \imath\delta\Omega_* & -\Gamma + \imath(\Omega - \delta\Omega_*) \end{bmatrix} \begin{bmatrix} \rho_{+-} \\ \rho_{-+} \end{bmatrix}, \quad (4.142)$$

with new energy level shift

$$\delta\Omega_\star \equiv \frac{1}{\pi} \int_0^\infty d\varepsilon \mathcal{P} \left[ \frac{1}{\varepsilon - \Omega} \right] \Gamma(\varepsilon). \quad (4.143)$$

In addition to differing from the frequency shift without the RWA, it also contains a higher order cutoff sensitivity. For approximately local dissipation, the sensitivity was logarithmic but is now linear.

#### 4.4.3.6 Application to Quantum Brownian Motion

Again utilizing the second-order master equation, we find the RWA interaction Hamiltonian yields

$$\dot{\rho} = -i[\mathbf{H}_L^\star, \rho] - i\Gamma[\mathbf{X}, \{\mathbf{P}, \rho\}] - MD_{PP}[\mathbf{X}, [\mathbf{X}, \rho]] - D_{XP}^\star[\mathbf{X}, [\mathbf{P}, \rho]], \quad (4.144)$$

with new frequency shift and anomalous diffusion coefficient

$$\Omega_L^\star = \Omega - \frac{1}{\pi} \int_0^\infty d\varepsilon \mathcal{P} \left[ \frac{\varepsilon}{\varepsilon - \Omega} \right] \Gamma(\varepsilon), \quad (4.145)$$

$$D_{XP}^\star = +\frac{1}{\pi} \int_0^\infty d\varepsilon \mathcal{P} \left[ \frac{1}{\varepsilon - \Omega} \right] \Gamma(\varepsilon) \varepsilon \coth\left(\frac{\varepsilon}{2T}\right). \quad (4.146)$$

For what was Ohmic coupling in the original model, the frequency shift has a different but still linear cutoff sensitivity. However the anomalous diffusion coefficient now also has a linear cutoff sensitivity. If the cutoff is very large, this could be very problematic.

#### 4.4.3.7 A Multipartite Example

Let us say that we have an array of, otherwise non-interacting, parallel q-bits all with  $\sigma_z$  Hamiltonians. A simple dipole interaction can be represented with the

bilinear interaction Hamiltonian

$$\mathbf{H}_I = \sum_n \sigma_{x_n} \mathbf{l}_n \quad (4.147)$$

where  $\sigma_{x_n}$  is the  $x$  spin component of the  $n^{\text{th}}$  qubit and  $\mathbf{l}_n$  is its corresponding collective environment coupling. The environmental coupling for a qubit at location  $\mathbf{r}_n$  is

$$\mathbf{l}_n = \sum_{\mathbf{k}} g_k \left\{ e^{+i\mathbf{k}\cdot\mathbf{r}_n} \mathbf{a}_{\mathbf{k}}^\dagger + e^{-i\mathbf{k}\cdot\mathbf{r}_n} \mathbf{a}_{\mathbf{k}} \right\}, \quad (4.148)$$

with  $g_k \propto 1/\sqrt{\varepsilon_k}$  for an scalar field environment such as will be discussed in Chapter 6. The resultant damping kernel corresponding to the  $\langle \mathbf{l}_n(t) \mathbf{l}_m(\tau) \rangle_B$  correlation is

$$\tilde{\gamma}_{nm}(\omega) = \tilde{\gamma}_0 \text{sinc}(r_{nm}\omega), \quad (4.149)$$

where  $\mathbf{r}_{nm} = \mathbf{r}_n - \mathbf{r}_m$  and therefore the damping is Ohmic or local for the autocorrelations where  $r_{nm} = 0$ . The cross-correlations, which are strictly nonlocal, vanish in the limit of large distance separation.

The pre-trace RWA interaction, which was considered for two qubits in [10], introduces the duplication of quantum noise sources

$$\mathbf{l}_n^{\text{RWA}} = \frac{1}{2} \sum_{\mathbf{k}} g_k \left\{ e^{+i\mathbf{k}\cdot\mathbf{r}_n} \mathbf{a}_{\mathbf{k}}^\dagger + e^{-i\mathbf{k}\cdot\mathbf{r}_n} \mathbf{a}_{\mathbf{k}} \right\}, \quad (4.150)$$

$$\mathbf{j}_n^{\text{RWA}} = \frac{i}{2} \sum_{\mathbf{k}} g_k \left\{ e^{+i\mathbf{k}\cdot\mathbf{r}_n} \mathbf{a}_{\mathbf{k}}^\dagger - e^{-i\mathbf{k}\cdot\mathbf{r}_n} \mathbf{a}_{\mathbf{k}} \right\}, \quad (4.151)$$

and one must now consider the correlations between all such operators. The resultant damping kernels can be organized

$$\begin{bmatrix} \tilde{\gamma}_{l_n l_m}^{\text{RWA}}(\omega) & \tilde{\gamma}_{l_n j_m}^{\text{RWA}}(\omega) \\ \tilde{\gamma}_{j_n l_m}^{\text{RWA}}(\omega) & \tilde{\gamma}_{j_n j_m}^{\text{RWA}}(\omega) \end{bmatrix} = \frac{\tilde{\gamma}_{nm}(\omega)}{4} \begin{bmatrix} 1 & -i \text{sign}(\omega) \\ +i \text{sign}(\omega) & 1 \end{bmatrix}, \quad (4.152)$$

with reference to the original damping kernel  $\tilde{\gamma}_{nm}(\omega)$ . The scenario is much the same. The damping rates will be correct, while the frequency shifts and basis corrections (including asymptotic entanglement) will be incorrect. There is no longer a white noise limit, even when the qubits are distantly separated.

However this remains a fairly reasonable physical theory, as the RWA interaction itself was fairly reasonable. The cross-correlations between *different* qubits still vanishes for large separations. The second-order master equation, being determined by the second-order cumulants or two-time correlations, will therefore reduce to that of qubits coupled to independent environments in the large separation limit.

## 4.5 The Second-Order Thermal State

In the limit of vanishing system-environment interaction the system should asymptote to the equilibrium state  $\rho_\beta \propto e^{-\beta\mathbf{H}}$ . But this is not the equilibrium state of the system + environment with non-vanishing interaction. In this section we will demonstrate the known consistency with the local equilibrium state and give some clarification to the global equilibrium state. More specifically we will show that both the master equation and Schrödinger equation are consistent with the equilibrium state of the system + environment with non-vanishing interaction.

Unlike the RWA-Lindblad equation, the second-order master equation is capable of producing not only the zeroth-order thermal Green's function  $e^{-\beta\mathbf{H}}$ , but also the off-diagonal perturbative corrections. From Eq. (4.81), the second-order

Green's function must be

$$\langle \omega_i | \rho_\beta | \omega_j \rangle \propto e^{-\beta \omega_i} \delta_{ij} - \frac{i}{\omega_{ij}} \langle \omega_i | \mathcal{L}_2 \{ e^{-\beta \mathbf{H}} \} | \omega_j \rangle_{i \neq j} \quad (4.153)$$

plus diagonal perturbations which rely upon the fourth-order Pauli master equation.

The second-order thermal Green's function in fact corresponds to the reduced thermal Green's function of the system + environment with nonvanishing interaction

$$\rho_\beta = \frac{1}{Z_C(\beta)} \text{Tr}_E [ e^{-\beta(\mathbf{H} + \mathbf{H}_E + \mathbf{H}_I)} ] , \quad (4.154)$$

at least as far as we have checked perturbatively. Obviously expressions (4.153) and (4.154) agree at zeroth order. To validate their agreement at second order will require a perturbative expansion of Eq. (4.154). There exists such a perturbative expansion of exponential matrices utilizing the identity

$$\frac{d}{d\epsilon} e^{\mathbf{A} + \epsilon \mathbf{B}} = e^{\mathbf{A} + \epsilon \mathbf{B}} \int_0^1 d\eta e^{-\eta(\mathbf{A} + \epsilon \mathbf{B})} \mathbf{B} e^{+\eta(\mathbf{A} + \epsilon \mathbf{B})} , \quad (4.155)$$

to obtain an operator-Taylor series in the perturbation  $\epsilon \mathbf{B}$ . After a fair amount of simplification, one can determine the second-order reduced thermal Green's function for a Gaussian environment to be

$$\rho_\beta \propto e^{-\beta \mathbf{H}} + e^{-\beta \mathbf{H}} \int_0^\beta d\beta' \int_0^{\beta'} d\beta'' \langle \underline{\mathbf{H}}_I(-i\beta') \underline{\mathbf{H}}_I(-i\beta'') \rangle_E , \quad (4.156)$$

in terms of the complex-time operators

$$\underline{\mathbf{H}}_I(-i\beta) \equiv e^{+\beta(\mathbf{H} + \mathbf{H}_E)} \mathbf{H}_I e^{-\beta(\mathbf{H} + \mathbf{H}_E)} , \quad (4.157)$$

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. For our sum of separable couplings the double integrals reduce to

$$\sum_{nm} \int_0^\beta d\beta' \int_0^{\beta'} d\beta'' \alpha_{nm}(-i\beta', -i\beta'') \underline{\mathbf{L}}_n(-i\beta') \underline{\mathbf{L}}_m(-i\beta''), \quad (4.158)$$

in terms of the complex-time operators

$$\underline{\mathbf{L}}(-i\beta) \equiv e^{+\beta \mathbf{H}} \mathbf{L} e^{-\beta \mathbf{H}}. \quad (4.159)$$

Finally expressions (4.153) and (4.158) can be compared term-by-term in the energy basis wherein the imaginary-time integrals of Eq. (4.158) can be easily resolved as the master equation operators were. The two expressions can be seen to be equivalent, even in their missing diagonal perturbations, after sufficient application of the thermal symmetry inherent in all thermal correlations, Eq. (2.200). This was also done for univariate noise in Ref. [118].

We know that Eq. (4.153) is missing diagonal perturbations; the same discrepancy in Eq. (4.158) stems from the expansion being inherently secular in  $\beta$ . Therefore neither result is sufficient to second-order in the low temperature regime. This is analogous to how the perturbative series for the propagator  $\mathcal{G}(t)$  in Eq. (2.6) is secular in time.

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

$$\boldsymbol{\psi} = \boldsymbol{\psi}_0 + \boldsymbol{\psi}_1 + \boldsymbol{\psi}_2 + \cdots, \quad (4.160)$$

$$\boldsymbol{\psi}_0 \equiv |0\rangle \otimes |0\rangle_{\text{E}}, \quad (4.161)$$

and then traces out the environment

$$\rho_\infty = |0\rangle\langle 0| + \left\langle \psi_2 \psi_0^\dagger + \psi_1 \psi_1^\dagger + \psi_0 \psi_2^\dagger \right\rangle_E + \dots, \quad (4.162)$$

here for Gaussian noise where the odd moments vanish. Without loss of generality let us set the ground-state energy of the system to zero. The second-order reduced ground state of ordinary perturbation theory can then be seen to coincide with the partial results of the second-order master equation

$$\langle \omega_i | \delta \rho_\beta | \omega_j \rangle = \sum_{nmk} \frac{C_{ijk}^{nm}}{Z_0(\beta)} \langle \omega_i | \mathbf{L}_m | \omega_k \rangle \langle \omega_k | \mathbf{L}_n | \omega_j \rangle, \quad (4.163)$$

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

$$C_{iik}^{nm} \equiv \text{An} \left[ e^{-\beta\omega_k} \frac{d}{d\omega_i} A_{nm}(\omega_{ik}) + e^{-\beta\omega_i} \frac{d}{d\omega_i} A_{mn}(\omega_{ki}) \right], \quad (4.164)$$

and off-diagonal coefficients given by

$$C_{ijk}^{nm} \equiv \text{An} \left[ e^{-\beta\omega_k} \frac{A_{nm}(\omega_{ik}) - A_{nm}(\omega_{jk})}{\omega_i - \omega_j} + \frac{e^{-\beta\omega_i} A_{mn}(\omega_{ki}) - e^{-\beta\omega_j} A_{mn}(\omega_{kj})}{\omega_i - \omega_j} \right], \quad (4.165)$$

with the resonance limit of the first off-diagonal term taken to be

$$\lim_{\omega_{ij} \rightarrow 0} \frac{A_{nm}(\omega_{ik}) - A_{nm}(\omega_{jk})}{\omega_i - \omega_j} = \frac{d}{d\omega_i} A_{nm}(\omega_{ik}), \quad (4.166)$$

and for the remaining term, the resonance limit can only be taken after the zero-temperature limit, where it would vanish trivially. Finite-temperature results derived from the master equation, and without use of degenerate perturbation theory, are not valid near resonance. Eq. (6.66) is exact for zero temperature and our best

guess for the positive-temperature coefficients: it has the correct functional dependence upon the Boltzmann weight and fourth-order master equation coefficients. At worst this is an interpolation of the zero and high-temperature states.

## 4.6 The Fluctuation-Dissipation Inequality

The *fluctuation-dissipation relation* (FDR) is usually formulated for a system interacting with a heat bath at finite temperature in the context of linear response theory, where only small deviations from the mean are considered. In Sec. 2.5.2.3 we briefly demonstrated that for an open quantum system interacting with a non-equilibrium environment, even where temperature is no longer a valid notion, a *fluctuation-dissipation inequality* (FDI) exists. Clearly stated, quantum fluctuations are bounded below by quantum dissipation, whereas classically the fluctuations can be made to vanish. The lower bound of this inequality is exactly satisfied by (zero-temperature) quantum noise and is in accord with the Heisenberg uncertainty principle (HUP). FDI violating quantum noise can be viewed as arising from HUP violating states of the environment and can induce HUP violating states in the open system.

Like the HUP, the FDI is mathematical in nature and is a precise inequality which must be obeyed in all regimes. Whereas, in its most general form the HUP relates two quantum operators of arbitrary systems, the FDI relates the two-time noise and dissipation (or equivalently, damping) kernels of arbitrary environments. Though it is exact, we will mostly discuss its context and physical implications in the

regime of weak coupling to an environment with stationary correlations. Moreover, for linear systems we additionally show that the non-equilibrium non-equilibrium FDR (which must satisfy the FDI) determines the non-equilibrium uncertainty relation (which must satisfy the HUP).

We have already derived the FDI in Sec. 2.5.2.3 and contrast it to equilibrium FDR, however here we will also work from the other end and motivate the FDI phenomenologically, but less generally. This result also produces the (weak-coupling) non-equilibrium uncertainty relation for quantum Brownian motion, which can be contrasted to the finite-temperature uncertainty relation [87, 88, 11, 7].

#### 4.6.1 Non-Equilibrium FDR & FDI

From the definitions of the multivariate noise kernel  $\boldsymbol{\nu}$  (2.168), dissipation kernel  $\boldsymbol{\mu}$  (2.169) and damping kernel  $\boldsymbol{\gamma}$  (2.176), one can prove the *fluctuation-dissipation inequality*:

$$\boldsymbol{\nu}(t, \tau) \geq \pm \boldsymbol{\mu}(t, \tau), \quad (4.167)$$

most generally, and in the case of stationary noise it takes the more useful form

$$\tilde{\boldsymbol{\nu}}(\omega) \geq \pm \omega \tilde{\boldsymbol{\gamma}}(\omega), \quad (4.168)$$

in the Fourier domain where the  $\omega$  would denote energy level transitions of the system. To prove this one simply notes that the noise kernel is the sum of two positive-definite kernels whereas the dissipation kernel is given by their difference. The essential point is that if there is any damping, or amplification, there will be quantum noise and Eq. (4.168) determines its lower bound. This is quite a departure

from classical physics where noise can be made to vanish in the zero-temperature limit, although the lower bound of this inequality is satisfied by zero-temperature quantum noise since  $\tilde{\alpha}(|\omega|) = 0$  in that case.

For the case of one collective system coupling, coupled to one or more environments, it is sufficient to define a fluctuation-dissipation relation

$$\tilde{\nu}(\omega) = \tilde{\kappa}(\omega) \tilde{\gamma}(\omega), \tilde{\kappa}(\omega) \equiv \frac{\tilde{\nu}(\omega)}{\tilde{\gamma}(\omega)}, \quad (4.169)$$

with  $\tilde{\kappa}(\omega)$  the fluctuation-dissipation kernel [84, 85] which relates fluctuations to dissipation. For multivariate noise one might use the symmetrized product

$$\tilde{\nu}(\omega) = \frac{1}{2} [\tilde{\kappa}(\omega) \tilde{\gamma}(\omega) + \tilde{\gamma}(\omega) \tilde{\kappa}(\omega)], \quad (4.170)$$

which would ensure  $\tilde{\kappa}(\omega)$  to be positive definite if  $\tilde{\gamma}(\omega)$  is, in accord with this being a (continuous) Lyapunov equation [18]. We will use this particular definition for quantum Brownian motion in the next section. Inequality (4.168) can now be restated as

$$\tilde{\kappa}(\omega) \geq |\omega|, \quad (4.171)$$

for damping environments. Typically  $\tilde{\kappa}(\omega)$  will contain dependence upon the precise nature of the environment couplings  $\mathbf{I}_n$ . As discussed in Sec. 2.5.3, *the* FDR:

$$\tilde{\kappa}_T(\omega) \equiv \omega \coth\left(\frac{\omega}{2T}\right), \quad (4.172)$$

is unique in that it is coupling independent, universal for equilibrium environments, and always provides detailed balance in a coupling invariant manner. One should be careful to note that the thermal FDR is not special because it exists,

nor because of its simple form, but because of its invariance to couplings (model-independence). “Observable dependence” for non-equilibrium correlations was also noticed in Ref. [110]. As a concrete example of an elegant yet non-equilibrium FDR, the late-time dominating stationary correlations for linear coupling to a squeezed thermal reservoir [83, 96] will produce the FDR kernel

$$\tilde{\kappa}_T^r(\omega) = \cosh[2r(\omega)] \omega \coth\left(\frac{\omega}{2T}\right), \quad (4.173)$$

where  $r(\omega)$  is the squeezing parameter, which may be allowed to vary with the energy scale. One can easily see that this FDR also obeys inequality (4.171) as it must.

## 4.6.2 Non-Equilibrium Uncertainty Principle

In the context of second-order perturbation theory, quantum noise is effectively Gaussian in the influence functional, and Gaussian noise is equivalent to that arising from linear coupling to a bath of harmonic oscillators. Therefore any violations of Eq. (4.168) must correspond to environment oscillators in a non-quantum state. In the phase-space or Wigner function representation [81], HUP violating states of the environment can be constructed which violate the quantum FDI. Such is the case for the classical vacuum, which has vanishing fluctuations yet finite damping. Now we shall show that FDI violating noise can also relax the system into a HUP violating state.

Let us consider weakly influencing a system of oscillators at resonance, all with mass  $M$  and frequency  $\omega$ , via position-position coupling to some phenomeno-

logical set of noise processes with resistive correlation  $\tilde{\alpha}(\omega)$ . We do not assume the system-environment couplings to be identical, nor do we neglect the presence of cross-correlations among the noise processes. From the results of Chapters 3 & 2 and applying the second-order master equation coefficients (6.18), the damping kernel  $\tilde{\gamma}(\omega)$  will play the role of the dissipation coefficients and the noise kernel  $\tilde{\nu}(\omega)$  will play the role of the normal diffusion coefficients in the Fokker-Planck or master equation. Integrals over the two kernels will then provide the system renormalization and anti-diffusion coefficients respectively. Given sufficient dissipation and bandwidth-limited correlations, the system will relax into a Gaussian state which satisfies the Lyapunov equation

$$\tilde{\nu}(\omega) = \frac{1}{2} \left[ \left( \frac{2}{M} \sigma_{PP} \right) \tilde{\gamma}(\omega) + \tilde{\gamma}(\omega) \left( \frac{2}{M} \sigma_{PP} \right) \right], \quad (4.174)$$

for the momentum covariance, which has elements  $\langle P_i P_j \rangle$ , and

$$\sigma_{XX} = \frac{1}{(M\omega)^2} \sigma_{PP}, \quad (4.175)$$

$$\sigma_{XP} = \mathbf{0}, \quad (4.176)$$

for the remaining covariances in the phase-space (Wigner function) representation [81], and to lowest order in the system-environment interaction. Comparing Eq. (4.174) to Eq. (4.170), we can express our covariances

$$\sigma_{XX} = \frac{1}{2M\omega^2} \tilde{\kappa}(\omega), \quad (4.177)$$

$$\sigma_{PP} = \frac{M}{2} \tilde{\kappa}(\omega), \quad (4.178)$$

in terms of the FDR kernel  $\tilde{\kappa}(\omega)$ . So far our FDR kernel remains phenomenological and not microscopically derived, however it must be positive definite for this to

describe a physical state. As  $\tilde{\kappa}(\omega)$  is positive definite, we may transform to the basis in which it is diagonalized. If  $\tilde{\kappa}(\omega)$  is a scalar quantity, then this is simply the normal basis of the free system. For each mode in this basis we have the phase-space covariance

$$\boldsymbol{\sigma}_n = \begin{bmatrix} \sigma_n^{XX} & \sigma_n^{XP} \\ \sigma_n^{PX} & \sigma_n^{PP} \end{bmatrix} = \begin{bmatrix} \frac{1}{2M\omega^2}\tilde{\kappa}_n(\omega) & 0 \\ 0 & \frac{M}{2}\tilde{\kappa}_n(\omega) \end{bmatrix}, \quad (4.179)$$

which must then satisfy the generalized Heisenberg uncertainty relation due to Schrödinger [132, 43]:

$$\langle \Delta X^2 \rangle \langle \Delta P^2 \rangle - \frac{1}{2} \langle \{ \Delta X, \Delta P \} \rangle \geq \frac{1}{4}, \quad (4.180)$$

or in terms of the phase-space covariance

$$\det(\boldsymbol{\sigma}) \geq \frac{1}{4} \quad (4.181)$$

and, therefore, it must be the case that

$$\tilde{\kappa}_n(\omega) \geq \omega, \quad (4.182)$$

for all  $\omega > 0$ . But this is equivalent to our previous statement

$$\tilde{\kappa}(\omega) \geq \omega, \quad (4.183)$$

in terms of positive definiteness as  $\omega$  is a scalar quantity. So not only do FDI violating correlations arise from HUP violating states, they can also produce HUP violating states via dissipation and diffusion (and decoherence).

Furthermore we can say that in the weak-damping limit, the scalar FDR kernel  $\tilde{\kappa}(\omega)$  precisely determines the (asymptotic) non-equilibrium uncertainty product

$$\det(\boldsymbol{\sigma}) = \left( \frac{1}{2} \frac{\tilde{\kappa}(\omega)}{\omega} \right)^2, \quad (4.184)$$

for a single system mode of frequency  $\omega$ . Larger FDR kernels naturally produce larger uncertainty and minimal FDR kernels (zero temperature) produce minimal uncertainty. Non-perturbative results require evaluation of the exact expressions found in Chapter 3.

## 4.7 Decoherence Strength

Environment-induced decoherence is an essential process for a quantum system to acquire classical attributes [74, 124, 164]. To characterize how strong an environment can induce decoherence in an open quantum system it is desirable to come up with a measure of the “*decoherence strength*” of each environment acting on the system. For a Markovian environment, which for quantum Brownian motion refers to high temperatures and Ohmic spectral density, such a measure can be constructed by the product of its temperature and damping rate. However, such a measure may not exist for a *general* environment (with non-Ohmic spectral density functions and under low temperature conditions, see [84, 85, 64]), or, even more challenging, for nonequilibrium environments where the notion of temperature loses meaning. We show in this section that at least perturbatively under weak coupling between the system and these environments, low-temperature and non-equilibrium environments *can* be partially ordered. The object of comparison is, in fact, the correlation function of the collective coupling operators of the environment. As with matrices and kernels, quantum correlations can only be partially ordered. However, this does not rule out nontrivial comparisons; in our final subsection we explicitly detail how one

can compare decoherence strengths for combinations of thermal reservoirs without resorting to the concocted notion of an effective temperature, as in general it does not exist. Our general relation include some recently reported results as special cases [16].

#### 4.7.1 Quantum Correlations & Decoherence Strength

The theorem of Lindblad [106] and Gorini, Kossakowski and Sudarshan [76] specifically characterizes the *algebraic generators*  $\Phi$  for all completely-positive maps  $e^{\eta\Phi}$ , where  $\eta > 0$  parameterizes the semi-group [106, 76].

$$\Phi \rho = \underbrace{-i [\Theta, \rho]}_{\text{unitary}} + \underbrace{\sum_{ij} \Delta_{ij} \left( \mathbf{e}_i \rho \mathbf{e}_j^\dagger - \frac{1}{2} \{ \mathbf{e}_j^\dagger \mathbf{e}_i, \rho \} \right)}_{\text{decoherent}}. \quad (4.185)$$

Such generators and the dynamics they engender when the master equation has the Lindblad form have been extensively studied [97, 45, 46, 48, 6, 1, 14, 90, 107, 151]. Here,  $\Delta_{ij}$ , the (algebraic) *dissipator*, is a positive-definite coefficient matrix and  $\mathbf{e}_i$  denotes a particular basis of representation for the dissipator. The “dissipation” generated by the dissipator is that of states, including decoherence. Derived in Sec. 2.2.1.3, the second-order algebraic dissipator evaluates to

$$\Delta_{ii';jj'}(t) = \sum_{nm} \int_0^t d\tau \int_0^t d\tau' \langle i | \mathbf{L}_m(\tau) | i' \rangle \alpha_{nm}(\tau', \tau) \overline{\langle j | \mathbf{L}_n(\tau') | j' \rangle}, \quad (4.186)$$

in the interaction (Dirac) picture and evaluated in some basis  $|i\rangle\langle j|$ . Expression (4.186) will be shown to be a positive-definite form for all microscopically derived noise correlations, thus agreeing with the Lindblad-GKS theorem in as much as is required. Though the Markovian generators which appear in the Lindblad equation

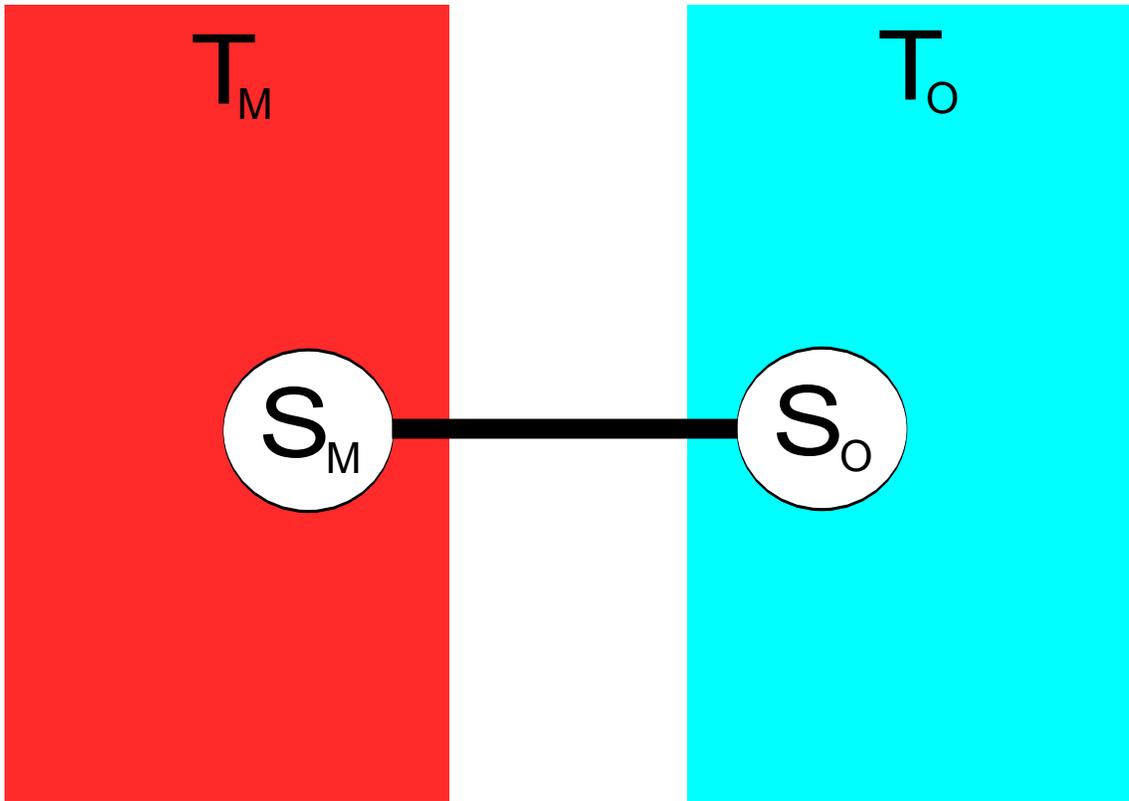


Figure 4.3: Consider, for instance, a system of nano-mechanical resonators  $S_M$  interacting with a system of optical modes  $S_O$ , wherein the optical modes experience dissipation and thermal noise  $T_O$  from the cavity field yet the resonators experience dissipation and thermal noise  $T_M$  from a phonon environment. The combined environment is non-equilibrium and, not obeying the fluctuation-dissipation relation, it is not described by a single spectral-density function and temperature. The decoherence of this multipartite system cannot be strictly argued from a temperature and dissipation coefficient.

are well known, to our knowledge these non-Markovian generators (which are strictly algebraic and do not appear in the master equation) are a novel discovery.

The key to accomplishing our stated goal rests in the comparison of perturbative dissipators. We first note that, from its microscopic origins, Eq. (2.19), the environment correlation function is Hermitian in the sense of

$$\boldsymbol{\alpha}(t, \tau) = \boldsymbol{\alpha}^\dagger(\tau, t), \quad (4.187)$$

and also positive definite in the sense of

$$\int_0^t d\tau_1 \int_0^t d\tau_2 \mathbf{f}^\dagger(\tau_1) \boldsymbol{\alpha}(\tau_1, \tau_2) \mathbf{f}(\tau_2) \geq 0, \quad (4.188)$$

for all vector functions  $\mathbf{f}(t)$  indexed by the noise. All quantum correlations are at least *nonlocally decoherent* as their algebraic Lindblad dissipator, Eq. (4.186), is necessarily positive definite. Nonlocal decoherence only implies that there is more net decoherent evolution than recoherent evolution. The more strict property of instantaneously decoherent evolution,  $\dot{\Delta}(t) > 0$ , can only be generally satisfied by *local decoherence* with delta correlations (Markovian processes) and would always produce a Lindblad master equation. However, some very restricted classes of system-environment interactions, such as the RWA-interaction Hamiltonian (see Sec. 4.4), can be constrained by their coupling to be locally decoherent. This characterizes the class of systems with non-Markovian dynamics whose master equation is, nevertheless, naturally of Lindblad form.

It is the key point of this work that the environment correlation function itself provides a natural comparison of state dissipation or *decoherence strength*.

If two correlation functions are ordered  $\alpha_+(t, \tau) > \alpha_-(t, \tau)$  in the sense of positivity (4.188), then their corresponding second-order Lindblad dissipators are also ordered  $\Delta_+(t) > \Delta_-(t)$ , and we can, therefore, say that one environment generates more state dissipation than the other, regardless of the system. For instance, the set of univariate Markov processes is totally ordered by the scalar magnitude of the respective delta correlations, e.g.  $2\delta(t - \tau) > 1\delta(t - \tau)$ . In general, the set of all quantum correlations is only partially ordered, but nontrivial orderings do exist. We illustrate this principle with a specific problem below.

## 4.7.2 Thermal Correlations

### 4.7.2.1 Individual Reservoirs

Constant coupling to a thermal reservoir will always produce environment correlations which can be expressed in the Fourier domain as

$$\tilde{\alpha}(\omega) = \tilde{\gamma}(\omega) [\tilde{\kappa}_T(\omega) - \omega] , \quad (4.189)$$

$$\tilde{\kappa}_T(\omega) \equiv \omega \coth\left(\frac{\omega}{2T}\right) , \quad (4.190)$$

in terms of the damping kernel  $\tilde{\gamma}(\omega)$  (anti-derivative of the dissipation kernel) and fluctuation-dissipation kernel  $\tilde{\kappa}_T(\omega)$ . This relation was given three derivations in Sec. 2.5.3: directly from first principles in Eq. (2.19), by demanding a coupling-invariant fluctuation-dissipation relation (FDR), and by demanding coupling-invariant detailed balance in the master equation.

Note that a Markovian quantum regime (complex white noise) necessarily implies a local damping kernel and high temperature (local FDR kernel). The

Markovian regime is reached when the system time scales are much slower than those of the environment, so that we can take the zero-frequency approximation

$$\lim_{\omega \rightarrow 0} \tilde{\alpha}(\omega) = \tilde{\gamma}(0) 2T . \quad (4.191)$$

Markovian processes can, therefore, be ordered in their decoherence strength by the product of their damping and temperature, a result which is well known. If one inquires as to the temperature of an unknown Markovian process, the FDR kernel (or noise-to-damping ratio) will always reveal this.

In general, nonlocal correlations (e.g. finite temperature) and thus decoherence strengths are not totally ordered. For a fixed temperature thermal correlations can be ordered by damping. On the other hand we have the inequality

$$\tilde{\kappa}_{\text{hot}}(\omega) > \tilde{\kappa}_{\text{cold}}(\omega) \geq |\omega| , \quad (4.192)$$

and so for fixed damping, correlations can also be ordered by their temperature. Therefore, finite-temperature thermal correlations are partially ordered by damping and temperature.

$$\tilde{\gamma}_{\text{strong}}(\omega) [\tilde{\kappa}_{\text{hot}}(\omega) - \omega] > \tilde{\gamma}_{\text{weak}}(\omega) [\tilde{\kappa}_{\text{cold}}(\omega) - \omega] , \quad (4.193)$$

and so it immediately follows that

$$\tilde{\alpha}_{\text{strong}}^{\text{hot}}(\omega) > \tilde{\alpha}_{\text{weak}}^{\text{cold}}(\omega) . \quad (4.194)$$

If one environment has a higher temperature but weaker damping, then the two correlations cannot be ordered – the implication is that different systems would decohere faster or slower for each environment, but not in a manner which can be strictly ordered.

From Eq. (4.193) we can now compare environments of low temperature and nonlocal damping. For fixed damping, the monotonic ordering of temperature is no surprise. While for fixed temperature, the ordering of damping is more subtle though also not surprising. The damping can be increased by an overall rescaling, say  $\tilde{\gamma}(\omega) \rightarrow 2\tilde{\gamma}(\omega)$ , but also by increasing the high-frequency response of the environment, say  $\Lambda \rightarrow 2\Lambda$  for any monotonic regulator (not necessarily Ohmic) with cutoff  $\Lambda$ .

#### 4.7.2.2 Multiple environments

Here we wish to duplicate the work of Beer & Lutz [16] wherein they compared the decoherence rates of collective environments with different temperatures and Ohmic cutoff frequencies, specifically for linear coupling to an oscillator and with both reservoirs at a relatively high temperature. For multiple environments we can make the same comparison by using the natural measure of decoherence strength from the Lindblad dissipator and environment correlation function. First we note that for coupling to one reservoir, the individual thermal correlations can be expressed via Eq. (6.13). Next we note that for any monotonic cutoff regulator, with fixed local limit  $\tilde{\gamma}(0)$  and variable cutoff  $\Lambda$ , then

$$\tilde{\gamma}_{\text{high}}(\omega) \geq \tilde{\gamma}_{\text{low}}(\omega), \quad (4.195)$$

where  $\Lambda_{\text{high}} > \Lambda_{\text{low}}$  (referred to as “fast” and “slow” in Ref. [16]). We can also compare the individual FDR kernels as per Eq. (4.192). Finally we can use the

above relations construct the mathematical inequality

$$[\tilde{\gamma}_{\text{high}}(\omega) - \tilde{\gamma}_{\text{low}}(\omega)]\{[\tilde{\kappa}_{\text{hot}}(\omega) - \omega] - [\tilde{\kappa}_{\text{cold}}(\omega) - \omega]\} > 0, \quad (4.196)$$

which can then be rearranged to show that

$$\tilde{\alpha}_{\text{high}}^{\text{hot}}(\omega) + \tilde{\alpha}_{\text{low}}^{\text{cold}}(\omega) > \tilde{\alpha}_{\text{low}}^{\text{hot}}(\omega) + \tilde{\alpha}_{\text{high}}^{\text{cold}}(\omega), \quad (4.197)$$

which is consistent with the results of Ref. [16], when we interpret the left and right-hand sides of Eq. (4.197) as comparing the decoherence strengths of two different collective (non-equilibrium) environments. Note that as the individual reservoirs are Gaussian and independent, one may simply add their correlations in determining the collective correlation. Our result applies more generally than that of Ref. [16], in terms of coupling and temperature, though we do not calculate a specific decoherence time. Their work relied upon what is essentially the exact FDR kernel, but which has been referred to as an *effective temperature* [40] in the classical regime. We would rather avoid this nomenclature given that such environments will lead to an asymptotic stationary state which is not thermal in general (sufficiently simple systems can reach a thermal state, but the corresponding temperature will be different for different systems).

## 4.8 Discussion

In Sec. 4.2 we have shown that even when provided with a stationary master equation describing dynamics that are amenable to perturbative solution, the solutions to an order- $2n$  perturbative master equation are, in general, only accurate

to order- $(2n-2)$ , a step down from that of the master equation itself. This has a wide-range of implications upon the common use of second-order master equations and related master equations derived from second-order dynamics: the Redfield, Born-Markov, and many Lindblad equations. Moreover, not even a nonlocal representation, such as with the Nakajima-Zwanzig master equation can avoid this effect. This is to be expected as a thorough analysis of time-local and nonlocal dynamics shows their asymptotics to be perturbatively the same [63].

To be more specific, the second-order master equation can provide all second-order timescales and off-diagonal density matrix elements (in the free energy basis). However it can only provide the diagonal matrix elements with zeroth-order accuracy, and the missing information is the most relevant to positivity in the low-temperature regime. Therefore the second-order master equation can produce second-order positivity violations, whereas the full second-order solutions are positive to second-order. Likewise, the steady state of the second-order master equation may only agree with the steady state of the full master equation to zeroth order. More generally, the predicted expectation of observables will typically be off by a second-order amount, and this certainly includes the energy or other quantities that were conserved at zeroth order. We have shown that this inaccuracy manifests itself in the case of quantum Brownian motion through an underestimation of the position uncertainty stationary limit. The same issue also affects the predicted dynamics for a collection of atoms interacting with a shared field [61], where the complete second-order solution is required make correct predictions about the sudden death of entanglement.

There are three mathematical limits in which the second-order master equation will give solutions accurate to second order: The first is early times, where  $t$  is small compared to any of the second-order damping time scales. The second is the Markovian limit, because in this limit the second-order master equation is exact. The third is the limit employed by Davies [45] where one rewrites the master equation in terms of the rescaled time parameter  $\tau = g^2 t$  and then takes the limit  $g \rightarrow 0$  (for  $\tau \neq 0$  this effectively amounts to taking a simultaneous  $t \rightarrow \infty$  limit). In this limit all corrections to the eigenoperators of the Liouvillian vanish, and the only effect of the environment is to introduce damping rates through corrections to the eigenvalues, which are correctly captured by the perturbative master equation. Thus, the inaccuracies of second-order master equation we have addressed may be sufficiently suppressed even at late times if a physical system is sufficiently close to being described by one of these limits. Therefore our results should be most heeded in the non-Markovian regime of low temperature or long-ranged correlations in contexts where  $\mathcal{O}(g^2)$  discrepancies are not negligible.

In Sec. 4.4 we have systematically examined the rotating-wave approximation by using a master equation for an open quantum system weakly-coupled to a general environment. There are, in fact, two distinct rotating-wave approximations: The pre-trace RWA is an approximation performed on the interaction Hamiltonian before the environment is traced out which yields a somewhat modified Hamiltonian dynamics from which the reduced dynamics can be derived. The post-trace RWA is performed on the master equation for the reduced density matrix after the environment has already been traced out. Using the general framework of Chap-

ter 2 we have compared the master equation describing the dynamics of this open system under the pre and post-trace RWA to the dynamics without making such an approximation. We have specifically addressed the master equations for a two-level system and for a linear oscillator, two models in which the RWA is often invoked.

We find the post-trace RWA to be more innocuous than the pre-trace RWA. It can be seen as an often reasonable approximation in which the full weak-coupling Liouvillian (which is time-local and of pseudo-Lindblad form) is projected onto a Lindblad-form Liouvillian. We call the resulting master equation the RWA-Lindblad equation. We find that for a general open quantum system the post-trace RWA will yield exactly the same timescales as perturbative solutions of the weak-coupling master equation. The perturbative corrections to eigen-operators of the Liouvillian are neglected in the RWA-Lindblad equation, so the predicted quantum state will differ. In particular, the steady state solution of the RWA-Lindblad master equation will differ from the true steady state by an amount that is perturbative in the coupling. These results are consistent with what Agarwal found [5, 3, 4] for the two-level atom and the linear oscillator in the Born-Markov approximation. One context in which the discrepancy in the steady state could be important is examining the late-time behaviour of entanglement dynamics at low temperature. When the system is bipartite and the ground state is separable, the RWA-Lindblad equation will give an asymptotically separable state, whereas a more complete master equation leaves open the possibility of asymptotic entanglement. In view of our findings, we can say that generally the post-trace RWA is suitable if one only wants the perturbative timescales of the dynamics, but it may not be appropriate if one wants

more detailed information about the quantum state of the system, as it misses some corrections introduced by the coupling to the environment, and it will also not be appropriate when perturbative timescales fail, i.e. for near-resonance in the energy level splittings.

We find the pre-trace RWA to be more problematic. When the environment contains many frequencies with a spread comparable to the frequencies of the system, then the pre-trace RWA in general does not provide a faithful representation of the true solution. We also find that the pre-trace RWA results in two strongly correlated sources of environmental noise that together have no Markovian limit. The cross-correlations between the noise sources are such that if the autocorrelations are white then the cross-correlations are strongly coloured. This issue has been noticed before [152]. Finally, we have shown that, unlike the post-trace RWA, the pre-trace RWA in general does not correctly obtain all perturbative timescales for the dynamics, yielding incorrect frequency shifts. This finding based on a more extended theoretical framework agrees with results obtained for specific cases studied before for the two-level atom [5, 3, 4].

We caution that the way the RWA is applied also matters. For Markovian processes in closed and open systems, certain liberties can be taken with the master equation; terms can be mixed and matched rather haphazardly. A Markovian process will generally produce a master equation of Markovian representation and Lindblad form. The RWA can also produce a master equation of Markovian representation and Lindblad form, and therefore one might assume that these liberties can also be taken with the RWA-Lindblad master equation. But this is not the case as

the underlying stochastic process remains non-Markovian and the master equation coefficients contain memory despite their Markovian representation. A haphazard construction of RWA-Lindblad master equations for multipartite systems, or those with external forcings, can produce an evolution which is completely positive and yet totally unphysical.

The pre-trace RWA as applied to quantum Brownian motion is additionally interesting because the interaction Hamiltonian in this case coincides with the Gardiner-Collett Hamiltonian [72] used to model the coupling between the intracavity field of a high-finesse electromagnetic cavity and external field modes. However, in this case the form of the Hamiltonian is not the result of a rotating-wave approximation and can be derived [54] from a “modes-of-the-universe” approach for a cavity with a partially transmitting mirror in the limit that transmission is weak. So in this case the pre-trace RWA Hamiltonian corresponds to the physical Hamiltonian of an actual system, and the solutions of the master equation have relevance directly, rather than as an approximation.

A single two-level atom is clearly a particularly simple quantum system. As such, some of the shortcomings of the RWA are obscured in this case. We find that the post-trace RWA gives the correct equilibrium state for a thermal environment in this case, in addition to the correct timescales, though it does miss some of the corrections to the transient quantum evolution that can be obtained from the weak-coupling master equation without the RWA. Thus, if one’s theoretical investigations are limited to those features that it captures correctly, then the post-trace RWA may be a suitable approach.

Our general analysis provides a reasonably good characterization of the validity of the RWA for constructing a master equation; however, many questions remain about its effects on the internal dynamics of more complex systems, such as the effect of either form of RWA on causality in multi-atom models with spatial separation [52] and its effect on entanglement dynamics [10].

In Sec. 4.5 we have considered the true equilibrium state of the system and of the system + environment. As we have shown, the master equation and Schrödinger equation both agree that the reduced equilibrium state of the system + environment is the asymptotic state of the system. This does not necessarily imply that the system + environment evolves into equilibrium, but only the coarse-grained open system. One must also consider that the infinite and continuum limit of the environment does not commute with many other limits. This is the key ingredient which leads to irreversible dissipation and decoherence in the system. The system + environment could be evolving coherently, and yet the open system appear to reach global equilibrium.

In Sec. 4.6 we have derived a fluctuation-dissipation inequality (FDI) for an open quantum system interacting with a non-equilibrium environment from the microscopically-derived environment correlation function and recovered the well-known fluctuation-dissipation relation (FDR) for a thermal environment. The FDI is a very general statement contrasting quantum noise to classical noise, and is satisfied even for non-equilibrium fluctuations. Simply put, quantum fluctuations are bounded below by quantum dissipation, whereas classically the fluctuations can be made to vanish. The lower bound of this inequality is exactly satisfied by

zero-temperature noise and is in accord with the Heisenberg uncertainty principle (HUP). FDI violating correlations arise from HUP violating states of the environment and can relax the open system into HUP violating states. Therefore, the FDI can be viewed as an open-system corollary to the HUP both from microscopic and phenomenological considerations. Analogously, the non-equilibrium FDR also determines the non-equilibrium uncertainty product, most directly in the limit of weak-damping [cf. Eq. (4.184)], and the corresponding FDI implies the HUP.

Finally, in Sec. 4.7, we have motivated a general notion of decoherence strength as that generated by the quantum correlations of the environment, which in turn determine the magnitude of the algebraic Lindblad dissipator and thus state dissipation itself. The ordering of decoherence strengths in this formalism is only partial, though when it occurs it is system and state independent. However, this is not to say that comparison of environment correlations is not as useful when there is no strict ordering. State-dependent decoherence is of particular interest in the search for decoherence-free states and how they emerge in certain classes of environments. If for two environments  $\tilde{\alpha}_1(\omega) - \tilde{\alpha}_2(\omega)$  is indefinite, then there *could* be states corresponding to the vectors  $\mathbf{f}$  in Eq. (4.188) which exploit this. As an explicit example, if two atoms are brought close together in a quantum field, then rapidly and slowly decaying states emerge which mirror the behavior of the eigenvalues of the correlation function (see Sec. 6.3).

## Chapter 5

### Strong-Coupling Perturbation

#### 5.1 Introduction

In Chapter 2 we worked from the prevailing perspective of weak-coupling perturbation, wherein the system-environment interaction energy was considered to be relatively weak. At the other end of the spectrum, there is also great interest in the strong-coupling regime. Naive expectations of behavior in this regime appeal to the limit in which the system Hamiltonian is taken to be vanishing and the system is primarily driven by the system-environment interaction. For a single (collective) coupling to the environment, or univariate noise, this produces a rapid decoherence in the basis of the system coupling operator. Ergo, for a strong position coupling to the environment, it is thought that the system should decohere into a position distribution. However, this limit has questionable applicability to small mechanical systems. Large noise fluctuations can generate large kinetic energies, which are then incompatible with the limit of vanishing system energy.

In this chapter we derive from first principles the dynamics of a quantum or classical system primarily driven by noise and dissipation. What we take to be perturbative is not the entire system Hamiltonian as with the “strong-coupling” *quantum Smoluchowski equation* [13, 38], but only the system potential, or (anharmonic) terms beyond quadratic order. The result is a class of master equations

which describe highly non-Markovian dynamics and seem to admit no generic form. Instead we have a procedure for generating said master equations through an iterative application of noise averaging and integration. The two critical assumptions in this work are that the influence of the environment is Gaussian (e.g. linear coupling to a bath of harmonic oscillators) and that system-environment coupling is moreover bilinear. For simplicity we will assume the Hamiltonian we perturb from to be time-independent.

Thus far, the results therein are semi-formal. We have obtained definite expressions for the master-equation coefficients, but these expressions are quite complicated and a fair amount of work remains in their application.

## 5.2 Linear Systems

Let us review the QBM Langevin-equation solutions from Chapter 3. Given system phase-space coordinates  $\mathbf{Z} = (\mathbf{X}, \mathbf{P})$  and environment phase-space coordinates  $\mathbf{z} = (\mathbf{x}, \mathbf{p})$ , the system solutions can be expressed

$$\mathbf{Z}(t) = \mathbf{\Phi}(t) \mathbf{Z}(0) + \mathbf{\Phi}(t) * \underbrace{\sum_k \mathbf{c}_k \mathbf{\Phi}_k(t) \mathbf{z}_k(0)}_{\mathbf{\Xi}(t)}, \quad (5.1)$$

where  $\mathbf{\Phi}(t)$  is the homogeneous propagator of the open system and where we have opted to represent the noise process directly in terms of its free propagators  $f_k(t)$

and system-environment couplings  $c_k$ , related by

$$\boldsymbol{\Phi}_k(t) \equiv \begin{bmatrix} m_k \dot{f}_k(t) & f_k(t) \\ m_k^2 \ddot{f}_k(t) & m \dot{f}_k(t) \end{bmatrix}, \quad (5.2)$$

$$\mathbf{c}_k \equiv \begin{bmatrix} 0 & 0 \\ 0 & c_k \end{bmatrix}. \quad (5.3)$$

Recall specifically that these solutions correspond to the characteristic curves (or classical-like trajectories) of the system + environment evolution while in the phase-space representation. Analogously, there are also solutions for  $\mathbf{z}(t)$  in terms of  $\mathbf{Z}(0)$  and  $\mathbf{z}(0)$ , however we will not need such relations.

The master equation which governs the evolution of the reduced Wigner function  $W(\mathbf{Z}, t)$  is given by

$$\frac{d}{dt} W(\mathbf{Z}, t) = \mathcal{L}(t) W(\mathbf{Z}, t), \quad (5.4)$$

$$\mathcal{L}(t) \equiv \nabla_{\mathbf{Z}}^T \mathcal{H}(t) \mathbf{Z} + \nabla_{\mathbf{Z}}^T \mathbf{D}(t) \nabla_{\mathbf{Z}}, \quad (5.5)$$

and takes the form of a classical Fokker-Planck equation, except that the diffusion coefficients describe quantum fluctuations. The time-local homogeneous and diffusion coefficients are given by

$$\mathcal{H}(t) = -\dot{\boldsymbol{\Phi}}(t) \boldsymbol{\Phi}^{-1}(t), \quad (5.6)$$

$$\mathbf{D}(t) = \frac{1}{2} \{ \mathcal{H}(t) \boldsymbol{\sigma}_T(t) + \boldsymbol{\sigma}_T(t) \mathcal{H}^T(t) + \dot{\boldsymbol{\sigma}}_T(t) \}, \quad (5.7)$$

where for time-constant Hamiltonians the thermal covariance is then given by

$$\boldsymbol{\sigma}_T(t) \equiv \int_0^t d\tau_1 \int_0^t d\tau_2 \boldsymbol{\Phi}(t-\tau_1) \boldsymbol{\nu}(\tau_1, \tau_2) \boldsymbol{\Phi}^T(t-\tau_2). \quad (5.8)$$

This is a special case of the two-time thermal covariance

$$\boldsymbol{\sigma}_T(t_1, t_2) \equiv \int_0^{t_1} d\tau_1 \int_0^{t_2} d\tau_2 \boldsymbol{\Phi}(t_1 - \tau_1) \boldsymbol{\nu}(\tau_1, \tau_2) \boldsymbol{\Phi}^T(t_2 - \tau_2), \quad (5.9)$$

which is the thermal contribution to the two-time correlation function

$$\langle \mathbf{Z}(t_1) \mathbf{Z}^T(t_2) \rangle_{\Xi} = \boldsymbol{\Phi}(t_1) \boldsymbol{\sigma}_0 \boldsymbol{\Phi}^T(t_2) + \boldsymbol{\sigma}_T(t_1, t_2), \quad (5.10)$$

given the initial correlation  $\boldsymbol{\sigma}_0 \equiv \langle \mathbf{Z}(0) \mathbf{Z}^T(0) \rangle$ . The first contribution is thus homogeneous and dissipative.

### 5.3 Dynamics in the Characteristics Picture

We now examine the quantum (or classical) dynamics of the combined system + environment generated by a Hamiltonian with bilinear coupling to the environment. In the phase-space representation, the Wigner function  $W$  (or density function  $\rho$ ) obeys

$$\frac{\partial}{\partial t} W[\mathbf{Z}, \mathbf{z}, t] = \mathcal{L}[\boldsymbol{\nabla}_{\mathbf{Z}}, \boldsymbol{\nabla}_{\mathbf{z}}, \mathbf{Z}, \mathbf{z}, t] W[\mathbf{Z}, \mathbf{z}, t], \quad (5.11)$$

where  $\mathcal{L}$  is the closed system + environment Liouville operator, and appears completely classical. Let us denote the combined system + environment coordinates  $\mathbb{Z} = (\mathbf{Z}, \mathbf{z})$ , so that we may state more succinctly

$$\frac{\partial}{\partial t} W[\mathbb{Z}, t] = \mathcal{L}[\boldsymbol{\nabla}_{\mathbb{Z}}, \mathbb{Z}, t] W[\mathbb{Z}, t], \quad (5.12)$$

Let us inspect the dynamics of quantum states along some curves  $\mathbb{Z}(t)$  which

are not assumed to be proper characteristics.

$$\frac{d}{dt}W[\mathbb{Z}(t), t] = \delta\mathcal{L}(t)W[\mathbb{Z}(t), t], \quad (5.13)$$

$$\delta\mathcal{L}(t) \equiv \left\{ \dot{\mathbb{Z}}(t)^T \nabla_{\mathbb{Z}(t)} + \mathcal{L}[\nabla_{\mathbb{Z}(t)}, \mathbb{Z}(t), t] \right\}. \quad (5.14)$$

The key point is that we will utilize curves  $\mathbb{Z}(t)$  such that  $\delta\mathcal{L}(t)$  is ordinarily a *system* operator and then calculate the corresponding perturbative open-system dynamics for the marginal distribution  $W(\mathbf{Z}, t)$ , which is equivalent to the reduced density matrix.

Any Gaussian influence acting on the system can be modeled via linear coupling to a linear environment, and the dynamical contributions of all such terms are first-order (dynamically classical) in the phase-space representation. Therefore we can always transform to a classically-evolving coordinate system, along which the dynamics of the environment are effectively integrated out, very similar to what is done in the Langevin equation. Moreover, the system Hamiltonian used for the classical curves is irrelevant for the specific purpose of integrating out the environment dynamics. We do not have to use the true system Hamiltonian for our characteristics, but for the purposes of perturbation theory we do want  $\delta\mathcal{L}(t)$  to be small. Therefore we generally want the classical (or linear) dynamics of these curves to reproduce as much of the quantum dynamics as possible, allowing for much cancellation in Eq. 5.14. Essentially, the noise average of Eq. 2.10 is the generalization of the stochastic description of QBM found in Chapter 3 to nonlinear forces. Whereas for linear systems the stochastic trajectories  $\mathbf{Z}(t)$  immediately provide the solution, here they only serve as an evolving coordinate system which encapsulates all effects

of the environment.

### 5.3.1 Perturbation Along Characteristics

The integral equation of motion corresponding to Eq. 5.13 is

$$W[\mathbb{Z}(t), t] = W[\mathbb{Z}(0), 0] + \int_0^t d\tau \delta\mathcal{L}[\nabla_{\mathbb{Z}(\tau)}, \mathbb{Z}(\tau), \tau] W[\mathbb{Z}(\tau), \tau], \quad (5.15)$$

which is amenable to perturbation via a Neumann series yielding the perturbative solutions

$$W_0[\mathbb{Z}(t), t] = W[\mathbb{Z}(0), 0], \quad (5.16)$$

$$W_1[\mathbb{Z}(t), t] = \int_0^t d\tau \delta\mathcal{L}[\nabla_{\mathbb{Z}(\tau)}, \mathbb{Z}(\tau), \tau] W[\mathbb{Z}(0), 0], \quad (5.17)$$

and so on, where  $W[\mathbb{Z}, t] = \sum_{n=0}^{\infty} W_n[\mathbb{Z}, t]$  and  $W_n[\mathbb{Z}, t] = \mathcal{O}(\delta\mathcal{L}^n)$ . Then we can apply the classical propagator (see Sec. 3.3.1) to transform back to the (initial) domain coordinates.

$$W_n[\mathbb{Z}(0), t] = \mathcal{G}_C(t, 0) W_n[\mathbb{Z}(t), t]. \quad (5.18)$$

Next we assume the initial state of the system + environment to be an uncorrelated product of marginal distributions,  $W(\mathbb{Z}, 0) = W(\mathbf{Z}, 0) W(\mathbf{z}, 0)$ . Upon tracing over the environment we obtain a perturbative expansion of the open-system propagator

$$\mathcal{G}(t) = \sum_{n=0}^{\infty} \mathcal{G}_n(t) : W(\mathbb{Z}, 0) \rightarrow W(\mathbb{Z}, t), \quad (5.19)$$

with the first two terms given by

$$\mathcal{G}_0(t) = \langle \mathcal{G}_C(t, 0) \rangle_{\mathbb{E}}, \quad (5.20)$$

$$\mathcal{G}_1(t) = \int_0^t d\tau \underline{\delta\mathcal{L}}(\tau, t) \mathcal{G}_0(t), \quad (5.21)$$

given the two-time open-system operator

$$\underline{\delta\mathcal{L}}(\tau, t) \equiv \langle \mathcal{G}_C(t, \tau) \delta\mathcal{L}(\tau) \mathcal{G}_C(\tau, 0) \rangle_E \langle \mathcal{G}_C(t, 0) \rangle_E^{-1}, \quad (5.22)$$

where the single-time closed-system operator

$$\delta\mathcal{L}(t) = \delta\mathcal{L}[\nabla_{\mathbf{Z}}, \mathbf{Z}, t] = \delta\mathcal{L}[\nabla_{\mathbf{Z}}, \mathbf{Z}, t], \quad (5.23)$$

is always taken to be exclusively a system operation. This formal result should encapsulate that of Chen, Lebowitz, and Liverani [34], who applied (anharmonic) perturbation theory from the influence-functional formalism.

In weak-coupling perturbation theory, perturbative solutions of this form are terribly secular in time. More generally, they do not respect Lie-group symmetries such as unitary or completely-positivity. To remedy both issues we can use this expansion to obtain the perturbative master equation. Using ordinary perturbation theory, our open-system master equation is then

$$\mathcal{L}_0(t) = \left[ \frac{d}{dt} \langle \mathcal{G}_C(t, 0) \rangle_E \right] \langle \mathcal{G}_C(t, 0) \rangle_E^{-1}, \quad (5.24)$$

$$\mathcal{L}_1(t) = \delta\mathcal{L}(t) + \int_0^t d\tau \left\{ \frac{d}{dt} - \text{Ad}[\mathcal{L}_0(t)] \right\} \underline{\delta\mathcal{L}}(\tau, t), \quad (5.25)$$

and so on, where  $\mathcal{L}(t) = \sum_{n=0}^{\infty} \mathcal{L}_n(t)$  is the open-system Liouvillian or time-evolution generator, such that

$$\frac{\partial}{\partial t} W[\mathbf{Z}, t] = \mathcal{L}(t) W[\mathbf{Z}, t]. \quad (5.26)$$

The lowest-order propagator  $\mathcal{G}_0(t)$ , given by Eq. 5.20, is the evolution operator for the classical Brownian motion problem, but with quantum noise, making

it potentially exact for linear systems.  $\mathcal{L}_0(t)$ , given by Eq. 5.24, is the corresponding Fokker-Plank equation but with quantum noise, e.g. the HPZ equation [84]. Essentially, if one can solve the classical Brownian motion problem (with quantum fluctuations) then one can determine the quantum corrections due to the combination of nonlinear forces and noise. If one is in the classical, linear, Markovian or noiseless regime, then all perturbative corrections can vanish in this formalism given the appropriate characteristics to perturb from.

Our hierarchy of approximation schemes is therefore not only determined by the order to which we calculate the master equation, the  $n$  of  $\mathcal{L}_n$ , but also the amount of detail about our system captured in the characteristics we use. Perturbing off the damped (but otherwise) free characteristics is strong-damping perturbation as  $\mathcal{L}_n$  is  $\mathcal{O}(V^n)$ . Perturbing off of the damped oscillator characteristics is a kind of quasi-linear feedback approximation. And perturbing off of the full classical characteristics is a kind of semi-classical feedback approximation. At least formally, one might also imagine an extension beyond the classical characteristics into the quantum characteristics [99], possibly with an additional (partial) semi-classical expansion therein. Each class of characteristics encompasses the previous.

Note that the system operation  $\delta\mathcal{L}(t)$  is fully retained in the first-order correction  $\mathcal{L}_1(t)$ ; only its effect through the environment (back-action or feedback) is approximate. To zeroth order, the environment perceives the system evolution as being linear or classical and its feedback upon the environment is therefore approximate in that manner. At first order we pick up the full nonlinearity of the system forces in  $\delta\mathcal{L}(t)$  along with feedback corrections from the time integral in Eq. 5.25.

Essentially  $\underline{\delta\mathcal{L}}(\tau, t)$  is a two-time and open-system generalization of  $\delta\mathcal{L}(t)$ , such that  $\underline{\delta\mathcal{L}}(t, t) = \delta\mathcal{L}(t)$  and  $\underline{\delta\mathcal{L}}(0, t) = \mathcal{G}_0(t) \delta\mathcal{L}(t) \mathcal{G}_0^{-1}(t)$ , which is integrated over the past in order to compensate for what is missed in the lower-order approximation.

This is an analogous formalism to the more common weak-coupling perturbation wherein the system and environment are uncoupled at zeroth order. In weak-damping perturbation the higher-order generators involve integrals over the past which contain the free propagators acting upon the system-environment interaction. Here in strong-damping perturbation the higher-order generators involve integrals over the past which contain the approximate (yet interacting) propagators acting upon the nonlinear system forces.

## 5.4 Linear Back-Action

Perturbing off a linear system + environment is the most straightforward application of this formalism. This includes both the quasi-linear feedback and strong-coupling regimes. In evaluating the first-order Liouville operator in Eq. 5.25, we aim to calculate the two-time and open-system operator

$$\underline{\delta\mathcal{L}}(\tau, t) = \langle \mathcal{G}_C(t, \tau) \delta\mathcal{L}(\tau) \mathcal{G}_C(\tau, 0) \rangle_E \langle \mathcal{G}_C(t, 0) \rangle_E^{-1}, \quad (5.27)$$

for all ordinary system operators  $\delta\mathcal{L}(t)$ . This turns out to be a complicated procedure and therefore there will be no simple strong-coupling master equation which we can write down for an arbitrary system potential. Instead one must apply the following procedures iteratively and work out the dynamics for specific system potentials.

## 5.4.1 Evaluation of Two-Time Open-System Operators

### 5.4.1.1 Transformation of Derivatives

As a simple example, let us consider the reduced (linear) derivative operator

$$\delta\mathcal{L} = \nabla_{\mathbf{Z}}^T, \quad (5.28)$$

$$\underline{\delta\mathcal{L}}(\tau, t) = \langle \mathcal{G}_C(t, \tau) \nabla_{\mathbf{Z}}^T \mathcal{G}_C(\tau, 0) \rangle_{\mathbb{E}} \langle \mathcal{G}_C(t, 0) \rangle_{\mathbb{E}}^{-1}. \quad (5.29)$$

First we move all system derivatives to the left by performing the transformation

$$\underline{\delta\mathcal{L}}(\tau, t) = \langle [\mathcal{G}_C(t, \tau) \nabla_{\mathbf{Z}}^T \mathcal{G}_C(\tau, t)] \mathcal{G}_C(t, 0) \rangle_{\mathbb{E}} \langle \mathcal{G}_C(t, 0) \rangle_{\mathbb{E}}^{-1}. \quad (5.30)$$

System derivatives transform with the transpose of the system + environment propagator

$$\nabla_{\mathbf{Z}}^T \rightarrow \underbrace{\nabla_{\mathbf{Z}}^T \Phi(t-\tau)}_{\text{system}} + \sum_k \underbrace{\nabla_{\mathbf{Z}_k}^T [\Phi * \mathbf{c}_k \Phi_k](t-\tau)}_{\text{environment}}, \quad (5.31)$$

where we have used the system portion of the full system + environment propagation detailed in Eq. (5.1). Homogeneous system derivatives pass through the noise average, whereas noise derivatives will be shown to trivially vanish in the noise average. Integrating over environment derivatives results in only boundary terms which should limit to zero. E.g.

$$\int_{-\infty}^{+\infty} dx_k \frac{\partial}{\partial x_k} W(x_k) = W(x_k)|_{-\infty}^{+\infty} = 0. \quad (5.32)$$

Therefore our reduced derivative is simply

$$\underline{\delta\mathcal{L}}(\tau, t) = \nabla_{\mathbf{Z}}^T \Phi(t-\tau), \quad (5.33)$$

when left of all other operations.

### 5.4.1.2 Transformation of Coordinates

As a simple example, let us consider the reduced (linear) coordinate operator

$$\delta\mathcal{L} = \mathbf{Z}, \quad (5.34)$$

$$\underline{\delta\mathcal{L}}(\tau, t) = \langle \mathcal{G}_C(t, \tau) \mathbf{Z} \mathcal{G}_C(\tau, 0) \rangle_E \langle \mathcal{G}_C(t, 0) \rangle_E^{-1}. \quad (5.35)$$

First we move our system coordinates to the right by considering the transformation

$$\underline{\delta\mathcal{L}}(\tau, t) = \langle \mathcal{G}_C(t, 0) [\mathcal{G}_C(0, \tau) \mathbf{Z} \mathcal{G}_C(\tau, 0)] \rangle_E \langle \mathcal{G}_C(t, 0) \rangle_E^{-1}. \quad (5.36)$$

System coordinates transform with the inverse of the propagator and so we have

$$\underline{\delta\mathcal{L}}(\tau, t) = \langle \mathcal{G}_C(t, 0) [\Phi(\tau) \mathbf{Z} + \Phi(\tau) * \Xi(\tau)] \rangle_{\Xi} \langle \mathcal{G}_C(t, 0) \rangle_{\Xi}^{-1}. \quad (5.37)$$

in the stochastic representation. Homogeneous system coordinates can pass through the noise average, but then exist sandwiched between open-system propagators. To simplify the homogeneous part, we construct the generic identity

$$\mathbf{A} \mathbf{Z} = \mathbf{A} \mathbf{Z} \langle \mathcal{G}_C(t, 0) \rangle_{\Xi} \langle \mathcal{G}_C(t, 0) \rangle_{\Xi}^{-1}, \quad (5.38)$$

which evaluates to

$$\langle \mathcal{G}_C(t, 0) [\mathbf{A} \Phi(t) \mathbf{Z} + \mathbf{A} \Phi(t) * \Xi(t)] \rangle_{\Xi} \langle \mathcal{G}_C(t, 0) \rangle_{\Xi}^{-1}, \quad (5.39)$$

and then we choose  $\mathbf{A} = \Phi(\tau, t)$  so that the homogeneous part of Eq. 5.39 matches with the homogeneous part of Eq. 5.37. Therefore we now have

$$\begin{aligned} \underline{\delta\mathcal{L}}(\tau, t) &= \Phi(\tau, t) \mathbf{Z} - \langle \mathcal{G}_C(t, 0) \Phi(\tau, t) \Phi(t) * \Xi(t) \rangle_{\Xi} \langle \mathcal{G}_C(t, 0) \rangle_{\Xi}^{-1} \\ &+ \langle \mathcal{G}_C(t, 0) \Phi(\tau) * \Xi(\tau) \rangle_{\Xi} \langle \mathcal{G}_C(t, 0) \rangle_{\Xi}^{-1}. \end{aligned} \quad (5.40)$$

This expression can also be written

$$\underline{\delta\mathcal{L}}(\tau, t) = \Phi(\tau, t) \mathbf{Z} + \left\langle \mathcal{G}_C(t, 0) \int_0^t d\tau' \Phi_f(\tau, \tau') \Xi(\tau') \right\rangle_{\Xi} \langle \mathcal{G}_C(t, 0) \rangle_{\Xi}^{-1}, \quad (5.41)$$

where the  $\Phi_f(\tau, \tau')$  is the final-value propagator (see Sec. 3.4.2.1), and is given by

$$\Phi_f(\tau, \tau') \equiv \theta(\tau - \tau') \Phi(\tau - \tau') - \Phi(\tau, t) \Phi(t - \tau'). \quad (5.42)$$

This would be the *advanced propagator* for local dissipation

$$\Phi_{\text{adv}}(\tau, \tau') \equiv -\theta(\tau' - \tau) \Phi(\tau - \tau'), \quad (5.43)$$

but for nonlocal dissipation no such propagator exists.

### 5.4.1.3 Noise Averages

Tracing over environment coordinates will yield moments of the noise. Most easily, the environment coordinates can be identified with the stochastic process  $\Xi(t)$ , a Gaussian noise process with noise correlation  $\langle \Xi(t) \Xi^T(\tau) \rangle_{\Xi} = \boldsymbol{\nu}(t, \tau)$ . Therefore we can evaluate our noise averages with the help of Novikov's formula (functional integration by parts), e.g.

$$\langle \mathcal{G}_C(t, 0) \Xi(\tau') \rangle_{\Xi} = - \int_0^t d\tau'' \left\langle \mathcal{G}_C(t, 0) \boldsymbol{\nu}(\tau', \tau'') \left[ \frac{\delta \mathbf{Z}(t)}{\delta \Xi(\tau'')} \right]^T \nabla_{\mathbf{Z}(t)} \right\rangle_{\Xi}, \quad (5.44)$$

where from Eq. 5.1, the functional derivative must be

$$\left[ \frac{\delta \mathbf{Z}(t)}{\delta \Xi(\tau'')} \right] = \Phi(t - \tau''), \quad (5.45)$$

and so for calculating 5.44 we have

$$\langle \mathcal{G}_C(t, 0) \Xi(\tau') \rangle_{\Xi} = - \int_0^t d\tau'' \boldsymbol{\nu}(\tau', \tau'') \Phi^T(t - \tau'') \nabla_{\mathbf{Z}} \langle \mathcal{G}_C(t, 0) \rangle_{\Xi}, \quad (5.46)$$

upon substitution and transforming the derivative through the propagator. All together the co-rotating coordinate becomes

$$\underline{\delta\mathcal{L}}(\tau, t) = \Phi(\tau, t) \mathbf{Z} - \int_0^\tau d\tau' \int_0^\tau d\tau'' \Phi_f(\tau, \tau') \nu(\tau', \tau'') \Phi^T(t - \tau'') \nabla_{\mathbf{Z}}, \quad (5.47)$$

which can be simplified to

$$\underline{\delta\mathcal{L}}(\tau, t) = \Phi(\tau, t) \mathbf{Z} + [\Phi(\tau, t) \sigma_T(t, t) - \sigma_T(\tau, t)] \nabla_{\mathbf{Z}}, \quad (5.48)$$

where  $\sigma_T(\tau, t)$  is the two-time thermal covariance in Eq. 5.9. As our noise is Gaussian, all operations can be reduced to integrals over the homogeneous propagator and two-time thermal covariance.

## 5.4.2 Consistent Results

### 5.4.2.1 External Forcing of an Oscillator

Consider the quantum damped oscillator with exact master equation given by Eq. 5.5 under the perturbation of an external force

$$\delta\mathcal{L}(t) = -\nabla_{\mathbf{Z}}^T \mathbf{F}(t), \quad (5.49)$$

in the microscopic theory. It demonstrated in Sec. 3.4.4 that this is not the force one finds in the open-system master equation. In evaluating the first-order correction, Eq. 5.25, we must invoke our rules for transforming system derivatives. Here we can specifically use Eq. 5.33 to obtain

$$\underline{\delta\mathcal{L}}(\tau, t) = -\nabla_{\mathbf{Z}}^T \Phi(t - \tau) \mathbf{F}(\tau), \quad (5.50)$$

$$\mathcal{L}_1(t) = \delta\mathcal{L}(t) - \int_0^t d\tau \left\{ \frac{d}{dt} - \text{Ad}[\mathcal{L}_0(t)] \right\} \nabla_{\mathbf{Z}}^T \Phi(t - \tau) \mathbf{F}(\tau). \quad (5.51)$$

The diffusion generator in  $\mathcal{L}_0(t)$  in Eq. 5.5 commutes with the external forcing, but the homogeneous generator does not commute and one easily finds

$$\mathcal{L}_1(t) = \delta\mathcal{L}(t) - \nabla_{\mathbf{Z}}^T \int_0^t d\tau \left\{ \frac{d}{dt} + \mathcal{H}(t) \right\} \Phi(t-\tau) \mathbf{F}(\tau). \quad (5.52)$$

which is *exactly* the correct effective force; note that this correction vanishes for local dissipation where  $\mathcal{H}(t)$  is constant and  $\dot{\Phi}(t) = -\mathcal{H}\Phi(t)$ . The true force is not seen in the non-Markovian master equation because, despite its Markovian representation, the response is still inherently nonlocal.

#### 5.4.2.2 Linear Forcing of a Free Particle

Finally we consider a perturbation of linear forces which take the form

$$\delta\mathcal{L}(t) = \nabla_{\mathbf{Z}}^T \mathbf{K}(t) \mathbf{Z}, \quad (5.53)$$

so that the perturbative solutions 5.2 to the Langevin equation are simply given by

$$\Phi = \Phi_0 + \Phi_1 + \dots, \quad (5.54)$$

$$\Phi_1 = - \int_0^t d\tau \Phi_0(t-\tau) \mathbf{K}(\tau) \Phi_0(\tau). \quad (5.55)$$

This is sufficient to calculate the perturbative master equation. The simplest coefficients to calculate are the homogeneous coefficients

$$\mathcal{H}(t) = \mathcal{H}_0(t) + \mathcal{H}_1(t) + \dots, \quad (5.56)$$

$$\mathcal{H}_1(t) = \mathbf{K}(t) + \int_0^t d\tau \left\{ \left[ \frac{d}{dt} + \mathcal{H}_0(t) \right] \Phi(t-\tau) \right\} \mathbf{K}(\tau) \Phi(\tau, t), \quad (5.57)$$

which would be trivial in the limit of local dissipation. Our new perturbative formalism requires evaluation of the reduced operator

$$\underline{\delta\mathcal{L}}(\tau, t) \equiv \langle \mathcal{G}_C(t, \tau) \nabla_{\mathbf{Z}}^T \mathbf{K}(\tau) \mathbf{Z} \mathcal{G}_C(\tau, 0) \rangle_E \langle \mathcal{G}_C(t, 0) \rangle_E^{-1}, \quad (5.58)$$

which from the results of Sec. 5.4 can be shown to be

$$\underline{\delta\mathcal{L}}(\tau, t) = \nabla_{\mathbf{Z}}^T \Phi(t - \tau) \mathbf{K}(\tau) \times \{ \Phi(\tau, t) \mathbf{Z} + [\Phi(\tau, t) \boldsymbol{\sigma}_T(t, t) - \boldsymbol{\sigma}_T(\tau, t)] \nabla_{\mathbf{Z}} \} . \quad (5.59)$$

It is then a straightforward calculation of Eq. 5.25 to show that the homogeneous terms here *exactly* reproduce those generated by 5.55. Comparison of the diffusion coefficients is considerably more taxing, but they also work out to be exactly the same.

### 5.4.3 New results

From Sec. 5.4.2 we now have for the external driving force

$$\delta\mathcal{L}(t) = -\nabla_{\mathbf{Z}}^T \mathbf{F}(t) , \quad (5.60)$$

$$\underline{\delta\mathcal{L}}(\tau, t) = -\nabla_{\mathbf{Z}}^T \Phi(t - \tau) \mathbf{F}(t) , \quad (5.61)$$

whereas for the linear force

$$\delta\mathcal{L}(t) = \nabla_{\mathbf{Z}}^T \mathbf{K}_0(t) \mathbf{Z} , \quad (5.62)$$

we have the two-time operator 5.59. In any case, the first-order master equation perturbative in these forces is given by

$$\mathcal{L}_1(t) = \delta\mathcal{L}(t) + \int_0^t d\tau \left\{ \frac{d}{dt} - \text{Ad}[\mathcal{L}_0(t)] \right\} \underline{\delta\mathcal{L}}(\tau, t) , \quad (5.63)$$

$$= \left\{ \frac{d}{dt} - \text{Ad}[\mathcal{L}_0(t)] \right\} \int_0^t d\tau \underline{\delta\mathcal{L}}(\tau, t) , \quad (5.64)$$

in terms of their respective two-time open-system forcing terms. These results are compatible with known results for the linear system and now we will proceed to

derive new results. First we consider an arbitrary quantum deformation of a one-dimensional system

$$\delta\mathcal{L}(t) = \frac{\partial^d}{\partial P^d} F_{db}(t) X^b, \quad (5.65)$$

and then express it in matrix notation as

$$\delta\mathcal{L}(t) = \left( \nabla_{\mathbf{Z}}^T \hat{\mathbf{P}} \right)^d F_{db}(t) \left( \hat{\mathbf{X}}^T \mathbf{Z} \right)^b, \quad (5.66)$$

where  $\hat{\mathbf{X}}$  and  $\hat{\mathbf{P}}$  are unit vectors in phase space. Applying the rules of Sec. 5.4.1, the two-time operator takes the form

$$\underline{\delta\mathcal{L}}(\tau, t) = \left[ \nabla_{\mathbf{Z}}^T \Phi(t-\tau) \hat{\mathbf{P}} \right]^d F_{db}(\tau) \sum_{k=0}^b \binom{b}{k} \left[ \hat{\mathbf{X}}^T \Phi(\tau, t) \mathbf{Z} \right]^{b-k} \Delta^{[k]}(\tau, t), \quad (5.67)$$

$$\Delta^{[k]}(\tau, t) \equiv \left\langle \mathcal{G}_{\mathbf{C}}(t, 0) \left[ \hat{\mathbf{X}}^T \int_0^t d\tau' \Phi_f(\tau, \tau') \Xi(\tau') \right]^k \right\rangle_{\Xi} \langle \mathcal{G}_{\mathbf{C}}(t, 0) \rangle_{\Xi}^{-1}, \quad (5.68)$$

where we have expanded the transformed coordinates via the binomial theorem. The final step is to evaluate the propagated noise moments via successive application of Novikov's formula. The zeroth moment is trivial and the first moment we have already calculated

$$\Delta^{[0]}(\tau, t) = 1, \quad (5.69)$$

$$\Delta^{[1]}(\tau, t) = \hat{\mathbf{X}}^T \left[ \Phi(\tau, t) \sigma_T(t, t) - \sigma_T(\tau, t) \right] \nabla_{\mathbf{Z}}, \quad (5.70)$$

while all higher-order operators can be determined recursively from integration by parts and the product rule.

$$\Delta^{[k]}(\tau, t) = \Delta^{[1]}(\tau, t) \Delta^{[k-1]}(\tau, t) - (k-1) \mathbf{s}(\tau, t) \Delta^{[k-2]}(\tau, t), \quad (5.71)$$

$$\mathbf{s}(\tau, t) \equiv \hat{\mathbf{X}}^T \left[ \sigma_T(\tau, \tau) + \Phi(\tau, t) \sigma_T(t, t) \Phi^T(\tau, t) - 2 \Phi(\tau, t) \sigma_T(t, \tau) \right] \hat{\mathbf{X}}, \quad (5.72)$$

and so  $\Delta^{[k]}(\tau, t)$  is a  $k^{\text{th}}$ -order differential operator. Thus we have explicit rules for all master equations with polynomial potentials.

### 5.4.3.1 Quadratic forcing

A quadratic correction to the spring force (with some cubic correction also in mind) can be written

$$\delta\mathcal{L}(t) = k_1(t) \left( \frac{\partial}{\partial p} x^2 - \frac{1}{12} \frac{\partial^3}{\partial p^3} \right), \quad (5.73)$$

where the second term arises only in the quantum Fokker-Planck equation; it carries an additional dimensional factor of  $\hbar^2$  and generates the quantum deformation of trajectories. To simplify calculation we rewrite these terms in matrix notation as

$$\delta\mathcal{L}(t) = k_1(t) \left( \nabla_{\mathbf{Z}}^T \hat{\mathbf{P}} \right) \left( \hat{\mathbf{X}}^T \mathbf{Z} \right)^2 - \frac{k_1(t)}{12} \left( \nabla_{\mathbf{Z}}^T \hat{\mathbf{P}} \right)^3. \quad (5.74)$$

Applying expansion formula 5.67, we find the two-time operator to take the form

$$\begin{aligned} \underline{\delta\mathcal{L}}(\tau, t) &= k_1(\tau) \left( \nabla_{\mathbf{Z}}^T \Phi(t-\tau) \hat{\mathbf{P}} \right) \left( \hat{\mathbf{X}}^T \Phi(\tau, t) \mathbf{Z} \right)^2 \\ &\quad + 2 k_1(\tau) \left( \nabla_{\mathbf{Z}}^T \Phi(t-\tau) \hat{\mathbf{P}} \right) \left( \hat{\mathbf{X}}^T \Phi(\tau, t) \mathbf{Z} \right) \Delta^{[1]}(\tau, t) \\ &\quad + k_1(\tau) \left( \nabla_{\mathbf{Z}}^T \Phi(t-\tau) \hat{\mathbf{P}} \right) \left( \Delta^{[1]}(\tau, t)^2 - \mathbf{s}(\tau, t) \right) \\ &\quad - \frac{k_1(\tau)}{12} \left( \nabla_{\mathbf{Z}}^T \Phi(t-\tau) \hat{\mathbf{P}} \right)^3. \end{aligned} \quad (5.75)$$

### 5.4.3.2 Cubic forcing

The cubic correction to the spring force can be written

$$\delta\mathcal{L}(t) = k_2(t) \left( \frac{\partial}{\partial p} x^3 - \frac{1}{4} \frac{\partial^3}{\partial p^3} x \right), \quad (5.76)$$

and in matrix notation

$$\delta\mathcal{L}(t) = k_2(t) \left( \nabla_{\mathbf{Z}}^T \hat{\mathbf{P}} \right) \left( \hat{\mathbf{X}}^T \mathbf{Z} \right)^3 - \frac{k_2(t)}{4} \left( \nabla_{\mathbf{Z}}^T \hat{\mathbf{P}} \right)^3 \left( \hat{\mathbf{X}}^T \mathbf{Z} \right), \quad (5.77)$$

which yields the two-time operator

$$\begin{aligned} \underline{\delta\mathcal{L}}(\tau, t) &= k_2(\tau) \left( \nabla_{\mathbf{Z}}^T \Phi(t-\tau) \hat{\mathbf{P}} \right) \left( \hat{\mathbf{X}}^T \Phi(\tau, t) \mathbf{Z} \right)^3 \\ &+ 2 k_2(\tau) \left( \nabla_{\mathbf{Z}}^T \Phi(t-\tau) \hat{\mathbf{P}} \right) \left( \hat{\mathbf{X}}^T \Phi(\tau, t) \mathbf{Z} \right)^2 \Delta^{[1]}(\tau, t) \\ &+ 2 k_2(\tau) \left( \nabla_{\mathbf{Z}}^T \Phi(t-\tau) \hat{\mathbf{P}} \right) \left( \hat{\mathbf{X}}^T \Phi(\tau, t) \mathbf{Z} \right) \left( \Delta^{[1]}(\tau, t)^2 - \mathbf{s}(\tau, t) \right) \\ &+ k_2(\tau) \left( \nabla_{\mathbf{Z}}^T \Phi(t-\tau) \hat{\mathbf{P}} \right) \left( \Delta^{[1]}(\tau, t)^3 - 3 \mathbf{s}(\tau, t) \Delta^{[1]}(\tau, t) \right) \\ &- \frac{k_2(\tau)}{4} \left( \nabla_{\mathbf{Z}}^T \Phi(t-\tau) \hat{\mathbf{P}} \right)^3 \left( \hat{\mathbf{X}}^T \Phi(\tau, t) \mathbf{Z} \right) \\ &- \frac{k_2(\tau)}{4} \left( \nabla_{\mathbf{Z}}^T \Phi(t-\tau) \hat{\mathbf{P}} \right)^3 \Delta^{[1]}(\tau, t). \end{aligned} \quad (5.78)$$

## 5.5 Discussion

We have derived a fairly general theory of strong-coupling perturbation for continuous variable systems. Our formalism makes heavy use of the phase-space representation as the zeroth-order problem is a linear one and the phase-space representation is perhaps the simplest formalism for that case. Essentially we are perturbing off of QBM, where the QBM-like model provides us with stochastic coordinates which do not solve the problem, but integrate the environment dynamics. This results in a combined nonlinear (in terms of forces) and stochastic dynamics of the unraveled system. From there we may apply the standard perturbation theory of master equations, one wherein the perturbative time-translation generator is calculated. This method retains important Lie-group symmetries and grants the

possibility of non-secular behavior (thus allowing for late-time solutions). As one might expect our master equation describes highly non-Markovian dynamics (thus exhibiting much time dependence) and is highly model specific.

These results are given in a form which is calculable, but far more complicated than what one would desire at first order. At least two avenues of attack must be considered. In the larger view one must question if there is a more suitable formalism which includes all necessary ingredients: the stochastic map, open and closed-system propagation, etc.. The linear forces of the zeroth-order dynamics is simplest here in the phase-space representation, but perhaps we could trade some of that simplicity for a representation which is more apt for the nonlinear forces. Or perhaps a combined approach could best utilize properties of different representations. On the other hand, in the more localized view one must address some simplified calculation of the noise averages which predominantly rely upon knowledge of the two-time thermal correlation and integrals thereof. We have demonstrated that a true strong-coupling master equation exists, and we have explicitly given its form, but a large amount of work remains to apply our results to representative physical setups.

## Chapter 6

### Entanglement Dynamics of Field-Immersed Multipartite Systems

#### 6.1 Introduction and Overview

Atomic systems constitute an important setting for the investigation of quantum decoherence and entanglement phenomena essential for quantum information processing considerations [36, 131, 116, 101]. The physical principles underlying these systems are quite well understood, and they can be controlled and measured with great precision. One aspect of quantum entanglement dynamics that has received significant attention is the phenomenon of entanglement sudden death, or finite-time disentanglement, while energy and local coherences only decay away exponentially in time [89, 159, 162]. A common setting for theoretical discussion of this phenomenon is atomic systems interacting with the electromagnetic field [159, 157, 158, 58, 9, 10], serving as an environment in the quantum open system (QOS) perspective.

In this chapter we consider otherwise non-interacting atoms held fixed in a shared quantum field. In Sec. 6.2 we discuss the influence kernels and statistical correlations of the relativistic scalar and electromagnetic fields. These quantities will then be applied to two different system models in the later sections. Only in Chapter 7 will we consider motional dynamics in our atoms, which leads to the infamous Abraham-Lorentz radiation reaction.

In Sec. 6.3 we derive the stochastic equations and consider the non-Markovian dynamics of a system of multiple two-level atoms in a common quantum field. We make only the dipole approximation for the atoms and assume weak atom-field interaction, but no Born-Markov (BMA) or rotating-wave approximation (RWA). These more accurate solutions are necessary if one wants to determine a) whether late-time asymptotic entanglement exists and b) whether any initial state can avoid sudden death, questions of practical importance for quantum information processing. We find that even at zero temperature all initial states will undergo finite-time disentanglement (or eventually meet with ‘sudden death’), in contrast to previous work. We also use our solution without invoking RWA to fully characterize the necessary conditions for the sub-radiant dark state, which can be used to preserve coherence and entanglement for long times. For sub-radiance and super-radiance to be achieved, the atoms must be held close in relation to their resonant wavelength, and they must be tuned closely in relation to the normal dissipation rate. This latter regime cannot be described by Lindblad equations. Temperature does not alter the existence of such states. We discuss how the phenomena of sub and super-radiance can be viewed as an *interference phenomenon* among the noise processes and give a simple explanation of why the super-radiance emission rate scales like the number of atoms squared. We also give an in-depth treatment of renormalization, which takes into account the correlated influences between atoms and the importance of time dependent renormalization in preserving causality.

In Sec. 6.4 we apply the exact formalism of Chapter 3 and make a similar analysis of multiple local oscillators in a common scalar field. There has been

a great deal work on this model by Lin and Hu (see e.g., [105]), mostly within the confines of the resonant and weak-coupling regime. It should also be noted that Paz and Roncaglia [122, 123] effectively considered the entanglement of two resonant oscillators at close proximity, but without assuming weak-coupling. Using our Chapter 3 we are able to provide calculations which encompass all of the above regimes and more. This also extends beyond the regimes accessible for two-level systems, where we are more restricted by perturbation theory.

## 6.2 Relativistic Fields

### 6.2.1 Interacting Hamiltonians

We begin with the QED Hamiltonian of non-relativistic charged particles moving in potentials  $V_{nm}(\mathbf{X})$  and coupled to quantized electromagnetic field modes of wavelength  $2\pi/k$  and in the Coulomb gauge.

$$\mathbf{H}_{\text{QED}} = \sum_n \frac{[\mathbf{P}_n - e_n \mathbf{A}(\mathbf{X}_n)]^2}{2M_n} + \sum_{nm} V_{nm}(\mathbf{X}_n - \mathbf{X}_m) + \mathbf{H}_{\text{field}}, \quad (6.1)$$

$$\mathbf{H}_{\text{field}} = \iiint d^3k \sum_{\epsilon_k} \omega_k \mathbf{a}_{\mathbf{k},s}^\dagger \mathbf{a}_{\mathbf{k},s}, \quad (6.2)$$

where the vector potential  $\mathbf{A}$  is given by the spatial Fourier and polarization decomposition

$$\mathbf{A}(\mathbf{X}) = \frac{1}{(2\pi)^{3/2}} \iiint d^3k \sum_{\epsilon_k} \mathbf{A}_{\mathbf{k},\epsilon_k}(\mathbf{X}), \quad (6.3)$$

$$\mathbf{A}_{\mathbf{k},\epsilon_k}(\mathbf{X}) = \frac{\epsilon_k}{\sqrt{2\epsilon_0\omega_k}} \left\{ e^{+i\mathbf{k}\cdot\mathbf{X}} \mathbf{a}_{\mathbf{k},\epsilon_k} + e^{-i\mathbf{k}\cdot\mathbf{X}} \mathbf{a}_{\mathbf{k},\epsilon_k}^\dagger \right\}, \quad (6.4)$$

where  $k = \omega_k/c$  and where  $\boldsymbol{\epsilon}_k$  denote the polarization vectors perpendicular to  $\mathbf{k}$  [114, 39]. As we have considered the non-relativistically appropriate Coulomb gauge, all electrostatic potentials are to be included in  $V(\mathbf{X})$  and the vector potential is purely transverse.

For two-level atoms at rest we will consider the related dipole interaction (derived in [8], App. A), where only the lowest-level excitations of the Dirac field are considered, so that ignoring the electrostatic interactions (neutral atoms far apart) we have

$$\mathbf{H}_{2LA} = \sum_n \Omega_n \boldsymbol{\sigma}_{+n} \boldsymbol{\sigma}_{-n} + \sum_n \sigma_{x_n} \mathbf{d}_n \cdot \mathbf{A}(\mathbf{R}_n) + \mathbf{H}_{\text{field}}, \quad (6.5)$$

where  $\boldsymbol{\sigma}$  denote the Pauli matrices and  $\mathbf{d}$  corresponds to the lowest-energy levels of the electron's dipole matrix [3, 150, 8]. Note that we have neglected the  $\mathbf{A}^2$  term here. As we only consider a second-order analysis of the open system, it is then sufficient to consider only  $\mathbf{H}_{\text{field}}$ , specifically in calculating the second-order environment correlations. Contributions from  $\mathbf{A}^2$  would only enter into the fourth-order correlations, and thus the fourth-order master equation.

In the remainder of this chapter we will refer to static locations with  $\mathbf{R}$ , as we do not want to confuse field evaluation locations with dynamical observables. For our Brownian oscillator atoms we essentially consider the atom-field coupling  $\mathbf{X} \cdot \mathbf{A}(\mathbf{R})$ , carefully distinguishing between the spatial location of the atom  $\mathbf{R}$  and the “position”  $\mathbf{X}$  which corresponds to the atom's internal degree of freedom. Our Brownian atoms will also be taken to be at rest.

## 6.2.2 Environment Correlations

We will now proceed to detail the electromagnetic correlation function.

$$\boldsymbol{\alpha}_{nm}(t, \tau) = \frac{e_n e_m}{M_n M_m} \langle \underline{\mathbf{A}}(\mathbf{R}_n; t) \underline{\mathbf{A}}^T(\mathbf{R}_m; \tau) \rangle_{\text{E}}, \quad (6.6)$$

where for simplicity we will assume that all system masses are  $M$  and all system charges are  $q$ . In addition to the indices  $n$  and  $m$ , which run over the system particles,  $\boldsymbol{\alpha}$  is also a  $3 \times 3$  matrix in accord with  $\mathbf{A}$  being a 3-dimensional vector.

### 6.2.2.1 Scalar-Field Correlations

First let us consider the case of a scalar field, wherein one essentially neglects the polarization vectors as if  $\mathbf{A}$  were scalar and thus  $\boldsymbol{\alpha}_{nm}$  is also scalar. In this case we calculate the associated damping kernel to be

$$\tilde{\gamma}_{nm}(\omega) = \tilde{\gamma}_0 \text{sinc}(R_{nm}\omega), \quad (6.7)$$

where  $\mathbf{R}_{nm} = \mathbf{R}_n - \mathbf{R}_m$  is the difference vector for the positions of the two atoms and  $\tilde{\gamma}_0 = \tilde{\gamma}_{nn}(\omega)$  is the self-damping coefficient. Restoring factors of  $c$ , the sinc function appears to act as a relativistic regulator for  $\omega \ll c/R$ . Therefore, in the nonrelativistic regime one only has local damping for charges at precisely the same location. In fact, this must always be the case for linear (field) coupling to a relativistic field. The damping kernel is deterministic and the same whether in the classical or quantum regimes. Thus it is necessarily constrained to the light cone.

Note for instance the temporal representation of the scalar-field damping kernel

$$\gamma_{nm}(t) = \frac{\tilde{\gamma}_0}{2} \delta_{R_{nm}}(t), \quad (6.8)$$

$$\delta_R(t) \equiv \frac{\theta(R-|t|)}{2R}, \quad (6.9)$$

where  $\theta$  is the Heaviside step function. This kernel strictly adheres to the light cone.

### 6.2.2.2 Electromagnetic-Field Correlations

The electromagnetic damping kernel is slightly more complicated as it contains orientation dependence from the polarization vectors.

$$\tilde{\gamma}_{nm}(\omega) = \tilde{\gamma}_0 \left\{ \text{FS}_1(R_{nm}\omega) \mathbf{1} - \frac{1}{2} \text{FS}_0(R_{nm}\omega) \hat{\mathbf{R}}_{nm} \hat{\mathbf{R}}_{nm}^T \right\}, \quad (6.10)$$

in the Fourier domain. Instead of a sinc functions, we have the related entire functions

$$\text{FS}_1(z) \equiv \frac{3(z^2 - 1) \sin(z) + z \cos(z)}{z^3}, \quad (6.11)$$

$$\text{FS}_0(z) \equiv 3 \frac{(z^2 - 3) \sin(z) + 3z \cos(z)}{z^3}. \quad (6.12)$$

In Fig. 7.1 we compare these functions. One can see that the scalar-field correlations are very similar to that of the electromagnetic field when  $\mathbf{d}_n \parallel \mathbf{d}_m \perp \mathbf{R}_{nm}$ , when considering  $\mathbf{d}_n^T \tilde{\gamma}_{nm}(\omega) \mathbf{d}_m$ . As we will wish to maximize cross correlations, we will primarily work with the scalar-field correlations, which one can think of as being very similar to the parallel dipoles.

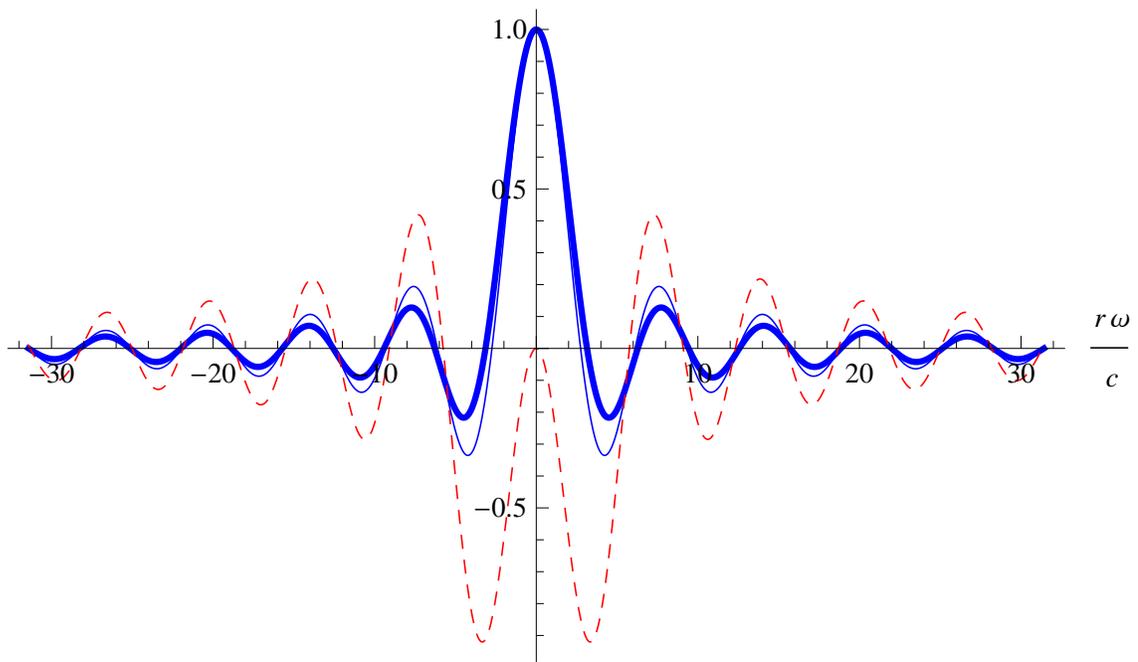


Figure 6.1: Comparison of sinc (bold), FS<sub>1</sub>, and FS<sub>0</sub> (dashed). Sinc and FS<sub>1</sub> are extremely qualitatively similar, both being unity at zero whereas FS<sub>0</sub> vanishes at zero.

### 6.2.2.3 Noise Kernel

The fluctuation-dissipation relation allows us to express the environmental correlations in terms of the damping kernel as

$$\tilde{\alpha}(\omega) = \tilde{\gamma}(\omega) \frac{\omega}{\sinh\left(\frac{\omega}{2T}\right)} e^{-\frac{\omega}{2T}} = 2 \tilde{\gamma}(\omega) \omega [\bar{n}(|\omega|, T) + \theta(-\omega)] , \quad (6.13)$$

and also the noise kernel as

$$\tilde{\nu}(\omega) = \tilde{\gamma}(\omega) \omega \coth\left(\frac{\omega}{2T}\right) = \tilde{\gamma}(\omega) |\omega| [\bar{n}(|\omega|, T) + 1] , \quad (6.14)$$

where  $\bar{n}(\omega, T)$  is the thermal average photon number in a mode of frequency  $\omega$ . The role of these kernels was explained in Sec. 2.5. Very briefly, the damping kernel  $\tilde{\gamma}(\omega)$  characterizes dissipation, the noise kernel  $\tilde{\nu}(\omega)$  characterizes diffusion, and the full quantum correlation  $\tilde{\alpha}(\omega)$  can be thought to characterize decoherence. For our model, the near and far correlations are not ordered and therefore we cannot make any general statements regarding one limit always providing more dissipation, diffusion, and decoherence than the other. However, for two very close and parallel dipoles the off-diagonal entries of the kernels approach the diagonal values, and in doing so an eigen-value must also vanish. At resonance this damping and decoherence deficit can give rise to a “dark state” as we explain more thoroughly in Sec. 6.3.2.3.

## 6.3 Two-Level Systems

Much of the theoretical work on atom-field systems is derived using the *rotating-wave approximation* (RWA) [5, 150, 32] (Sec. 4.4). When considering atomic dynam-

ics with an open-system approach, where there is a continuum of field modes that are treated as a reservoir, the *Born-Markov approximation* (BMA) (Sec. 2.2.1.5) is commonly invoked, usually in combination with a RWA [5, 32, 22]. However, if such systems are central to many important investigations of broad relevance, both experimental and theoretical, and one wishes to examine subtle features such as entanglement dynamics, then great care must be taken in the use of approximations and the accuracy of results so derived. Both of these approximations require an assumption of weak system-environment coupling to be justified.

A fully non-Markovian treatment of multiple two-level atoms in a common quantum field has yet to be carried out in a manner which can predict entanglement evolution fully and address critical issues such as sudden death of entanglement. There are several important reasons for this. (1) Calculations have usually involved perturbative master equations, either explicitly or by invoking the BMA or RWA which both implicitly assume a perturbative coupling to the environment. However, second-order master equations fundamentally cannot rule out second-order amounts of entanglement in many situations. This is due to the little-known fact that second-order (non-Markovian) master equations are not generally capable of providing full second-order solutions except at early times (Sec. 4.2). (2) Use of a RWA will lead to inaccurate predictions of late-time asymptotic entanglement differing by second-order terms from the actual value, and at low temperatures this can potentially lead to entirely different and erroneous qualitative features of entanglement dynamics and sudden death. The RWA only captures certain timescales to second order, while at any instance density matrix elements can have second-order inaccuracies

(Sec. 4.4) (except at early times). However we would note that a non-perturbative (or higher-order) treatment of the model with an RWA system-environment interaction is still qualitatively interesting. (3) Use of the RWA also does not allow for consideration of near resonance (as additional near-stationary terms are needed in the Dirac picture)<sup>1</sup>. The existence of a sub-radiant *dark state* generically requires the resonance condition, but determining how critical this is requires some analysis of the near-resonance regime.

In this section, rather than employing the BMA or RWA, we use a conceptually straight-forward implementation of perturbation theory, assuming only weak system-environment coupling as laid out in Chapter 2. We make careful and justified use of the second-order master equation for the dynamics, paying attention to the expected accuracy of the solutions. We use an alternative (but compatible) means of calculating the late-time asymptotics. In this way we are able to show that the two atoms in a single field are not asymptotically entangled, even when near resonance and very close together — which is the criterion for a dark state. This asymptotic behavior turns out to be rather opposite to that of two oscillators in a field, which can be asymptotically entangled (Sec. 6.4). In fact, we find that the entanglement of any pair of atoms will always undergo sudden death, regardless of the initial state. Furthermore we make a detailed analysis of how coherence can be long lived amongst the ground state and dark states, and we proceed to describe all relevant timescales of the atom-field system. We explore what conditions are

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<sup>1</sup>Near resonant terms can be preserved in implementing the RWA, but this will then lead to a master equation not of Lindblad-form as in [136].

required for sub and super-radiance in terms of proximity, tuning, and dissipation. In brief, to achieve dark and bright states one requires proximity better than the resonant wavelength and tuning better than the ordinary dissipation rates. Temperature only appears to change the nature of these states and does not diminish their existence (other than increasing any positive decay rates linearly).

In physical terms the sub- and super-radiance of the dark and bright states are ultimately a result of *interference* among the multiple noise processes provided by the field modes evaluated at different locations. As such, one cannot simply add the emission rates of two isolated atoms. Some special mention should also be given to our treatment of renormalization. Previous works have only considered renormalization of the atoms individually, which is sufficient if the atoms are far apart, and also simultaneous in time, which is sufficient in the late-time regime. Here we “dress” the joint system in its entirety, which gives rise to an immersed dynamics more similar to the free system and also more well behaved. Our counter terms are also introduced along the light cone, which keeps the full-time theory causal, and not across all of space simultaneously.

We now describe the perturbative second-order master equation in Sec. [6.3.1](#). Then in Sec. [6.3.2](#) we explain our method of solution, the resulting dynamics, and the accuracy of these solutions. We also discuss the asymptotic state and entanglement dynamics specifically in Sec. [6.3.2.5](#).

## 6.3.1 Second-Order Master Equation

### 6.3.1.1 System-environment coupling and correlations

We wish to investigate the properties of multiple atoms interacting with a common electromagnetic field in free space, which serves as the environment in the open quantum system description. We will use the two-level approximation to describe the atoms, so that they are an array of, otherwise non-interacting, qubits. Thus we can write the Hamiltonian of our system + field as in Eq. (6.5).

$$\mathbf{H}_{2\text{LA}} = \underbrace{\sum_n \Omega_n \sigma_{+n} \sigma_{-n}}_{\mathbf{H}_S} + \underbrace{\sum_n \sigma_{x_n} \mathbf{d}_n \cdot \mathbf{A}(\mathbf{R}_n)}_{\mathbf{H}_I} + \underbrace{\mathbf{H}_{\text{field}}}_{\mathbf{H}_E}, \quad (6.15)$$

where  $\sigma$  denote the Pauli matrices and  $\mathbf{d}$  corresponds to the lowest-energy levels of the electron's dipole matrix [8]. The interaction of our system with the environment is represented such that  $\sigma_{x_n}$  denotes the  $x$  spin component of the  $n^{\text{th}}$  qubit and  $\mathbf{R}_n$  is the location of the qubit where the field  $\mathbf{A}$  is evaluated.

We assume the atoms to be relatively stationary and assume that the atomic transition in each atom will produce linearly polarized photons (i.e., both ground and excited state are eigenstates of some component of angular momentum with the same eigenvalue). Under these assumptions relevant field correlations are then given by Eq. (6.10) and the fluctuation-dissipation relation (6.13). We wish to maximize induced interactions between the atoms and therefore, from the analysis in Sec. 6.2.2.2, we will primarily consider a lattice of parallel dipoles with  $\mathbf{d} \perp \mathbf{R}_{nm}$ . Qualitatively speaking, the scalar-field correlations are then a good approximation, and so we apply Eq. (6.7) for simplicity.

### 6.3.1.2 Master Equation and Coefficients

The second-order master equation for the reduced density matrix of the dipoles can be expressed following the results of Chapter 2.

$$\dot{\rho} = (\mathcal{L}_0 + \mathcal{L}_2) \rho, \quad (6.16)$$

in terms of the zeroth and second-order Liouville operators

$$\mathcal{L}_0 \rho = [-\imath \mathbf{H}, \rho], \quad (6.17)$$

$$\mathcal{L}_2 \rho = \sum_{nm} [\sigma_{x_n}, \rho (\mathbf{A}_{nm} \diamond \sigma_{x_m})^\dagger - (\mathbf{A}_{nm} \diamond \sigma_{x_m}) \rho], \quad (6.18)$$

where  $\mathbf{H} = \mathbf{H}_S$  and with the second-order operator most easily represented by the ladder operators as

$$(\mathbf{A}_{nm} \diamond \sigma_{x_m}) = A_{nm}(+\Omega_m) \sigma_{+m} + A_{nm}(-\Omega_m) \sigma_{-m}, \quad (6.19)$$

$$\sigma_{\pm} \equiv \frac{1}{2} [\sigma_x + \imath \sigma_y], \quad (6.20)$$

and the second-order coefficients being related to the correlation function as

$$A_{nm}(\omega) = \frac{1}{2} \tilde{\alpha}_{nm}(\omega) - \imath \mathcal{P} \left[ \frac{1}{\omega} \right] * \tilde{\alpha}_{nm}(\omega), \quad (6.21)$$

here in the late-time limit (as compared to system and cutoff timescales), where  $\mathcal{P}$  denotes the Cauchy principal value. Higher-order master equation coefficients will entail convolutions over several copies of the correlation function combined with several products of the system coupling operator.

The first portion of the second-order coefficient, or Hermitian part (here real), is immediately given by Eq. (6.13). Whereas the second term, or anti-Hermitian

part (here imaginary), must be evaluated via the convolution

$$\text{Im}[A_{nm}(\omega)] = -\frac{1}{2\pi} \int_{-\infty}^{+\infty} d\varepsilon \mathcal{P} \left[ \frac{1}{\omega - \varepsilon} \right] \tilde{\alpha}_{nm}(\varepsilon), \quad (6.22)$$

and together they form a causal response function. These are the coefficients which often require regularization and renormalization. For now let us simply evaluate the bare coefficients for non-vanishing  $R$ . For finite temperatures, the coefficients exactly evaluate to

$$\begin{aligned} \text{Im}[A_{nm}(\omega)] = & +\frac{\tilde{\gamma}_0}{R_{nm}} \frac{1}{\pi} \text{Im} \left[ \Phi_1 \left( \frac{\imath \omega}{2\pi T}; 2\pi T R_{nm} \right) \right] \\ & - \frac{\tilde{\gamma}_0}{R_{nm}} \left\{ \frac{T}{\omega} - \frac{1}{2} \left[ \coth \left( \frac{\omega}{2T} \right) - 1 \right] \cos(R_{nm}\omega) \right\}, \end{aligned} \quad (6.23)$$

in terms of the Lerch  $\Phi_1$  function

$$\Phi_1(z; \lambda) \equiv \sum_{k=1}^{\infty} \frac{e^{-k\lambda}}{k+z}. \quad (6.24)$$

This functional representation is exact, though best for positive temperature. Conversely, one also has the low-temperature expansion

$$\begin{aligned} \text{Im}[A_{nm}(\omega)] = & \frac{\tilde{\gamma}_0}{R_{nm}} \frac{\text{sign}(\omega)}{\pi} \sum_{k=1}^{\infty} S_k \\ & - \frac{\tilde{\gamma}_0}{R_{nm}} \frac{1}{\pi} [\sin(R_{nm}\omega) \text{ci}(|R_{nm}\omega|) - \cos(R_{nm}\omega) \text{si}(R_{nm}\omega)], \end{aligned} \quad (6.25)$$

in terms of the summand

$$S_k = \frac{\text{Ei}[(+k\beta + \imath R_{nm})|\omega|]}{e^{(+k\beta + \imath R_{nm})|\omega|}} + \frac{\text{Ei}[-k\beta + \imath R_{nm})|\omega|] - \imath \pi}{e^{(-k\beta + \imath R_{nm})|\omega|}}, \quad (6.26)$$

and where the trigonometric integrals are defined

$$\text{si}(z) \equiv - \int_z^{\infty} dz' \frac{\sin(z')}{z'}, \quad (6.27)$$

$$\text{ci}(z) \equiv - \int_z^{\infty} dz' \frac{\cos(z')}{z'}, \quad (6.28)$$

$$\text{Ei}(z) \equiv - \int_{-z}^{\infty} dz' \mathcal{P} \left[ \frac{e^{-z'}}{z'} \right], \quad (6.29)$$

however, for positive temperatures this expansion is not well behaved for small energy differences. For zero temperature, the exact relation (the second line in (6.25)) is well behaved and matches perfectly to the zero-temperature limit of Eq. (6.23).

### 6.3.1.3 Asymptotic Regularization and Renormalization

Note that  $\text{sinc}(\omega/\Lambda)$  is a high frequency regulator:  $\text{sinc}(z) : [0, \infty) \rightarrow [1, 0)$  sufficiently fast for all of our integrals to converge. Therefore we don't need to consider any additional regularization in our damping kernel if we do not evaluate  $\text{sinc}(R\omega)$  for vanishing  $R$ . Instead of allowing  $R$  to vanish for self-correlations, we impose a high frequency cutoff  $R_0 = \Lambda^{-1}$ , perhaps motivated by the non-vanishing physical size of the dipole. The more common alternative is to introduce cutoff regularization directly into the field operator  $\mathbf{A}$  in Eq. (6.3), often by treating the coupling strength as a form factor with some gradual  $k$ -dependence. Different choices of cutoff regulators will yield the same results to highest order in  $\Lambda$ , and the theory should be somewhat insensitive to these details in the end.

Given some form of regularization, the coefficients are then bounded yet cutoff sensitive. The remaining cutoff sensitivity is reduced through a renormalization scheme. The typical scheme in quantum open systems is to subtract off the zero-energy correction

$$\text{Im}[A_{nm}(0)] = -\frac{\tilde{\gamma}_0}{2 R_{nm}}. \quad (6.30)$$

There are numerous reasons for this choice of renormalization given in Sec. 2.5.2.1. Most importantly, it keeps the homogeneous dynamics of the interacting system

most closely resembling that of the free system. However, in electrodynamics there are forms of non-dissipative backreaction which enter at second order and are extremely important. At lowest order they are, in fact, the magnetostatic interactions. In this thesis we will momentarily ignore these very interesting  $1/r$  interactions, including the self interactions which must be renormalized. We shall address this physics in the near future.

#### 6.3.1.4 Full-Time Regularization and Renormalization

For the full-time evolution of initially uncorrelated states, one must apply the full-time coefficients

$$A_{nm}(\omega; t) = \int_0^t d\tau e^{-i\omega\tau} \alpha_{nm}(\tau), \quad (6.31)$$

which must exhibit causal behavior in as much as the field correlations are causal. At zero temperature there is a  $(R-t)^{-1}$  pole in the integrand which can be encapsulated by contour integrals for  $t > R$ . The encapsulation of this pole produces an activation *jolt* in the master equation coefficients precisely at  $t = R$  which we plot in Fig. 6.2. Prior to the jolt, the master equation coefficients are roughly zero; whereas after the jolt, the coefficients are roughly their asymptotic value. For positive temperatures there is an infinite series of poles increasingly attenuated by the rising temperature.

With the consideration of renormalization, one becomes even more directly confronted with causality. If renormalization is applied to the entire system simultaneously, e.g.

$$\text{Im}[A_{nm}(\omega; t)] \rightarrow \text{Im}[A_{nm}(\omega; t)] - \text{Im}[A_{nm}(0; \infty)], \quad (6.32)$$

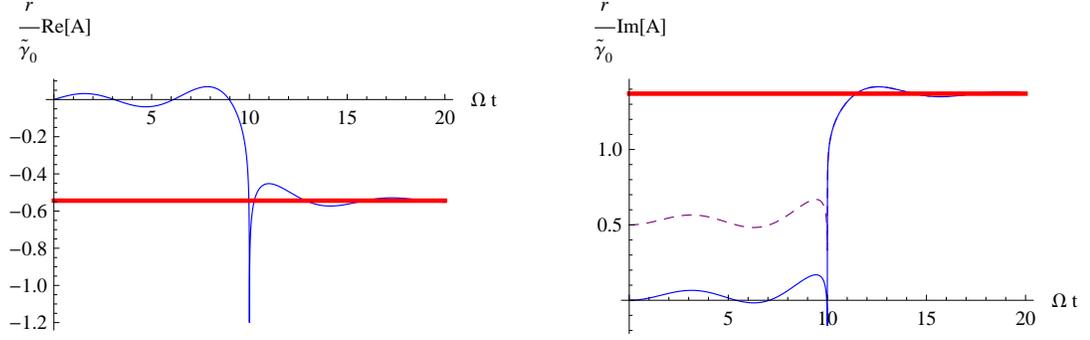


Figure 6.2:  $\text{Re}[A_{nm}(-\Omega_m; t)]$  (left) and renormalized  $\text{Im}[A_{nm}(-\Omega_m; t)]$  (right) for a zero temperature reservoir at  $R_{nm} = 10/\Omega_m$ . The bold line denotes the asymptotic coefficients. For the latter plot, the dashed curve is the result of simultaneous renormalization and is acausal.

then the renormalization will be felt instantaneously over finite distances. Effectively such an acausal renormalization is introducing a counter term into the Hamiltonian at  $t = 0$  for which there is nothing to counter until  $t > R$ . Whereas if renormalization is applied at a retarded time, e.g.

$$\text{Im}[A_{nm}(\omega; t)] \rightarrow \text{Im}[A_{nm}(\omega; t)] - \theta(t - R_{nm}) \text{Im}[A_{nm}(0; \infty)], \quad (6.33)$$

where  $\theta(z)$  denotes the unit-step function, then the renormalized theory will be as causal in its behavior as the non-renormalized theory. Renormalization (and any state preparation, see Sec. 4.3) must be performed in a causal manner (along the light cone) if one desires causal evolution. Improper renormalization, in the context of a factorized initial state of the system and environment, will create (apparently) mediated interactions between the atoms which are activated before mediation can actually occur. Such a theory is Hamiltonian, but not relativistic.

### 6.3.2 Second-Order Solutions

From an analysis of the full-time coefficients (see Fig. 6.2), one can see that each coefficient jolts on at  $t = R$ . [The jolting (here logarithmic divergence) is a result of the factorized initial conditions and would become a more smooth activation upon considering properly correlated initial states or switching on the interaction gradually.] So for  $t < R$  the atoms roughly evolve independently (equivalent to  $R \rightarrow \infty$ ) and then for  $t > R$  the atoms become aware of each other's presence and evolve jointly. If there is any acausal behavior, such as creation of entanglement outside of the light cone, then it would have to be very small.

Because the master equation coefficients mostly asymptote to constant values quite quickly here in the weak-coupling regime, as can be seen in Fig. 6.2, it is sufficient to consider a sequence of constant Liouvillians [65]. E.g. for two atoms

$$\mathcal{L}_{[R]}(t) \approx \begin{cases} \mathcal{L}_{[\infty]}(\infty) & t < r \\ \mathcal{L}_{[R]}(\infty) & t > r, \end{cases} \quad (6.34)$$

and therefore the full-time propagator can be sufficiently approximated by a chain of exponential propagators, here

$$\mathcal{G}_{[R]}(t) \approx \begin{cases} e^{t\mathcal{L}_{[\infty]}(\infty)} & t < r \\ e^{t\mathcal{L}_{[R]}(\infty)} e^{r\mathcal{L}_{[\infty]}(\infty)} & t > r. \end{cases} \quad (6.35)$$

A more accurate full-time treatment would be sensitive to initial conditions, and our factorized initial conditions are not reasonable enough to warrant that level of scrutiny for any physical applications. For the remainder of the paper, we will be interested in the  $t \gg R$  regime. The challenge then lies in calculating the evolution due to  $e^{t\mathcal{L}_{[R]}(\infty)}$ .

### 6.3.2.1 Dynamics

The open-system dynamics are described approximately by the time-independent Liouvillian  $\mathcal{L}_{[R]}(\infty)$ , which we will now write simply as  $\mathcal{L}$ . The time evolution is then approximately  $e^{t\mathcal{L}}$ , and it can be computed (analogously to the time-independent Schrödinger equation) simply from the solutions of the eigen-value problem

$$\mathcal{L}\{\mathbf{o}\} = f\mathbf{o}, \quad (6.36)$$

where  $f$  is an eigen-frequency and  $\mathbf{o}$  a right eigen-operator (super-vector). The canonical perturbation theory for this calculation was detailed in Chapter 2. Because the master equation itself is perturbative, there is no loss in accuracy by finding the solutions perturbatively.

Due to unavoidable degeneracy (including any resonant frequencies), the second-order operator perturbations actually require the fourth-order Pauli master equation (see Sec. 4.2). In Sec. 6.3.2.5 we use an alternative means to calculate corrections to the asymptotic or reduced thermal state using only the second-order coefficients, as per the results of Sec. 4.5. To briefly summarize, in general the matrix elements of the solution  $\rho(t)$  expressed in the (free) energy basis will be accurate to  $\mathcal{O}(\gamma)$  off the diagonal but only to  $\mathcal{O}(1)$  on the diagonal (though timescales are known to  $\mathcal{O}(\gamma)$ ). This inaccuracy in the diagonals is an inherent limitation of any perturbative master equation, including those derived under the RWA or the BMA. With the RWA, however, *all* matrix elements are only good to  $\mathcal{O}(1)$ <sup>2</sup>.

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<sup>2</sup>When looking only at observables time-averaged over many system periods  $2\pi/\Omega$  some of these additional discrepancies generated by the RWA can be greatly reduced.

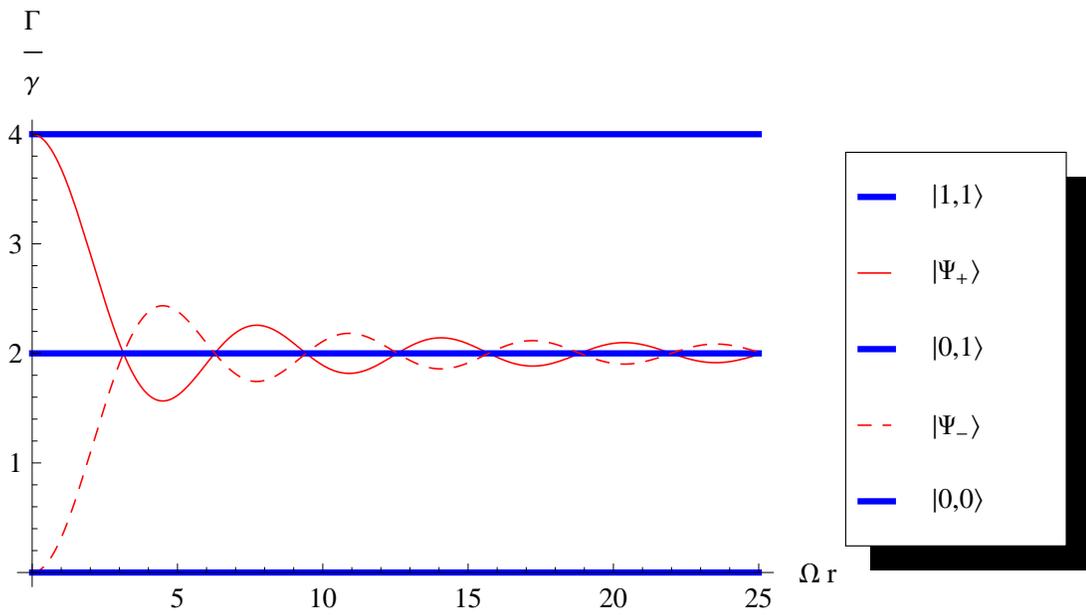


Figure 6.3: Decay rates of the (zeroth-order) stationary operators for two resonant atoms in a zero-temperature environment at varying separation distance. The legend indicates the pure states they approximately correspond to in the order they occur at the vertical axis.

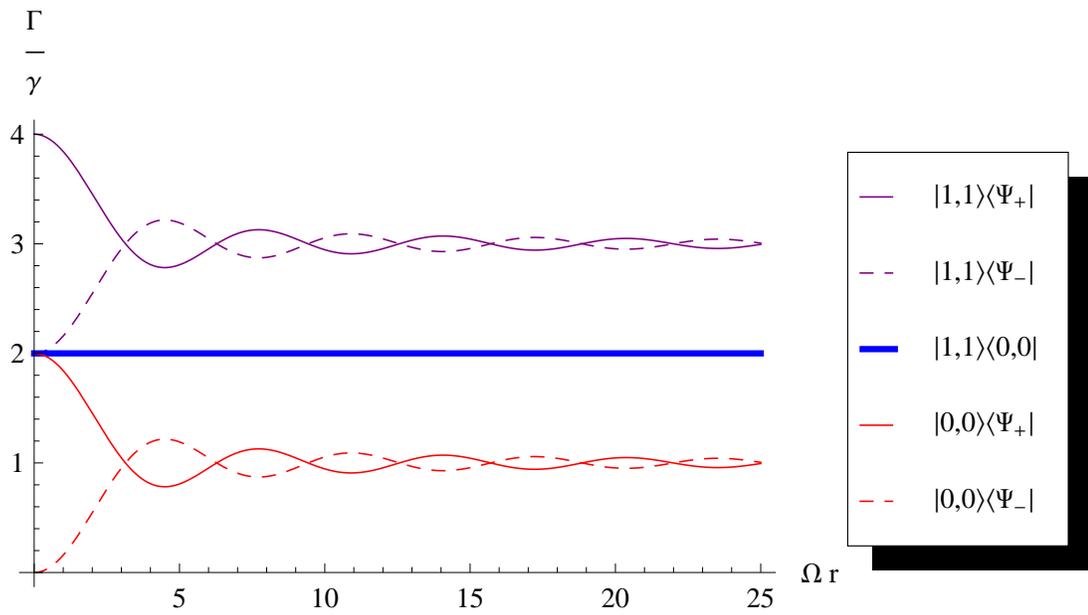


Figure 6.4: Decoherence rates of the (zeroth-order) non-stationary operators for two resonant atoms in a zero-temperature environment at varying separation distance. The legend indicates the matrix elements they correspond to in the order they occur at the vertical axis.

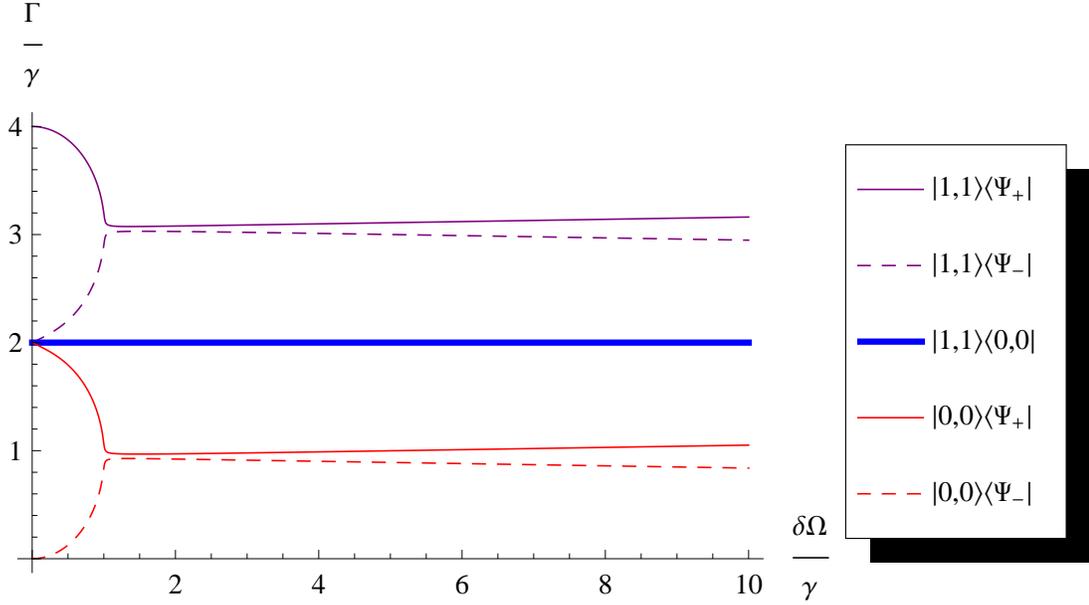


Figure 6.5: Decoherence rates of the (zeroth-order) non-stationary operators for two atoms in a zero-temperature environment at varying detuning and vanishing separation,  $R_{12} \ll \Omega_1, \Omega_2$ , with  $\gamma = \langle \Omega \rangle / 100$ . The legend indicates the matrix elements they approximately correspond to for small detunings (in the order they occur at the vertical axis) as to compare with Fig. 6.4.

In Figs. 6.3–6.4 we plot all relaxation rates associated with the two-atom system as a function of proximity, where  $\gamma$  is specifically the decoherence rate of a single isolated atom. For large separation the decay rates for  $|\Psi_{\pm}\rangle \equiv (|0,1\rangle \pm |1,0\rangle) / \sqrt{2}$  are  $1 + 1$  times  $\gamma$  (which would be  $N\gamma$  for  $N$  atoms), as the noise processes are independent and the decay rates are additive. Whereas at proximity they become 0 and  $N^2$  times  $\gamma$  for  $|\Psi_{-}\rangle$  and  $|\Psi_{+}\rangle$  respectively, as the noise processes are maximally correlated and display destructive and constructive interference. In Fig. 6.5 we plot all non-stationary decoherence rates associated with the two-atom system as a function of detuning. To achieve a dark state, the tuning of the two atoms

must be much better than the dissipation,  $\delta\Omega \ll \gamma$ , which counter-intuitively implies that weak-dissipation is not always desirable to preserve coherence. However, this condition makes more sense if thought of in another way: The dark state arises from the destructive interference of the emission from the two atoms. If the emission from each atom is characterized by center frequency  $\Omega_n$  and an emission line width  $\gamma$ , then the condition  $\delta\Omega \ll \gamma$  simply specifies that the emission lines of the atoms must overlap enough that their emissions are not distinguishable from one another. This allows the required destructive interference.

### 6.3.2.2 The Atomic Seesaw

One behavior which is qualitatively different from the closed-system evolution is the damped oscillations between the singly-excited states. More specifically for any initial state of the form

$$\rho_0 = \begin{array}{l} |0,1\rangle \\ |1,0\rangle \end{array} \begin{bmatrix} a + \delta & +i b \\ -i b & a - \delta \end{bmatrix} \begin{array}{l} \langle 0,1| \\ \langle 1,0| \end{array}, \quad (6.37)$$

with all positive coefficients, then in addition to the Bell state decay one will also have damped oscillations of the form

$$\begin{aligned} & [\delta \cos(f_1 t) - b \sin(f_1 t)] e^{-\gamma t} (|0,1\rangle\langle 0,1| - |1,0\rangle\langle 1,0|) \\ & + i [b \cos(f_1 t) + \delta \sin(f_1 t)] e^{-\gamma t} (|0,1\rangle\langle 1,0| - |1,0\rangle\langle 0,1|) \end{aligned} \quad (6.38)$$

which can oscillate from one excited state to the other excited state. But this happens very slowly, with the frequency

$$f_1 = 2\tilde{\gamma}_0 \frac{1 - \cos(\Omega R)}{R}, \quad (6.39)$$

for all temperatures. The oscillation is a timescale perturbation and should be present in conventional calculations using the RWA. This particular frequency vanishes for small separation; without our choice of regularization and renormalization, as detailed in Sec. 2.5.2.1, it would become increasingly large at proximity.

### 6.3.2.3 The Dark State

All stationary (and thus decoherence-free) states  $\rho_D$  of the open-system must satisfy the relation

$$\mathcal{L} \rho_D = 0, \quad (6.40)$$

and are thus right eigen-supervectors of the Liouvillian with eigenvalue 0. As the Liouvillian is not Hermitian, there is no trivial correspondence between the left and right eigen-supervectors. The super-adjoint of the master equation (see Sec. 2.4.1) time-evolves system observables and for closed systems can be contrasted

$$\mathcal{L}_0 \rho = -i[\mathbf{H}, \rho], \quad (6.41)$$

$$\mathcal{L}_0^\dagger \mathbf{S} = +i[\mathbf{H}, \mathbf{S}]. \quad (6.42)$$

The left eigen-supervector  $\mathbf{S}_D^\dagger$  corresponding to  $\rho_D$  must therefore satisfy

$$\mathcal{L}^\dagger \mathbf{S}_D = 0. \quad (6.43)$$

So for every stationary or decoherence-free state  $\rho_D$  there is a symmetry operator  $\mathbf{S}_D$  whose expectation value is a constant of the motion. The thermal state or reduced thermal state is such a state. In the limit of vanishing coupling strength, this state

is the familiar Boltzmann thermal state. One can check that the symmetry operator in this case is proportional to the identity and corresponds to  $\text{Tr}[\rho]$  being a constant of the motion.

For two resonant dipoles, with  $\Omega_n = \Omega$ , there is another stationary state in the limit of vanishing separation  $R_{12} = R$ . Because of degeneracy, any superposition of states

$$|\Psi\rangle = a_1 |1,0\rangle + a_2 |0,1\rangle, \quad (6.44)$$

is also an energy state and therefore annihilated by both  $\mathcal{L}_0$  and  $\mathcal{L}_0^\dagger$ . Further note that for vanishing separation, the noise processes arising from  $\mathbf{A}(\mathbf{R}_n, t)$  become exactly correlated and identical. Their contributions to the interaction Hamiltonian can then be collected into

$$\mathbf{H}_{I_1} + \mathbf{H}_{I_2} = (\sigma_{x_1} + \sigma_{x_2}) \mathbf{A} = \Sigma_x \mathbf{A}. \quad (6.45)$$

Next we note the equality

$$\Sigma_x |1,0\rangle = \Sigma_x |0,1\rangle, \quad (6.46)$$

so that for the Bell states

$$|\Psi_\pm\rangle \equiv \frac{1}{\sqrt{2}} \{|1,0\rangle \pm |0,1\rangle\}, \quad (6.47)$$

the noise adds destructively for  $|\Psi_-\rangle$  and constructively for  $|\Psi_+\rangle$ . Therefore  $|\Psi_-\rangle$  is a decoherence-free state (dark state) of the open system for vanishing separation and at resonance, regardless of coupling strength or temperature. And whereas  $|\Psi_-\rangle$  appears dark (sub-radiant),  $|\Psi_+\rangle$  appears bright (super-radiant). [Note that for anti-parallel dipoles, these roles will be reversed due to the anti-correlated noise.]

In this particular case the left and right eigen-supervectors are equivalent, and so it is the dark-state component  $\langle \Psi_- | \rho | \Psi_- \rangle$  which is a constant of the motion. However, unlike the thermal state, if the separation is no longer vanishing then this is not some perturbative limit of a stationary state but of a very long-lived state. The final constant of motion, which we have validated by analyzing the eigen-system of  $\mathcal{L}$ , corresponds to the coherence between the ground state and the dark state or  $\langle 0,0 | \rho | \Psi_- \rangle$ . Using these constants of motion, for two very close dipoles in a zero-temperature environment with initial state  $\rho_0$ , the system will relax into the state

$$\rho_1 = (1 - b) |0,0\rangle\langle 0,0| + b |\Psi_- \rangle\langle \Psi_-| + c |0,0\rangle\langle \Psi_-| + c^* |\Psi_- \rangle\langle 0,0| , \quad (6.48)$$

$$b \equiv \langle \Psi_- | \rho_0 | \Psi_- \rangle , \quad (6.49)$$

$$c \equiv \langle 0,0 | \rho_0 | \Psi_- \rangle , \quad (6.50)$$

to zeroth order in the system-environment coupling, whereupon the system has bipartite entanglement  $b$ .

While our (regularized) model is well behaved in the mathematical limit  $R \rightarrow 0$ , it is important to remember that physically the model is no longer valid for sufficiently small  $R$ . At small enough  $R$  other terms would come into play, including electrostatic interaction, and eventually the atoms would cease to even be distinct. We are assuming that this scale is much smaller than all other scales in our model (except perhaps the cutoff). This means that we can sensibly consider cases where  $R$  is small compared to the other parameters, but  $R$  cannot vanish completely.

Since the coefficients of our master equation are continuous in  $R$ , it is useful to consider  $R = 0$  to understand the limiting behavior as  $R$  becomes small. The

existence of the dark state we've discussed at  $R = 0$  means that this state will be almost completely dark when  $R$  is small; thus, any initial state  $\rho_0$  will first relax approximately into the state given in Eq. (6.48) within the ordinary relaxation timescale  $\gamma$ , and then on a much longer relaxation timescale  $\tau$ , where roughly  $1/\tau \approx \gamma(\Omega R)^2$  for small  $R$ , the system will fully thermalize. However, this expression for the dark state is only to zeroth-order in the system-environment coupling. In order to understand the subsequent final state of decay one needs the second-order asymptotics that we discuss in Sec. 6.3.2.5.

Finally we would note that this “dark state” is a very general feature of resonant multipartite systems with similar linear couplings to a shared environment. One can rather easily work out that for a pair of resonant linear oscillators with these same noise correlations the sum mode is thermalized, and the difference mode is decoherence free for vanishing separation. The separation dependence of the entanglement dynamics of two resonant oscillators was considered in Ref. [105] and Sec. 6.4, while that of (effectively) two very close oscillators was considered in Ref. [122, 123].

#### 6.3.2.4 $N$ -Atom dark and bright states

The sub-radiant dark state achieves destructive interference in the environmental noise (and thus little-to-no emission) while the bright state achieves constructive interference in the noise (and thus near-maximal emission). For the super-radiant bright state one essentially couples the system to  $N$  copies of the same noise process from  $\mathbf{A}(\mathbf{R}, t)$  and therefore the super-radiant emission rate *can* be proportional to

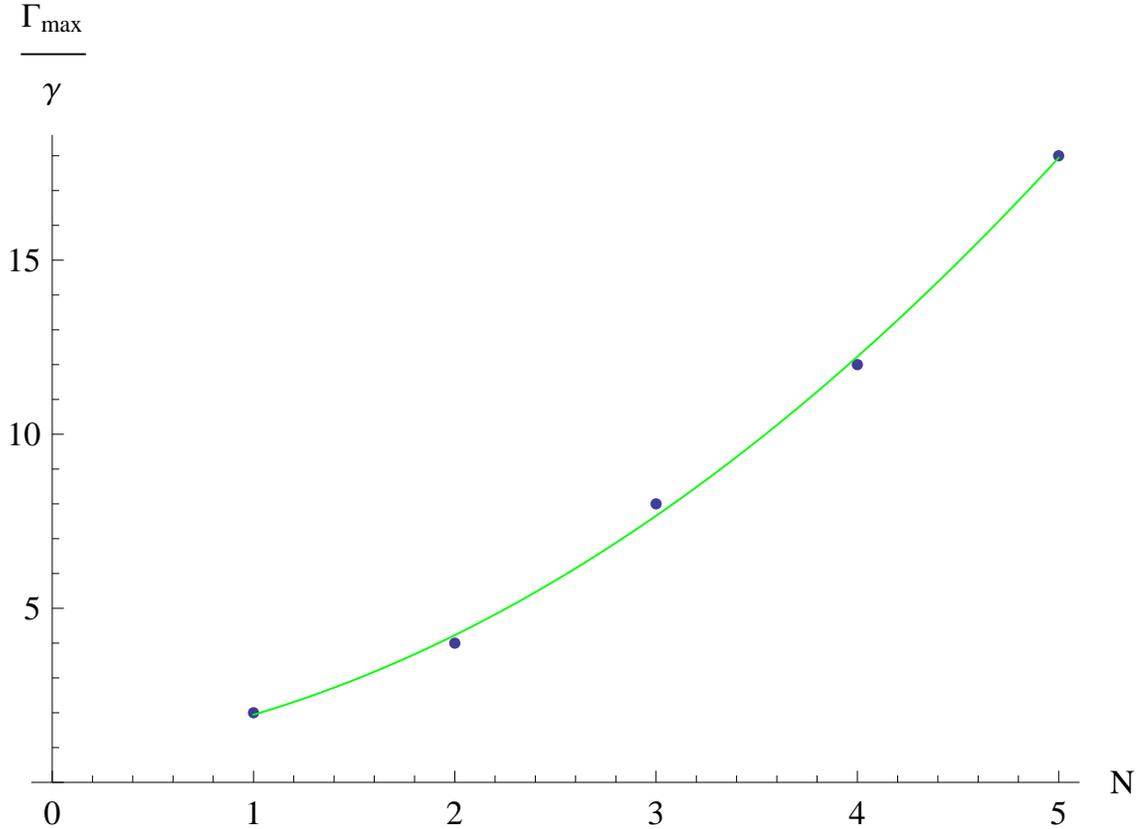


Figure 6.6: Maximal (over states) second-order decay rates as a function of the number of atoms  $N$ , at zero temperature and in close proximity. The solid curve denotes the best quadratic fit and has a corresponding p-value of 2.4%, which is fairly significant in corroborating an  $N^2$  dependence.

$N^2$ . An  $N^2$  dependence does appear the case as we demonstrate in Fig. 6.6. The emission rate is (perturbatively) determined by the noise correlation (the square of the noise process). Both results differ having from  $N$  independent noise processes where one can simply add the  $N$  independent noise correlations which results in an emission rate at most proportional to  $N$ .

Following the previous approach, we define a *proper dark state* as an atomic

state annihilated by  $\mathcal{L}_0$  and  $\mathbf{H}_I$  regardless of the state of the environment. Let us consider an assembly of  $N$  resonant dipoles at close proximity. We first note that the superposition

$$|\Psi\rangle = \sum_{\sum s_n=S} a_{s_1, s_2, \dots, s_N} |s_1, s_2, \dots, s_N\rangle, \quad (6.51)$$

of energy states with the same total excitement  $S$  is also an energy state and therefore annihilated by  $\mathcal{L}_0$ . Defining the collective spin operator

$$\Sigma_x = \sum_n \sigma_{x_n}, \quad (6.52)$$

such that the interaction Hamiltonian can be expressed

$$\mathbf{H}_I = \Sigma_x \mathbf{A}(\mathbf{R}_n); \quad (6.53)$$

a proper dark state must then satisfy  $\Sigma_x |\Psi_-\rangle = 0$  and will be decoherence free. For  $N = 2$  this is the familiar Bell state that we've already labeled  $|\Psi_-\rangle$ .

In considering large  $N$  the structure is essentially just what was studied by Dicke [50], so following that approach we define collective  $y$  and  $z$  spin operators  $\Sigma_y$  and  $\Sigma_z$  as well as raising and lowering operators  $\Sigma_+$  and  $\Sigma_-$ , analogously to Eq. (6.52), as well as  $\Sigma^2 = \Sigma_x^2 + \Sigma_y^2 + \Sigma_z^2$ . And we can note that the free Hamiltonian for the atoms only differs from  $\Sigma_z$  by a multiple of the identity, so all the eigenstates of that Hamiltonian are also eigenstates of  $\Sigma_z$ . A basis for the Hilbert space of the system can be specified by the eigenstates of  $\Sigma^2$  with eigenvalues  $j(j+1)$  and  $\Sigma_z$  with eigenvalues  $m$  (though for  $N > 2$  there will be degeneracy, so that additional quantum numbers are needed to identify a specific state). The dark state we seek must then satisfy  $\Sigma_z |\Psi_-\rangle = m |\Psi_-\rangle$  and  $\Sigma_x |\Psi_-\rangle = 0$ . As the discussion in [50]

implies, only states with  $j = 0$  and  $m = 0$  can satisfy these requirements simultaneously. Such states only occur when  $N$  is even, and that set of states has dimension  $N! / [(N/2 + 1)!(N/2)!]$ . These are also the dark states in the RWA, as they are in the null space of both  $\Sigma_+$  and  $\Sigma_-$ . For  $N = 4$  these states take the form

$$|\Psi_-\rangle = a_1(|0,0,1,1\rangle + |1,1,0,0\rangle) + a_2(|0,1,0,1\rangle + |1,0,1,0\rangle) + a_3(|0,1,1,0\rangle + |1,0,0,1\rangle), \quad (6.54)$$

$$0 = \sum_n a_n, \quad (6.55)$$

where every pair in parenthesis is spin-flip symmetric. One can easily check that any such state is annihilated by  $\Sigma_x$ .

More generally we define an *improper dark state* as one only annihilated by  $\mathcal{L}$  and not  $\mathbf{H}_I$  (i.e., stationary in the coarse-grained open-system dynamics but not in the full closed system dynamics), thus being dependent upon the state of the environment and even the coupling strength. In the simplest case we can consider the zero-temperature environment. For the second-order dynamics, upward transitions are automatically ruled out from the lack of thermal activation. The only term that could lead to population of higher excitation states is the second term in Eq. (??), which vanishes at  $T = 0$ . Rather than investigating the master equation, we can then simply demand that the lowest-order decay transitions are vanishing, meaning that if  $|\Psi_-^S\rangle$  has total excitation  $S$ , then  $\langle S' | \Sigma_x | \Psi_-^S \rangle = 0$  for all  $S' \leq S$  lesser and equally excited states. We can also state this in terms of the collective spin operators we have defined, by saying that we demand that  $|\Psi_-\rangle$  is an eigenstate of  $\Sigma_z$  with eigenvalue  $m$ , and that all matrix elements onto states with lower  $m'$

values must vanish. Since  $\Sigma_x = \frac{1}{2}(\Sigma_+ + \Sigma_-)$ , we know that there will be non-vanishing matrix elements onto states with  $m' = m - 1$  unless  $m = -j$ . So any state with  $m = -j$  is an imperfect dark state at zero temperature, and there are  $N!(2j + 1)/[(N/2 + j + 1)!(N/2 - j)!]$  such states [50]. Interestingly, in the RWA such states (when combined with a vacuum field) are also stationary states but of the closed-system dynamics. For  $N = 3$  and at zero temperature, all such dark states can be expressed

$$|\Psi_-\rangle = a_1 |1,0,0\rangle + a_2 |0,1,0\rangle + a_3 |0,0,1\rangle, \quad (6.56)$$

$$0 = \sum_n a_n, \quad (6.57)$$

for weak coupling to the field. These dark states also exist for positive temperature, but they take on a different form.

### 6.3.2.5 Asymptotics

To zeroth order in the system-environment interaction, the asymptotic steady state is Boltzmann, which can be expressed

$$\rho_T = \prod_n \rho_{T_n}, \quad (6.58)$$

$$\rho_{T_n} \equiv \frac{1}{2} \left[ 1 - \tanh\left(\frac{\Omega_n}{2T}\right) \sigma_{z_n} \right], \quad (6.59)$$

in terms of Pauli matrices. The asymptotic state of the second-order master equation is consistent with this result and can additionally provide some of the second-order corrections  $\delta\rho_T$  via the constraint

$$\mathcal{L}_0\{\delta\rho_T\} + \mathcal{L}_2\{\rho_T\} = 0. \quad (6.60)$$

These will specifically be the off-diagonal or non-stationary perturbations. In general, to find the second-order corrections to the diagonal elements of the density matrix one needs to compute contributions from the fourth-order Liouvillian (see Sec. 4.2).

Following Sec. 4.5, for non-vanishing interaction with the environment the off-diagonal elements of the asymptotic state match the reduced thermal state

$$\boldsymbol{\rho}_\beta \equiv \frac{1}{Z_C(\beta)} \text{Tr}_E \left[ e^{-\beta(\mathbf{H} + \mathbf{H}_E + \mathbf{H}_I)} \right], \quad (6.61)$$

where  $Z_C(\beta)$  is the partition function of the system and environment with non-vanishing interaction. We will refer to  $\boldsymbol{\rho}_\beta$  as the thermal Green's function; this function can be expanded perturbatively in the system-environment coupling as

$$\boldsymbol{\rho}_\beta = \frac{1}{Z_0(\beta)} e^{-\beta \mathbf{H}} + \boldsymbol{\delta\rho}_\beta + \dots, \quad (6.62)$$

where  $Z_0(\beta)$  is the partition function of the free system. The second-order corrections are given by

$$\langle \omega_i | \boldsymbol{\delta\rho}_\beta | \omega_j \rangle = \sum_{nmk} \frac{R_{ijk}^{nm}}{Z_0(\beta)} \langle \omega_i | \boldsymbol{\sigma}_{x_m} | \omega_k \rangle \langle \omega_k | \boldsymbol{\sigma}_{x_n} | \omega_j \rangle. \quad (6.63)$$

All terms with  $\omega_i = \omega_j$  are zero, so that this expression gives no correction to the diagonal elements of the density matrix. Otherwise, the (non-resonant) off-diagonal coefficients are given by

$$R_{ijk}^{nm} \Big|_{\omega_i \neq \omega_j} \equiv \text{Im} \left[ e^{-\beta\omega_k} \frac{A_{nm}(\omega_{ik}) - A_{nm}(\omega_{jk})}{\omega_i - \omega_j} + \frac{e^{-\beta\omega_i} A_{mn}(\omega_{ki}) - e^{-\beta\omega_j} A_{mn}(\omega_{kj})}{\omega_i - \omega_j} \right], \quad (6.64)$$

with the free ground-state energy set to zero. These coefficients agree perturbatively with those from Eq. (6.60). Because such an expansion is inherently secular in  $\beta$ , it

is valid only at a sufficiently high temperature such that the perturbations are small compared to the smallest Boltzmann weight,

$$\frac{\gamma}{\Omega} \ll e^{-\beta(\Omega_n + \Omega_m)} = \left( \frac{\bar{n}(\Omega_n, T)}{\bar{n}(\Omega_n, T) + 1} \right) \left( \frac{\bar{n}(\Omega_m, T)}{\bar{n}(\Omega_m, T) + 1} \right). \quad (6.65)$$

The expansion does not apply at lower temperatures. Reliability of the expansion at higher temperature suggests that the diagonal corrections to the asymptotic state must be suppressed there.

Since neither the second-order master equation nor the perturbative expansion of the thermal Green's function can give the full low-temperature solution, including diagonal corrections, it appears that in general this will require the fourth-order master equation coefficients. However, at zero temperature the thermal state is simply the ground state of the total system-environment Hamiltonian. This ground state can be calculated perturbatively from the Hamiltonian as usual in a closed system, and the zero-temperature reduced thermal state follows directly. All three of these formalisms are fully consistent as shown in Sec. 4.5. At zero temperature the off-diagonal second-order corrections to the asymptotic state are still of the form given in Eqs. (6.63) and (6.64), with the coefficients evaluated in the limit  $\beta \rightarrow \infty$ . The diagonal (and resonant) perturbations are given by

$$\lim_{\beta \rightarrow \infty} R_{ijk}^{nm} \Big|_{\omega_i = \omega_j} = \lim_{\beta \rightarrow \infty} \text{Im} \left[ e^{-\beta\omega_k} \frac{d}{d\omega_i} A_{nm}(\omega_{ik}) + e^{-\beta\omega_i} \frac{d}{d\omega_i} A_{mn}(\omega_{ki}) \right], \quad (6.66)$$

where only a handful of terms are non-vanishing. We note that the expression inside the limit in Eq. (6.66) has both the correct low and high-temperature limits, so it may be roughly correct for all temperatures, but we have yet to fully investigate the fourth-order master equation.

For most regimes the second-order thermal state can now be expressed entirely in terms of the second-order master equation coefficients and limits thereof, therefore we can say that the environmentally induced correlations do vanish for large separations with a power-law decay like  $1/R$  and  $1/R^2$ .

### 6.3.2.6 Entanglement of Two Atoms

Now we will consider the bipartite entanglement between any two atoms, labeled  $n$  and  $m$  in a common quantum field. We begin with some remarks that apply to any system of two qubits. We focus on the late-time dynamics of this system; we will compute the reduced density matrix for their asymptotic state  $\rho_{nm}$  and derive the asymptotic value of entanglement between these two atoms. We will see that this computation will also allow us to show that all entangled initial states become disentangled at a finite time.

To quantify the bipartite entanglement we will use Wootters' concurrence function [154], which is a monotone with a one-to-one relationship to the *entanglement of formation* for two qubits. The concurrence is defined as

$$C(\rho_{nm}) = \max \{0, \underline{C}(\rho_{nm})\} \quad (6.67)$$

$$\underline{C}(\rho_{nm}) = \sqrt{\lambda_1} - \sqrt{\lambda_2} - \sqrt{\lambda_3} - \sqrt{\lambda_4} \quad (6.68)$$

where  $\lambda_1 \geq \lambda_2 \geq \lambda_3 \geq \lambda_4$  are the eigenvalues of the matrix

$$\rho_{nm} \tilde{\rho}_{nm} \equiv \rho_{nm} (\sigma_{y_n} \sigma_{y_m} \rho_{nm}^* \sigma_{y_n} \sigma_{y_m}) , \quad (6.69)$$

which are always non-negative. A two-qubit state is entangled if and only if  $\underline{C} > 0$ .

It is important to note that  $\underline{C}(\rho)$  is a continuous function of the matrix elements

of  $\rho$  (since the eigenvalues of a matrix can be written as a continuous function of the matrix elements [143]); this then implies that any density matrix with  $\underline{C} < 0$  lies in the interior of the set of separable states (with every sufficiently nearby state also separable), while states with  $\underline{C} > 0$  lie in the interior of the set of entangled states. States with  $\underline{C} = 0$  are separable but include states that lie on the boundary between the two sets, infinitesimally close to both entangled states and the interior of the separable states. Any separable pure state lies on this boundary [82].

Given the late-time asymptotic state of two atoms  $\rho_{nm}$ , one can easily compute the asymptotic entanglement from  $\underline{C}(\rho_{nm})$ . Based on the preceding paragraph, however, we know that this will also tell us something qualitatively about the late-time entanglement dynamics. If  $\underline{C}(\rho_{nm}) < 0$  then (assuming only continuous evolution in state space) every initial state must become separable at some finite time as it crosses into the set of separable states. Likewise, if  $\underline{C}(\rho_{nm}) > 0$  then all initial states lead to entanglement at sufficiently late time and any sudden death of entanglement must be followed by revival. In models such as ours which have a unique asymptotic state, it is only when  $\underline{C}(\rho_{nm}) = 0$  that this qualitative feature of the late-time behavior will depend on the initial state, with some entangled states remaining separable after some finite time and others becoming disentangled only asymptotically in the limit  $t \rightarrow \infty$  as in [159, 58]. Previous work has pointed out that the late-time entanglement dynamics can be determined by the asymptotic state in this way [160, 42], with Yu and Eberly [161] discussing the role of  $\underline{C}$  in predicting sudden death. In Refs. [161, 42] the authors consider models with multiple steady states, which introduces additional dependence on initial conditions.

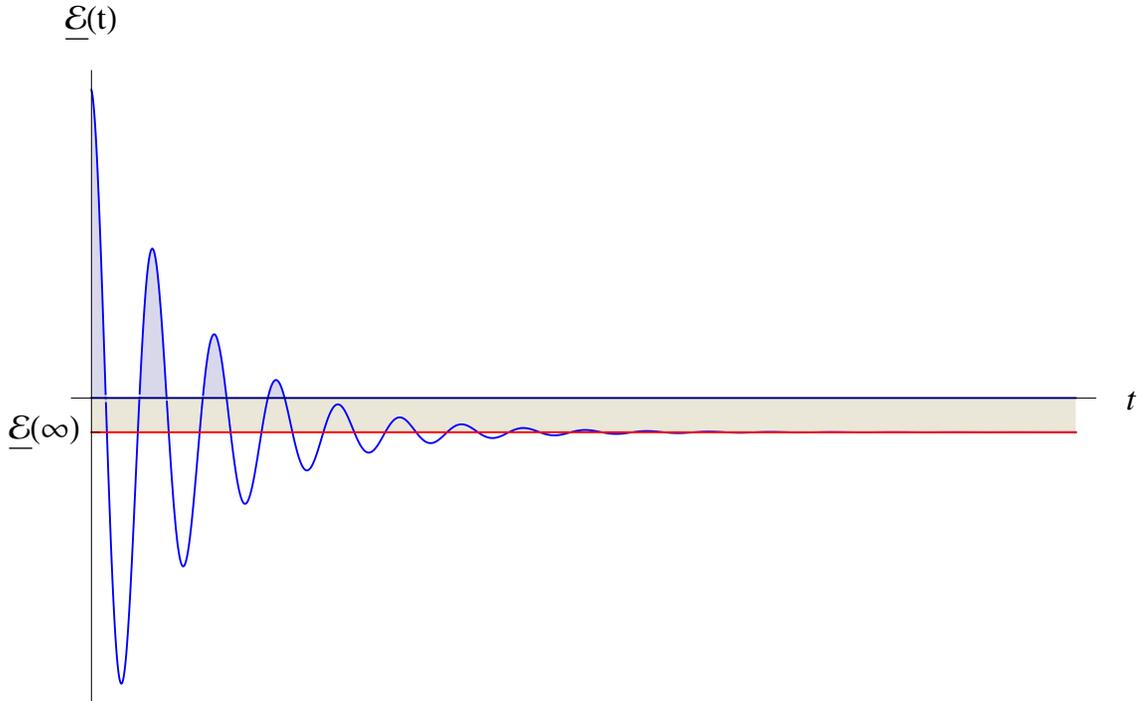


Figure 6.7: Qualitative plot of an (unmaximized) entanglement function showing dynamics including entanglement sudden death, revival, and asymptotic separability.

It can be seen that none of the foregoing discussion is specific to the concurrence; it would apply to any quantity that is a continuous function of the density matrix, takes on negative values for some separable states, and is an entanglement monotone when non-negative. If we have such an unmaximized entanglement function  $\underline{\mathcal{E}}$  from which an entanglement monotone can be defined by  $\mathcal{E} = \max\{0, \underline{\mathcal{E}}\}$ , then we can use it just as we have discussed using  $\underline{\mathcal{C}}$  above. As illustrated qualitatively in Fig. 6.7, entanglement sudden death occurs because the unmaximized entanglement function asymptotes towards a negative value, whereas any entanglement monotone (derived from  $\underline{\mathcal{E}}$  or otherwise) cannot go below zero, leading to an

abrupt sudden death of entanglement when  $\underline{\mathcal{E}}$  becomes negative.

An important point arises from the facts we have noted about  $\underline{C}$  and separability: At sufficiently low temperature the  $\mathcal{O}(\gamma)$  corrections to the asymptotic state are required to calculate the sign of  $\underline{C}(\rho_{nm})$  and, therefore, even the qualitative features of late-time entanglement dynamics. At zero temperature, the zeroth-order asymptotic state is simply the ground state of the system, assuming no degeneracy at the ground energy, according to Eq. (6.58). So the zeroth-order asymptotic state is a pure separable state. This means that it lies on the boundary between the entangled and separable states, and in general some initial states will suffer sudden death while others will not, as depicted in Fig. 6.8a. But any non-zero perturbation, however small, can lead to asymptotic entanglement or can place the asymptotic state in the interior of the separable states, implying sudden death for all initial conditions. Fig. 6.8b shows each of these situations. Thus, knowing only the zeroth-order asymptotic state one can make no meaningful prediction about late-time entanglement dynamics, and this will always be the case when using the rotating-wave approximation, because it neglects the second-order corrections to the asymptotic state [62]. This makes calculations such as [58] incapable of correctly predicting these features.

At positive temperature the zeroth-order asymptotic state is simply the Boltzmann state  $\rho_T$ , which lies in the interior of the set of separable states [160], and

$$\rho_T \tilde{\rho}_T = \frac{e^{-(\Omega_n + \Omega_m)/T}}{Z_0(T)^2} \mathbf{1}, \quad (6.70)$$

so that  $\underline{C}(\rho_T) = -2e^{-(\Omega_n + \Omega_m)/(2T)}/Z_0(T)$ . The  $\mathcal{O}(\gamma)$  corrections to  $\rho_{nm}$  will yield

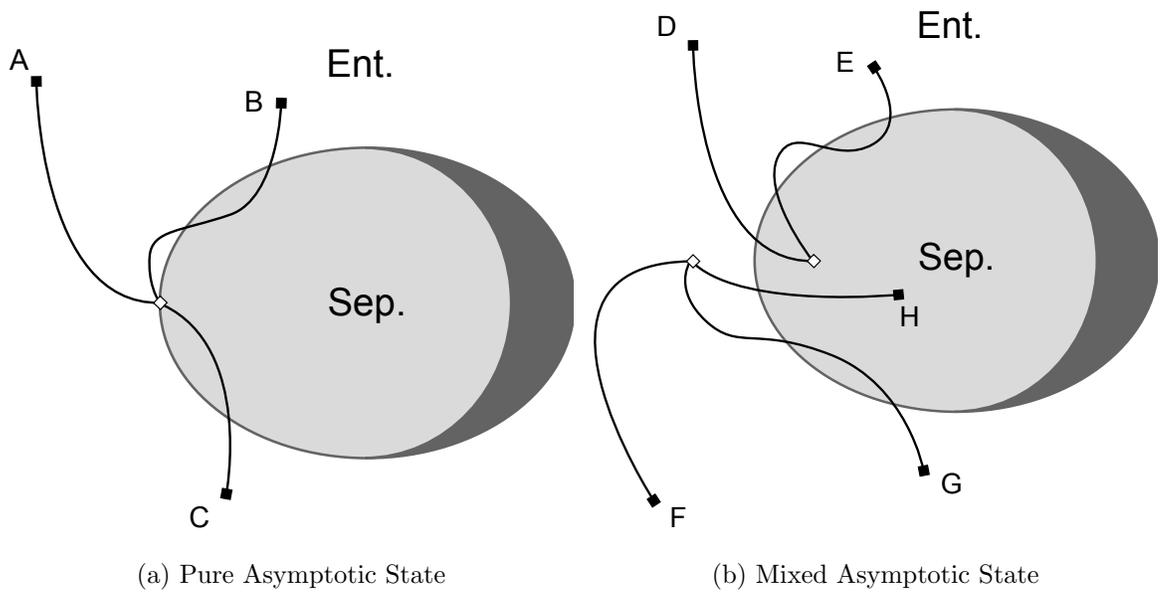


Figure 6.8: A schematic representation of the evolution in state space. The white area represents entangled states ( $\underline{C} > 0$ ), while the gray areas represent separable states  $\underline{C} \leq 0$  with the dark gray representing states with  $\underline{C} = 0$ . The asymptotic state is represented by  $\diamond$ , while initial states are represented by  $\blacksquare$ . In (a) we have the asymptotic state on the boundary as in the zeroth-order at  $T = 0$ . In (b) two scenarios are shown that can arise from a small perturbation moving the asymptotic state off the boundary, into the interior of one of the two sets. This illustrates how such a perturbation qualitatively changes the late-time entanglement dynamics.

order  $\mathcal{O}(\gamma)$  corrections to  $\rho_{nm}\tilde{\rho}_{nm}$ . Then simply from the definition of  $\underline{C}$  we know that so long as the temperature is sufficiently high that Eq. (6.65) is satisfied the corrections to  $\rho_{nm}$  will cause at most  $\mathcal{O}(\gamma)$  corrections to  $\underline{C}(\rho_{nm})$  so that it must remain negative. Consequently, the second-order asymptotic state still lies in the interior of the separable states, and all initial states will suffer entanglement sudden death at sufficiently late times. For lower temperatures it does not appear that the sign of  $\underline{C}(\rho_{nm})$  can be generically predicted, and one must find the late-time asymptotic state for the specific system in question which generally requires terms from the fourth-order master equation.

Returning to the specifics of the particular model examined in this paper, from Eq. (6.63) it can be readily seen that the atoms are correlated in the asymptotic state at all temperatures, and from our second-order coefficients these correlations experience power-law decay with separation. However, we find based on Eqs. (6.62), (6.63), and (6.64) that when the high-temperature expansion is valid (according to Eq. (6.65)) the asymptotic state has  $\underline{C}(\rho_{nm}) < 0$ . At zero temperature, Eqs. (6.64) and (6.66) also give  $\underline{C}(\rho_{nm}) < 0$ . In both cases the asymptotic state lies in the interior of the separable states, and all initial states become separable permanently after some finite time. With this property upheld for zero and high temperatures, we suspect this to be true at all temperatures, making entanglement sudden death a generic feature which happens in every case in this model. Of course, as discussed in Sec. 6.3.2.3, for closely spaced atoms there can be a dark state, so that entanglement persists over a long timescale before eventually succumbing to sudden death. It should also be noted that, while this examination of the asymptotic behavior tells

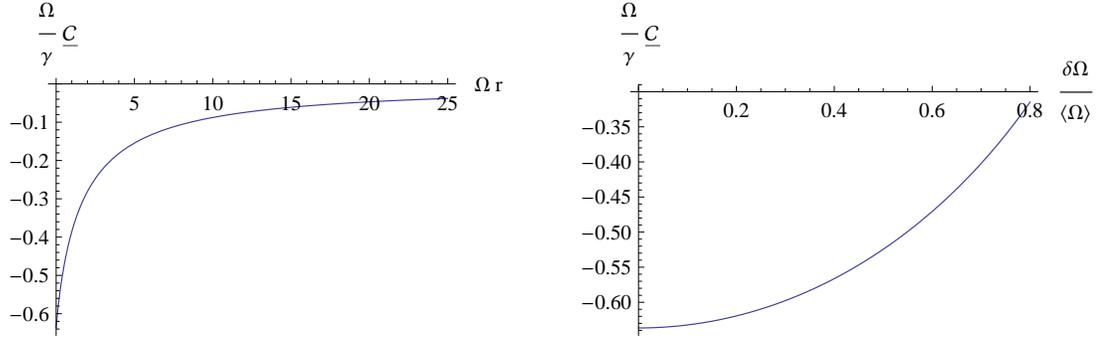


Figure 6.9: Unmaximized concurrence for two resonant atoms at various separation distance (left) and two close atoms at various frequency detunings (right) at zero temperature and for  $\gamma = \langle \Omega \rangle / 100$ .

us that entanglement always remains zero after some finite time, we do find  $\mathcal{O}(1)$  sudden death and revival of entanglement at earlier times for some initial states (similar to [58]).

In Fig. 6.9 we plot  $\underline{C}$  as it varies with separation distance and frequency detuning. As a consistency check we also calculated the logarithmic negativity and found it to be consistent with the concurrence to second order. The behavior of the entanglement is markedly different from that of two oscillators in a field, as we will cover in Sec. 6.4. For two oscillators, there can be asymptotic entanglement if they are held very close and near enough to resonance with each other. Separation and detuning then causes the entanglement monotones to decay away. For the two-atom case studied here asymptotic entanglement does not exist, and resonant tuning with proximity will only exacerbate the problem. Permanent sudden death of entanglement occurs because the unmaximized entanglement functions can trend

below zero within a finite amount of time and without the need of any asymptotic limit. We would finally note that while the concurrence function does appear to be increasing for large detuning, the parameters drift outside of the weak-coupling regime as one of the frequencies becomes very small.

## 6.4 Harmonic Atoms

The results of Chapter 3 have given us the tools to properly examine the entanglement dynamics of multiple (local) oscillators residing in the same scalar field. One can think of this model as analogous to the previous model of parallel and dipolar 2-level atoms in the electromagnetic field. With our non-perturbative formalism we are able to generalize some of the results of Lin and Hu [105] who have long investigated this problem at resonance and for weak coupling. However, to some degree we face many of the same difficulties which Lin and Hu faced, as the influence kernels of relativistic fields have a rather complicated structure related to their causal nature. Formally this problem is exactly solvable, but for computational reasons we do ultimately resign ourselves to separate study of the weak-coupling and close-proximity regimes. Off-resonant detuning is not problematic in our formalism.

### 6.4.1 System Modes in a Common Environment

Specifically we consider identical local oscillators (Unruh-deWitt detectors)  $X_i$  in a shared environment (a massless scalar field), but at different locations  $\mathbf{R}_i$  as considered in Ref. [105]. The coupling is local and linear between the system

positions (more typically momenta) and field operator. The damping kernel for this model was given in Sec. 6.2.2.1, and can be expressed

$$\tilde{\gamma}_{ij}(\omega) = 2\gamma_0 \operatorname{sinc}(R_{ij}\omega), \quad (6.71)$$

$$\hat{\gamma}_{ij}(s) = \gamma_0 \frac{1 - e^{-R_{ij}s}}{R_{ij}s}, \quad (6.72)$$

in the Fourier and Laplace domains respectively, where  $\mathbf{R}_{ij} = \mathbf{R}_i - \mathbf{R}_j$ . The Markovian limit is reached here when the environment is at high temperature and the detectors are taken to be very far apart, in which case one recovers  $\gamma_0$  as the damping rate of the individual system oscillators, or very close together, in which case one has effectively local cross-damping. At finite temperature it is well known that QBM requires a finite cutoff regulator for Ohmic coupling to the environment, and this is also the case here. Note that distance variation naturally takes the form of an Ohmic regulator in the limit of small separations. Following Sec. 6.3 which considered the same environment, we will not evaluate the correlations more precisely than to some smallest scale  $r_0$  in the sense that we take

$$\lim_{\mathbf{R}_i \rightarrow \mathbf{R}_j} R_{ij} = r_0 \equiv \Lambda^{-1}, \quad (6.73)$$

to provide a natural cutoff of  $\Lambda$ . Essentially this dictates that the detector is not truly local in the field.

For a pair of identical detectors  $X_1$  and  $X_2$ , the dynamics naturally decompose into the sum and difference  $X_{\pm} = X_1 \pm X_2$  under the influence of the kernels

$$\hat{\gamma}_{\pm}(s) = \hat{\gamma}_{11}(s) \pm \hat{\gamma}_{12}(s), \quad (6.74)$$

for both dissipation and noise (by the FDR), so when the two detectors are very

close their difference  $X_-$  experiences vanishing dissipation. Therefore their difference comprises the decoherence-free subspace, or *dark state*, but only in the limit of minimal separation so that their environments appear identical and are perfectly correlated. This is essentially the regime considered in Ref. [122] for the entanglement dynamics of two oscillators in a common environment. But more generally, for small separations the sum  $X_+$  relaxes quickly and then on a much longer timescale the difference  $X_-$  will also thermalize. The resultant asymptotic state, which is Gaussian in the sum and difference, can also be entangled to higher-order in the coupling.

More generally Ref. [105] considered finite separation distances and determined the oscillators to be asymptotically entangled when near the cutoff scale in separation. Our more general formalism allows for non-resonant detectors which cannot be transformed into a pair of individual quantum Brownian oscillators, and so we can determine how resonant the detectors must be for this asymptotic entanglement to ensue.

## 6.4.2 Regulation and Integration

The non-perturbative late-time covariance (3.178) evaluated with our exponential cutoff-regulator (6.72), is exceptionally oscillatory and does not lend it self to well-behaved numerics, especially in the near regime. In evaluating the late-time covariance, the exact regulator in the Laplace domain

$$\chi(z) = \frac{1 - e^{-z}}{z}, \quad (6.75)$$

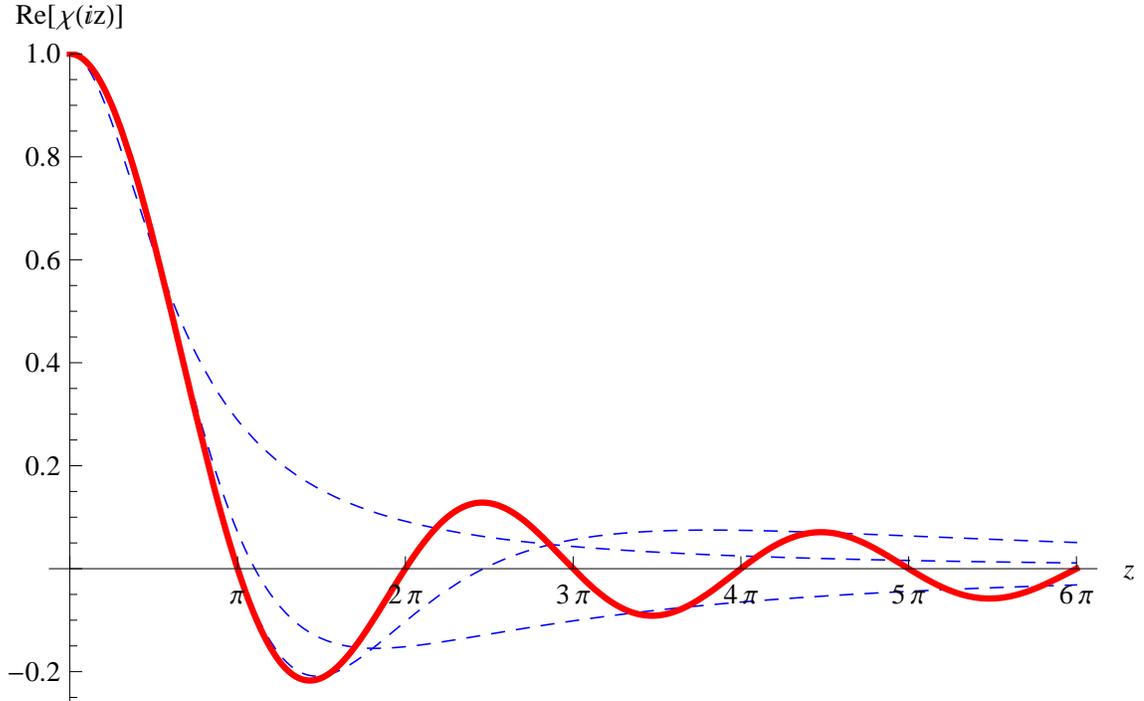


Figure 6.10:  $\cdots$  The first three  $\chi_{[n/n+1]}(z)$  Padé approximants of the  $\bullet$  exact regulator.

must be evaluated at imaginary arguments, and it becomes both oscillatory and asymptotically  $\mathcal{O}(1/\imath z)$ . Therefore the appropriate small- $R$  approximations of this regulator are the  $\chi_{[n/n+1]}(z)$  Padé approximants. These are the best local (small- $z$  expansion) and rational (ratio of polynomials) approximations which are also  $\mathcal{O}(1/\imath z)$ . We plot the first three such approximants in Fig. 6.10. One can see that they are good approximations up to  $R \sim n\pi/2\Omega$ , similar to the Taylor series convergence for sinusoidal functions. In this work we primarily have considered the first-order Padé approximant

$$\chi_{[0/1]}(z) = \frac{1}{1 + \frac{z}{2}}, \quad (6.76)$$

which is also an Ohmic regulator for small separations. This local approximation is most accurate in the high-cutoff and small- $R$  regime, which is precisely where we need to investigate more carefully, but it also has the correct asymptotics. Resultant calculations can be obtained exactly and are very well behaved. In addition to numeric solutions and analytic small- $R$  solutions, we also utilize analytic small- $\gamma_0$  solutions using weak-coupling perturbation of Eq. (3.149).

### 6.4.3 Entanglement Dynamics

In calculations of entanglement we consider both the Peres-Horodecki criterion  $\Sigma$  [140] the logarithmic negativity  $E_{\mathcal{N}}$  [148] as a consistency check. In all graphs, positive quantities denote entanglement and negative quantities separability. We consider the zero-temperature regime only as it emphasizes entanglement.

Our generic analysis of entanglement dynamics involves two factors: (1) the relevant timescales for decoherence and (2) *unmaximized* entanglement monotones of the asymptotic state [41]. In this linear model, decoherence comes about primarily from the thermal covariance  $\sigma_T$  smearing away quantum interference in the Wigner function and secondarily via dissipation blindly erasing all details of the initial state. A more thorough explanation of the general evolution is given in Chapter 3.

Traditionally defined entanglement monotones are not sufficient to give a cursory analysis of entanglement sudden death as they do not distinguish between separable states. Given some operationally defined entanglement monotone  $\mathcal{E} = \max[0, \underline{\mathcal{E}}]$ , so that  $\mathcal{E}$  is zero for all separable states, then the unmaximized func-

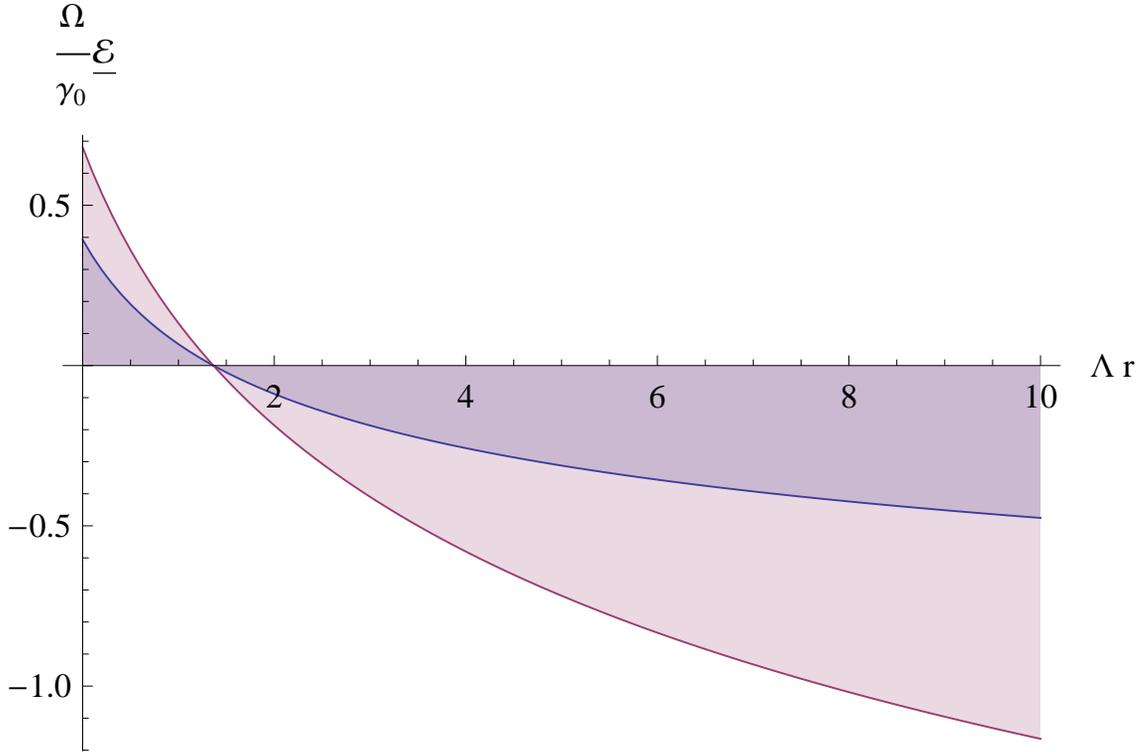


Figure 6.11: The asymptotic (unmaximized) entanglement monotones of two resonant oscillators as a function of separation where  $\gamma_0 = \Omega/2$ .

tion  $\underline{\mathcal{E}}$  is more useful if its evolution is continuous. As we qualitatively plot in Fig. 6.7, entanglement sudden death occurs because the unmaximized entanglement monotone asymptotes towards a negative value, whereas the traditionally defined entanglement monotone does not asymptote towards zero. Therefore, given that decoherence in linear models is fairly well understood, we will focus primarily upon analysis of the asymptotic (unmaximized) entanglement.

### 6.4.3.1 Near Detectors

In agreement with Ref. [105] we find the resonant oscillators to be asymptotically entangled when near the cutoff scale. Fig. 6.11 denotes the asymptotic entanglement measures of the resonant oscillators on the near cutoff scale where asymptotic entanglement can emerge. It is curious that, despite the same environmental correlations and despite the emergence of a dark state under the same conditions, for a pair of two-level atoms the possibility of asymptotic entanglement does not emerge (at second-order) for perfect cross-correlations (see Fig. 6.9). Moreover the behavior is rather opposite to that here, with the unmaximized entanglement monotonous becoming more negative at proximity.

### 6.4.3.2 Off-Resonant Detectors

We know that asymptotic entanglement will ensue when the two oscillators are very close together and at resonance. Therefore the question arises, how close and how at resonance must the two oscillators be to have asymptotic entanglement. The first question has been answered: to have asymptotic entanglement, the oscillators must be extremely close together, on the order of the inverse cutoff. The second question requires a more general multivariate treatment, as we can provide in our formalism.

Here we take the two oscillators to be very close in position but with different frequencies  $\Omega_{\pm} = \Omega_0 \pm \delta\Omega$ . Expressed in this manner with average frequency  $\Omega_0$  and difference  $2\delta\Omega$ , we find the asymptotic entanglement to be very much insensitive to

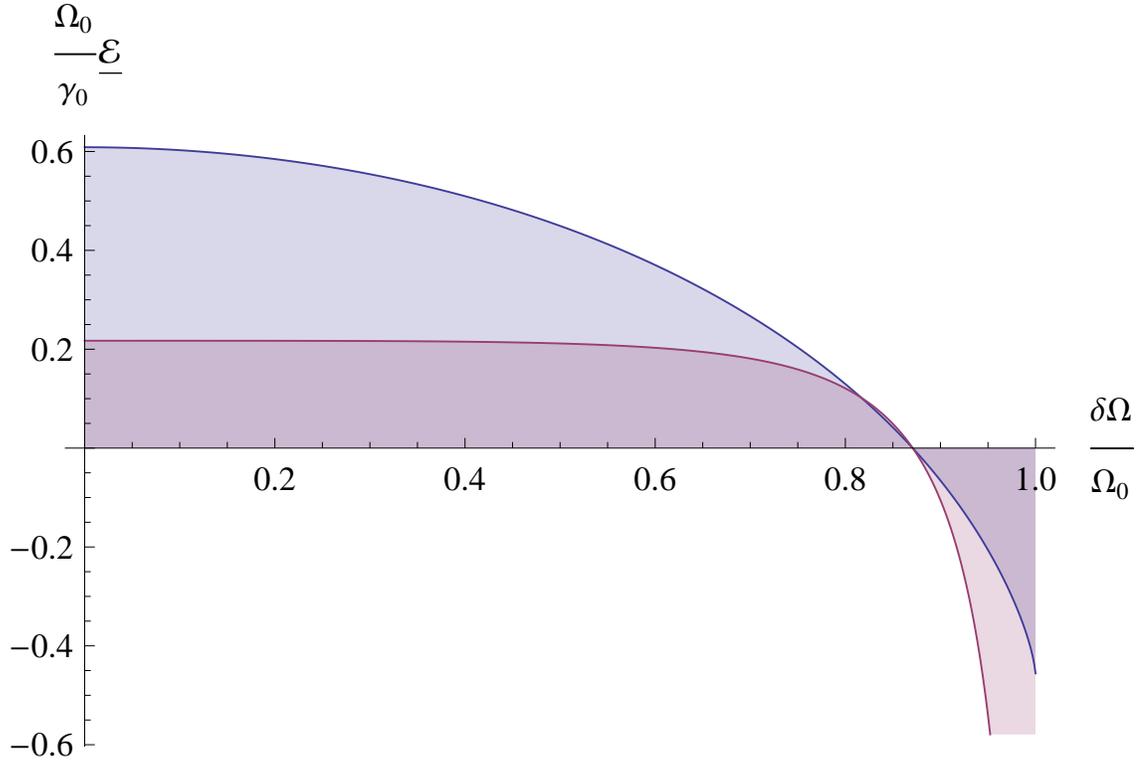


Figure 6.12: The asymptotic entanglement measures of two oscillators as a function of detuning where  $\gamma_0 = \Omega_0/2$ .

$\delta\Omega$ . Fig. 6.12 denotes the asymptotic entanglement measures of the two oscillators for a moderate coupling strength. The stronger the environmental interaction is, the more asymptotic entanglement ensues and the less resonance sensitivity there is. But even for weak interactions, resonance does not appear to be a very stringent criteria for asymptotic entanglement, at least nothing like nearness.

#### 6.4.4 Sub and Super-radiance

We know that the dark and bright states emerge the two oscillators are very close together and at resonance. Therefore the question arises, how close and how

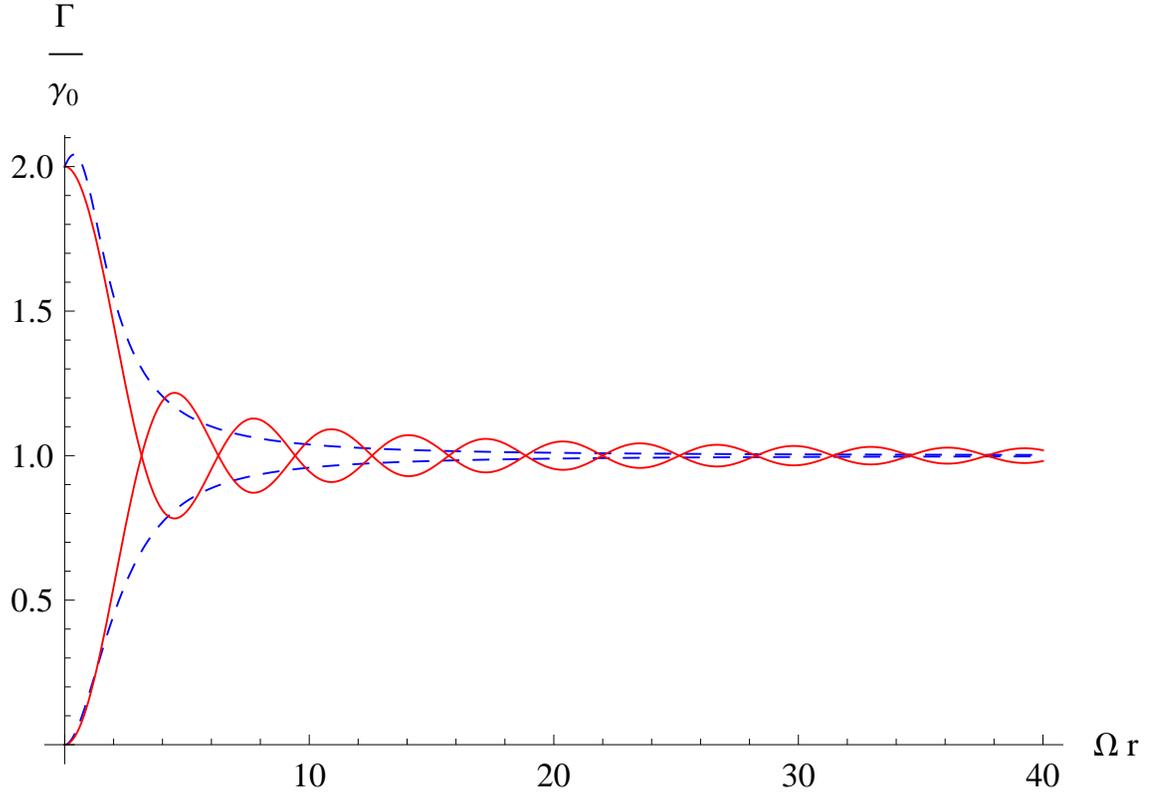


Figure 6.13: Phenomenological decay rates as a function of separation for two resonant oscillators using **· weak-coupling perturbation** and **··· the small- $r$  Padé approximation** where  $\gamma_0 = \Omega/10$ .

at resonance must the two oscillators be to have sub and super-radiance.

In Fig. 6.13 we plot the phenomenological damping rates  $\Gamma$  as a function of separation between two resonant oscillators. The sub-radiant decay rate starts at zero while the super-radiant decay rate starts doubled. Both then asymptote to the same value for large separations. For the exact regulator, the intermediate behavior of the decay rates is somewhat oscillatory and the separation timescales play a fairly different (and more causal) role than a simple decay rate. But the important information to extract is that the existence of the dark and bright states is not

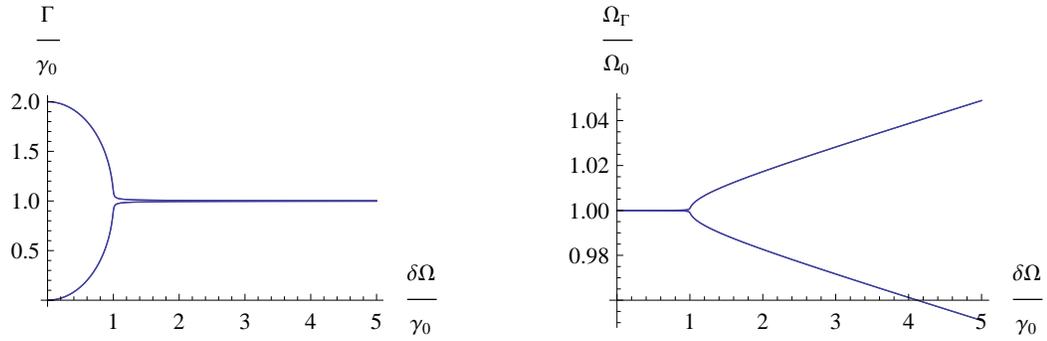


Figure 6.14: Homogeneous timescales as a function of detuning for two close oscillators where  $\gamma_0 = \Omega/100$ .

particularly sensitive to a lack of proximity (where our approximation is valid). The relevant length scale appears to simply be the resonant wavelength.

In Fig. 6.14 we plot the phenomenological damping and oscillation rates  $\Gamma$  and  $\Omega_\Gamma$  as a function of detuning between two close oscillators' free parameters. To be more specific, these oscillators have effective modes which evolve with frequencies  $\Gamma \pm i\tilde{\Omega}_\Gamma$  where  $\tilde{\Omega}_\Gamma = \sqrt{\Omega_\Gamma^2 - \Gamma^2}$ . As with two-level atoms, the tuning needs to be better than the damping rate (see Fig. 6.4). Here we can also see that the frequency of the damped oscillators initially resists any display of detuning until this critical threshold is reached. The bright and dark modes do oscillate at different rates, as their damping rates are very different, but they would appear to have more similar undamped frequencies than they actually do.

## 6.5 Discussion

In this chapter we have considered the entanglement dynamics of multipartite systems in common relativistic field. In particular we have considered scalar and electromagnetic field correlations as they influence to two-level and harmonic oscillator atoms at fixed locations.

In Sec. 6.3 we derived the dynamics of a collection of two-level atoms under a dipole approximation interacting with a common quantized electromagnetic field assuming only weak coupling and not the Born-Markov approximation (BMA) or rotating-wave approximation (RWA). The solution we have derived here therefore yields greater accuracy than those derived using the RWA, which is assumed in most prior studies of such systems. We have also presented a method of finding the zero-temperature asymptotic state to higher accuracy than is possible directly with a second-order master equation. We have used this to show that even at zero temperature the bipartite entanglement between any pair of atoms will undergo sudden death for all initial atomic states, in contrast to the predictions of previous theoretical treatments [58] under BMA or RWA. Finally, we have characterized the various decay rates that are present in this solution without the RWA and the sub- and super-radiant states that exist.

We have argued that in the RWA there can be inaccuracies in all entries of the density matrix that are of the order of the damping rate  $\gamma$ . By contrast, when represented in the (free) energy basis the solution we have derived here will have off-diagonal elements that are accurate at second-order, having  $\mathcal{O}(\gamma^2)$  errors. Even

in this solution diagonal matrix elements (and matrix elements between any two degenerate energy states) can still have  $\mathcal{O}(\gamma)$  errors, due to a fundamental limitation of any weak-coupling master equation. However, the expectation of any operator that has vanishing diagonals in the energy basis (including atomic dipole operators), will have only  $\mathcal{O}(\gamma^2)$  errors. Moreover, unlike some other methods of solution, our solution can be applied when the atoms have distinct frequencies.

At sufficiently low temperature, the zeroth-order asymptotic state (given by the RWA) is near the boundary between the separable and entangled states, and the small perturbation induced by the environment at  $\mathcal{O}(\gamma)$  can push it into either set. Depending on which set the perturbed asymptotic states fall into, all states may experience entanglement sudden death or all may become entangled asymptotically. We have presented a second-order solution for the asymptotic state of any two two-level atoms, which allows us to say decisively that the zero-temperature asymptotic state of those atoms is separable, and pairwise entanglement of all atoms experiences sudden death regardless of the initial state. This is in stark contrast to the oscillator atoms we considered in Sec. 6.4, which actually asymptotically entangle when placed at close proximity.

It should be noted that, for example, in some optical-frequency atomic systems the  $\mathcal{O}(\gamma)$  corrections we discuss can be quite small, with  $\gamma/\Omega$  being perhaps something on the order of  $10^{-9}$ . Though lowest order corrections to the timescales cannot be ignored (as they are responsible for the presence of dissipation), corrections of this size to the values of the density matrix elements at any instant can easily be considered negligible. However, in the case of a theoretical study of entanglement

sudden death, where one wishes to distinguish asymptotic decay to zero from vanishing in finite time, small perturbations can become vitally important, as they do at low temperature. And in optical frequency atomic systems at room temperature the thermal-average photon number will be far smaller than  $10^{-9}$ , placing the system deep into what we are considering the low-temperature regime for entanglement dynamics.

We have characterized the sub- and super-radiant states that exist in this model when the RWA is not used. We have shown that there is still a long-lived, highly-entangled dark state when the atoms have small enough separation, and sudden death of entanglement occurs only on the much longer timescale of decay of this state (assuming it had some population in the initial state). In this simple model, decoherence-free dark states are achievable for arbitrary temperature and dissipation, whereas typically these factors together are the primary cause of decoherence. This result is achieved through interference phenomena in the noise processes themselves. Both destructive and constructive interference occur, producing dark states and bright states. In this model the number of such states can be fairly large, which is a favorable condition for QIP.

We close with a few remarks: 1) With the knowledge of distance dependence, to preserve entanglement in time one should place the atoms very close to each other in the field, so as to produce strong cross correlations in the noise. But at some proximity one must also consider further atom-atom interactions, perhaps self-consistently within the confines of field theory. 2) Qualitative differences between systems under the two-level and dipole-interaction approximations and harmonic

systems suggests a degree of model dependence in some of the phenomena considered; this merits further investigation into the consequences of these approximations.

3) Many other sorts of level structures are relevant to experimental systems, both in terms of the number of levels involved and the angular momentum exchange with the field. The methodology and conceptions developed in this work can be applied for the analysis of the non-Markovian dynamics of more general systems, from which one can perhaps better understand how model-dependent the entanglement behavior considered herewith is.

## Chapter 7

### The Abraham-Lorentz Force

#### 7.1 Introduction

##### 7.1.1 Background

The Backreaction of a charged particle interacting with an electromagnetic field involves a number of famous problems including acausality, in the form of pre-acceleration, and runaway solutions to the Abraham-Lorentz equation [91]. A number of approaches to resolving these problems have been developed, including replacing point particles with extended objects [156, 133, 112], treating the electromagnetic field interaction perturbatively and truncating at a specific order [15], or replacing time-local differential equations of motion with nonlocal integro-differential equations of motion [67, 70]. Although these approaches yield models with causal dynamics, or alternatively it is observed that point particle models are at best effective models and the pathological solutions simply indicated a breakdown in their validity, nevertheless there remain important open questions about the stability of backreaction for particle plus field models, and the complex nonequilibrium dynamics that can arise from the interactions between a localized particle and nonlocal field degrees of freedom.

The physical problem of point particles self-interacting via the electromagnetic

field can be viewed in the context of quantum open systems. In such problems the system (or particles) are treated as ordinary quantum-mechanical degrees of freedom whereas the environment (a reservoir or field) has an uncountable number of degrees of freedom. Therefore the environment may induce irreversible behavior upon the system, in the form of dissipation and decoherence, and also stochastic behavior upon the system, in the form of noise. For equilibrium environments, these influences are related by the fluctuation-dissipation relation, including the decoherence (see Sec. 4.7).

It is well known that such models require a finite U.V. cutoff in the quantum domain, as to limit response to zero-point fluctuations of the environment  $\Lambda$  (see Chapter 3). Physically one can interpret this cutoff sensitivity as being related to the effective nature of the theory. The quantum-mechanical system and field are not being treated on equal footing. The system is, in fact, more akin to some local excitations of a matter field (e.g. the Dirac field). As such, the system cannot be probed too closely. We will discuss how other physically-motivated resolutions of preventing runaway solutions are equivalent to enforcing a U.V. cutoff in Sec. 7.4. The peculiarity of the electrodynamic backreaction is not in that it requires a finite cutoff, but that it appears to require a finite cutoff even in the classical dissipation (as opposed to solely in the quantum noise) and that there is a strict upper bound on the allowable cutoff as it relates to the positivity of the particle's bare mass.

There are two methods of derivation for the electromagnetic backreaction. By far the simplest derivation of the Abraham-Lorentz equation, which we designate to be *Larmor derived*, is to consider the power (more correctly stress-energy and

momenta) radiated by the moving particle, and then infer from that a reactive force upon the particle. One can also consider a first-principles derivation, which we designate to be *Maxwell-Lorentz* or *Langevin derived*, by (1) solving for the fields as sourced by the system and then (2) feeding that solution back into the system dynamics to obtain an effective equation of motion for the open system, which in the Langevin case may also be made to include quantum and thermal noise. In the non-relativistic regime, these two methods produce what is essentially the same answer for a point charge, though only the Langevin equation may be considered quantum mechanically. Whereas in the classical regime noise can be made to vanish in the zero-temperature or vacuum limit, quantum mechanics necessitates noise which obeys a (non-vanishing) fluctuation-dissipation relation if it is thermal or a fluctuation-dissipation inequality more generally (see Sec. 4.6).

In the relativistic regime, the Larmor-derived theory is not unique and some degree of guesswork is required. Dirac motivated his Abraham-Dirac-Lorentz equation by simplicity, but there are an infinite number of equally valid equations with terms of higher order in  $v/c$  [51, 17]. Let us designate all such backreactions to be of the Dirac-Bhabha family. Furthermore, the Abraham-Dirac-Lorentz equation exhibits all of the same pathologies as the Abraham-Lorentz equation and no contending higher-order equations are known to avoid this behavior.

The Abraham-Dirac-Lorentz equation can be “tamed” by an iterative process, which produces a perturbatively different equation: the Eliezer [55] and Landau-Lifshitz [103] reaction formulas being the best examples. The Eliezer and Landau-Lifshitz backreactions are coincident in the nonrelativistic regime, whereas in the

relativistic regime they make different predictions from each other and even from Maxwell's equations, as they are perturbed outside of the Dirac-Bhabha family of backreactions. Therefore they cannot be viewed as fundamental. Spohn demonstrated that there is a measure-zero set of initial conditions for which the Abraham-Dirac-Lorentz equation will not exhibit runaway solutions, and that this critical manifold of solutions is unstable [142]. Resolving the effective equations of motion which constrain all dynamics to these non-runaway solutions also results in the Landau-Lifshitz equation. Aside from the lack of physical derivation and the conflict with Maxwell's equations, it is also unclear how these taming formalisms can be made to properly incorporate noise and extended into the quantum regime.

Though the derivation is much more challenging, there are numerous advantages to the Langevin-derived theory. Extension to the quantum regime is included as the stochastic dynamics of the open system are derived from a Hamiltonian. Issues of causality are also resolved as the initial conditions of the system and environment determine all subsequent dynamics for the open system. If the open system appears to require additional initial conditions, they can be calculated from the theory. From the first-principles perspective, it will also be clear as to why runaway solutions occur if they do. Coupling a system to a thermal reservoir (which is effectively what the electromagnetic field is to a particle) should relax the open system to global equilibrium, regardless of the initial conditions. This was rigorously proven to second-order in Sec. 4.5 even for nonlinear system-environment interactions.

From the Maxwell-Lorentz and Langevin perspectives it is clear that runaway solutions only occur in the Abraham-Lorentz equation when the bare mass of the

system is allowed to become negative. Given a negative bare mass, one has what is, in analogy, an inverted oscillator coupled to a dissipative reservoir. In this light, it is physically obvious that runaway solutions are almost certain to occur. In fact, the critical manifold of solutions described by Spohn are merely the solutions where the particle is unstably balanced atop energy maxima. Constraining solutions to this critical manifold by the Landau-Lifshitz reaction is therefore highly artificial. The system should instead relax to (or to some Brownian motion in the neighborhood of) energy minima.

### 7.1.2 New Results Herein

The Abraham-Lorentz-Langevin equation for a nonrelativistic and *structureless* point charge coupled to the electromagnetic field and driven by other external forces is given by

$$m \ddot{\mathbf{x}}(t) = \mathbf{F}_{\text{ext}}(t) + \underbrace{2e^2 \gamma_0 \ddot{\mathbf{x}}(t)}_{\text{backreaction}} - \underbrace{e \dot{\boldsymbol{\xi}}(t)}_{\text{noise}}, \quad (7.1)$$

$$\gamma_0 = \frac{1}{12\pi\epsilon_0 c^3}, \quad (7.2)$$

and it has been derived microscopically from the Hamiltonian theory [44, 67, 15]. This equation is not manifestly causal and exhibits runaway solutions which occur within perturbative timescales. These runaway solutions are not necessarily problematic here as Eq. (7.1) must be viewed as a truncated perturbative series in  $1/c$ . The Abraham-Lorentz backreaction is indeed the lowest-order dissipative correction from (relativistic) electrodynamics and emerges a mere one power after the  $1/c^2$  magnetostatic forces ( $\mu_0 = 1/\epsilon_0 c^2$ ), which in turn can be viewed as non-dissipative

backreaction from the electromagnetic field. Truly, if we are to be entirely honest in our analysis, then the particle cannot be perfectly nonrelativistic and left-hand side of Eq. (7.1) should contain a  $(v/c)^2 \ddot{\mathbf{x}}$  correction. However, this adjustment does not appear to make for any qualitative differences in the theory.

Ford & O’Connell [70] have shown that their time-nonlocal, causal and runaway-free theory of a *structured* point charge (also microscopically derived) is perturbatively consistent with the time-local, causal and runaway-free equation of motion

$$m \ddot{\mathbf{x}}(t) = \mathbf{F}_{\text{ext}}(t) + \frac{2e^2\gamma_0}{m} \frac{d}{dt} \mathbf{F}_{\text{ext}}(t), \quad (7.3)$$

where to derive this equation, one merely iterates Eq. (7.1) once and drops all high-order terms.

In this work, we derive Eq. (7.3) directly from the Hamiltonian theory by making our perturbatively-consistent adjustments at the level of the Hamiltonian instead of at the level of the Langevin equation. Note that this is not a means of deriving a more accurate theory, but hopefully a means of constructing a better perspective of the problem. In our formalism it is seen that the mass renormalization is of a very different form and the bare mass is never made negative, regardless of the cutoff. We believe this demonstrates that the nonrelativistic theory of electromagnetic backreaction is a perfectly-adequate effective theory. No such claim can readily be made about the relativistic theory. It is much more problematic, both physically and mathematically. However, it should be noted that one can construct a causal and runaway-free relativistic theory of scalar-field backreaction [92] and therefore there is no intrinsic pathology inherent in relativistic mechanics.

### 7.1.3 Overview

To find the backreaction of the charged particle, the field degrees of freedom are integrated out exactly, following well-known procedures [67], making the particle an open system. The field is effectively a bath of harmonic oscillators linearly driven by the system, allowing one to solve for the bath dynamics as driven by the system and then substitute the driven solution back into the system's dynamics. The system's influence upon itself, as mediated by the environment, is known as backreaction and this is distinguished from noise, which arises from the undriven portion of the environment's evolution.

The non-relativistic problem of electromagnetic backreaction can be related to supra-Ohmic Quantum Brownian Motion (QBM), which we first consider in Sec. 7.2. QBM is a theory which is very well understood and readers may find these calculations more palatable than the later derivations. Important characteristics of the QBM model, such as the renormalization and integration kernels are more thoroughly discussed in Chapter 3. The equations of motion which we derive apply to both the classical trajectories and the Heisenberg-picture operators. In the latter case, the induced noise will be operator valued and has a complex-valued noise correlation and (moreover state-independent) commutator.

In Sec. 7.3 we perform the (analogous) standard analysis of nonrelativistic quantum particles in the (relativistic) electromagnetic field without immediately taking any dipole approximation. There are two standard calculations which arise from different choices of coupling gauge. It is seen that the first choice has an peculiar

notion of thermal noise, whereas the second choice involves relatively pathological integration kernels. In Sec. 7.4 we briefly mention how other physically-motivated repairs to the theory, such as a finite-sized particle, are equivalent to the U.V. cutoff, which is the more natural repair in the Langevin formalism. Finally in Sec. 7.5 we present our effective Hamiltonian theory.

## 7.2 Quantum Brownian Motion

We begin our discussion with the Quantum Brownian Motion (QBM) Lagrangian which we have adapted in form and notation to better mirror the problem of backreaction in the electromagnetic field. This Lagrangian describes a quantum system bilinearly coupled to a bosonic bath of harmonic oscillators and is traditionally used to model ordinary motional damping in quantum mechanics.

$$\mathcal{L}_{\text{QBM}} = \mathcal{L}_{\text{sys}} + \mathcal{L}_{\text{int}} + \mathcal{L}_{\text{env}} , \quad (7.4)$$

$$\mathcal{L}_{\text{sys}} \equiv \frac{1}{2} m \dot{\mathbf{x}}^2 - U(\mathbf{x}) , \quad (7.5)$$

$$\mathcal{L}_{\text{int}} = e \mathbf{x} \cdot \dot{\mathbf{Q}} , \quad (7.6)$$

$$\mathcal{L}_{\text{env}} = \int_0^\infty dk \frac{1}{2} \{ \dot{\mathbf{q}}_k^2 - \omega_k^2 \mathbf{q}_k^2 \} , \quad (7.7)$$

where  $\mathbf{x}$  denotes the system position,  $\mathbf{q}_k$  denote the field-mode “positions”, and  $\mathbf{Q}$  is the collective field operator

$$\mathbf{Q} \equiv \int_0^\infty dk g_k \mathbf{q}_k . \quad (7.8)$$

The system + environment Hamiltonian is then given by

$$\mathbf{H}_{\text{QBM}} = \mathbf{H}_{\text{sys}} + \int_0^\infty dk \frac{1}{2} \{ (\boldsymbol{\pi}_k - e g_k \mathbf{x})^2 + \omega_k^2 \mathbf{q}_k^2 \}, \quad (7.9)$$

$$\mathbf{H}_{\text{sys}} \equiv \frac{\mathbf{p}^2}{2m} + U(\mathbf{x}), \quad (7.10)$$

where, as determined by the gauge of our Lagrangian,  $\mathbf{p}$  is the system momentum conjugate to  $\mathbf{x}$  and  $\boldsymbol{\pi}$  is the field “momentum” conjugate to  $\mathbf{q}$ .

Note that for  $m \geq 0$  and  $U(\mathbf{x})$  sufficiently well behaved, Hamiltonian (7.9) is bounded from below in its energy spectrum. Therefore, under these conditions runaway solutions will not occur when the environment is initially described by a thermal state. This statement will be rigorously proven in Sec. 7.2.5. Correspondence between the free and interacting theories is a separate matter.

Additionally note that the “bare” system potential in Eq. (7.9) is given by

$$U_{\text{bare}}(\mathbf{x}) = U(\mathbf{x}) + \left( e^2 \int_0^\infty dk \frac{g_k^2}{2} \right) \mathbf{x}^2, \quad (7.11)$$

and that the system + environment Hamiltonian can also be expressed

$$\mathbf{H}_{\text{QBM}} = \mathbf{H}_{\text{sys}}^{\text{bare}} - e \mathbf{x} \cdot \boldsymbol{\pi} + \int_0^\infty dk \frac{1}{2} \{ \boldsymbol{\pi}_k^2 + \omega_k^2 \mathbf{q}_k^2 \}, \quad (7.12)$$

in terms of the collective field operator

$$\boldsymbol{\pi} \equiv \int_0^\infty dk g_k \boldsymbol{\pi}_k. \quad (7.13)$$

The resulting Heisenberg equations of motion then dictate that the system is driven by the field

$$\dot{\mathbf{x}} = \frac{\mathbf{p}}{m}, \quad (7.14)$$

$$\dot{\mathbf{p}} = -\nabla U_{\text{bare}}(\mathbf{x}) + e \boldsymbol{\pi}, \quad (7.15)$$

whereas the field modes are driven by the system

$$\dot{\mathbf{q}}_k = \boldsymbol{\pi}_k - e g_k \mathbf{x}, \quad (7.16)$$

$$\dot{\boldsymbol{\pi}}_k = -\omega_k^2 \mathbf{q}_k. \quad (7.17)$$

Solving for the field-mode evolution as driven by the system, we obtain the homogeneous + driven solution

$$\boldsymbol{\pi}_k(t) = \boldsymbol{\xi}_k(t) + e g_k \omega_k^2 (G_k * \mathbf{x})(t), \quad (7.18)$$

$$\boldsymbol{\xi}_k(t) \equiv \dot{G}_k(t) \boldsymbol{\pi}_k(0) + \ddot{G}_k(t) \mathbf{q}_k(0), \quad (7.19)$$

$$G_k(t) \equiv \frac{\sin(\omega_k t)}{\omega_k}, \quad (7.20)$$

where the  $*$  product denotes the Laplace convolution

$$(A * B)(t) \equiv \int_0^t dt' A(t-t') B(t'). \quad (7.21)$$

The time-evolving field operator is then given by

$$\boldsymbol{\pi}(t) = \underbrace{\boldsymbol{\xi}(t)}_{\text{noise}} - \underbrace{2e(\mu * \dot{\mathbf{x}})(t)}_{\text{dissipation}}, \quad (7.22)$$

$$\boldsymbol{\xi}(t) \equiv \int_0^\infty dk g_k \boldsymbol{\xi}_k(t), \quad (7.23)$$

$$\mu(t) \equiv - \int_0^\infty dk \frac{g_k^2 \omega_k^2}{2} G_k(t), \quad (7.24)$$

where  $\mu(t, t') = \mu(t - t')$  is the stationary dissipation kernel and  $\boldsymbol{\xi}(t)$  is a Gaussian stochastic process for the initial conditions we assume: a factorized state of the system and environment, with the environment in a thermal state.

Next we introduce the related damping kernel

$$\mu(t, t') = -\frac{\partial}{\partial t'} \gamma(t, t'), \quad (7.25)$$

$$\gamma(t, t') \equiv \int_0^\infty dk \frac{g_k^2}{2} \cos[\omega_k(t-t')], \quad (7.26)$$

which is necessarily positive definite and independent of the (factorized) initial state of the environment. The backreaction can then be expressed

$$\underbrace{(\mu * \mathbf{x})(t)}_{\text{dissipation}} = \underbrace{(\gamma * \dot{\mathbf{x}})(t)}_{\text{damping}} + \underbrace{\gamma(t, 0) \mathbf{x}(0)}_{\text{slip}} - \underbrace{\gamma(t, t) \mathbf{x}(t)}_{\text{renormalization}}, \quad (7.27)$$

in terms of the positive-definite damping and where we have labeled the terms corresponding to the renormalization and initial short-time slip dynamics associated with the factorization of the initial state (see Sec. 7.2.3).

Feeding our field solutions into the system equations of motion, we obtain the quantum Langevin equation

$$m\ddot{\mathbf{x}}(t) + 2e^2(\gamma * \dot{\mathbf{x}})(t) + \nabla U(\mathbf{x}) = e\boldsymbol{\xi}(t) - e^2\gamma(t)\mathbf{x}(0), \quad (7.28)$$

which reduces to

$$m\ddot{\mathbf{x}}(t) + 2e^2(\gamma * \dot{\mathbf{x}})(t) + \nabla U(\mathbf{x}) = e\boldsymbol{\xi}(t), \quad (7.29)$$

after the transient slip is taken into account.

## 7.2.1 Ohmic Coupling and Local Damping

Considering the damping kernel, which is given by

$$\gamma(t) = \int_0^\infty dk \frac{g_k^2}{2} \cos(\omega_k t). \quad (7.30)$$

If assume  $\omega_k = ck$  and  $g_k \approx g$  up to some high-frequency cutoff  $\Lambda$ , then we may evaluate the integral as

$$\gamma(t) = \frac{g^2}{2c} \int_0^\Lambda d\omega \cos(\omega t) = \frac{g^2}{2c} \frac{\sin(\Lambda t)}{t}. \quad (7.31)$$

The damping kernel may then be expressed

$$\gamma(t) = 2\gamma_0 \delta_\Lambda(t), \quad (7.32)$$

$$\gamma_0 \equiv \frac{\pi g^2}{4c}, \quad (7.33)$$

$$\delta_\Lambda(t) \equiv \frac{\sin(\Lambda t)}{\pi t}, \quad (7.34)$$

in terms of the Dirac delta  $\delta_\Lambda(t)$ . In the high-frequency limit, the damping contribution to the Langevin equation becomes

$$\lim_{\Lambda \rightarrow \infty} (\gamma * \dot{\mathbf{x}})(t) = \gamma_0 \dot{\mathbf{x}}(t), \quad (7.35)$$

or local damping.

## 7.2.2 Renormalization

For Ohmic coupling or local damping the quantum Langevin equation described by Eq. (7.29) is phenomenological, in the sense that its various parameters entail precisely what they appear to. Assuming the Langevin equation to be phenomenological, note the bare system potential in Hamiltonian perspective (7.12) & (7.15) as compared to the phenomenological system potential  $U(\mathbf{x})$ .

$$U_{\text{bare}}(\mathbf{x}) = U(\mathbf{x}) + 2e^2 \gamma(0) \mathbf{x}^2, \quad (7.36)$$

where  $\gamma(0) = \frac{g^2}{2c}\Lambda$  for local damping with a hard cutoff regulator. The proper renormalization is a quadratic term, regardless of whether or not the original model contained such a term. The QBM model typically proceeds from an  $\mathbf{x}\cdot\mathbf{Q}$  interaction, where this renormalization does not naturally result from the Lagrangian theory.

### 7.2.3 Factorized Initial Conditions

If the operator noise  $\boldsymbol{\xi}(t)$  in our Langevin equation is to be sampled from a thermal distribution which is (initially) statistically independent from the system, then the initial state of the system and environment must be a product state of the form  $\boldsymbol{\rho}_{\text{sys+env}} = \boldsymbol{\rho}_{\text{sys}} \otimes \boldsymbol{\rho}_{\text{env}}$  or a product of marginal phase-space distributions in the classical regime, and with the environment initially in equilibrium. This is an important simplification in our (and most other, e.g., [57, 27]) analyses of the nonequilibrium dynamics of open systems.

The consequence of assuming an initially uncorrelated system and environment must be carefully examined when studying radiation reaction, however, since acausal behaviors arise during the same very short time scale where the unphysical nature of a factorized state is relevant. It is therefore an important aspect of our analysis that we are also able to apply the results of Chapter 3 showing that for classical or high-temperature electromagnetic noise ( $\hbar\omega_{\text{sys}} \ll k_{\text{B}}T$  in Eq. (7.39)) the initial evolution of factorized states (or distributions) quickly leads to physical, dressed particle states without reintroducing the pathologies or instabilities in the dynamics that our analysis is intended to avoid. In the semiclassical or quantum regime, use

of properly-correlated initial states can mitigate the unphysical aspects of assuming initially factorized states entirely, without otherwise spoiling the results in this paper (see Sec. 4.3).

### 7.2.3.1 The Slip

The transient *slip* in our Langevin equation is an initial-time pathology associated with vanishing correlations in the factorized initial conditions despite non-vanishing interaction strength between the system and field. In addition to the slip, there is a diffusive initial-time pathology, called *jolts*, which arise from correlation with the (quantum) zero-point fluctuations of the environment. The slip in particular was thoroughly analyzed in Chapter 3, where it was pointed out to generate the linear dynamical map

$$\rho \rightarrow e^{+i2e^2\gamma_0\mathbf{x}^2} \rho e^{-i2e^2\gamma_0\mathbf{x}^2}, \quad (7.37)$$

which maps states in a unitary fashion and preserves all kinematic moment invariants [53], including the uncertainty function. Therefore one can identify the post-slip state as a “renormalized” initial state which is more properly correlated with the environment and the pre-slip state as a “bare” initial state. If one only considers the classical regime, then jolting is not severe due to the lack of zero-point fluctuations in the environment. Moreover, for a classical zero-temperature environment there is no noise causing any diffusion. Thus for this case one only needs to consider the renormalized initial states, effectively discarding the slip term entirely.

## 7.2.4 The Fluctuation-Dissipation Relation

The Gaussian process  $\boldsymbol{\xi}(t)$  has the noise kernel

$$\nu(t, t') = \frac{1}{2} \langle \{\boldsymbol{\xi}(t), \boldsymbol{\xi}(t')\} \rangle_{\boldsymbol{\xi}} = \nu(t-t'), \quad (7.38)$$

which is stationary and positive definite for any stationary initial state of the environment. For an equilibrium initial state of the environment the noise kernel is related to damping kernel by the (quantum) fluctuation-dissipation relation (FDR)

$$\tilde{\nu}(\omega) = \tilde{\gamma}(\omega) \hbar \omega \coth\left(\frac{\hbar \omega}{2k_{\text{B}}T}\right), \quad (7.39)$$

where  $\tilde{\gamma}(\omega) = \int_{-\infty}^{+\infty} dt e^{-i\omega t} \gamma(t)$ . Essentially, the damping kernel and temperature completely characterize Gaussian, thermal noise.

Also note that as the coupling and environment are dynamically linear, the damping kernel, being determined by the commutator, is independent of the state of the environment and it is the same whether in the classical or quantum regimes. In the classical regime we have the limit

$$\lim_{\hbar \rightarrow 0} \tilde{\nu}(\omega) = \tilde{\gamma}(\omega) 2k_{\text{B}}T, \quad (7.40)$$

and the classical fluctuations vanish in the zero temperature limit. In this limit (the classical vacuum) we can neglect the stochastic process  $\boldsymbol{\xi}(t)$ .

In the quantum regime, the anti-commutator expectation value (7.38) is not sufficient to describe the statistics of the operator-valued stochastic process  $\boldsymbol{\xi}(t)$ .

One additionally requires the commutator expectation value

$$\mu(t, t') = \frac{1}{2i} \langle [\boldsymbol{\xi}(t), \boldsymbol{\xi}(t')] \rangle_{\boldsymbol{\xi}} = \mu(t-t'), \quad (7.41)$$

which is given by the dissipation kernel, a state-independent quantity. Here the dissipation kernel is not generating backreaction, but consistent time evolution for the non-commuting stochastic process. The full quantum correlation is therefore given by

$$\alpha(t, t') \equiv \langle \underline{\boldsymbol{\pi}}(t) \underline{\boldsymbol{\pi}}(t') \rangle_{\text{env}} = \langle \boldsymbol{\xi}(t) \boldsymbol{\xi}(t') \rangle_{\boldsymbol{\xi}}, \quad (7.42)$$

$$= \nu(t-t') + \imath \mu(t-t'), \quad (7.43)$$

where  $\underline{\boldsymbol{\pi}}(t)$  denotes the interaction-picture or Dirac-picture field operator and not the Heisenberg-picture field operator which we have already denoted  $\boldsymbol{\pi}(t)$ .

### 7.2.5 Proof of Stability

We will now show that the dynamics of the system are dissipative and stable under the very same conditions for which the system + environment Hamiltonian (7.9) has a lower bound in its energy spectrum. Let us denote the system Hamiltonian

$$\mathbf{H}_{\text{sys}} \equiv \frac{\mathbf{p}^2}{2m} + U(\mathbf{x}). \quad (7.44)$$

One may then calculate an energy constraint from either the Heisenberg equations of motion for  $\mathbf{H}_{\text{sys}}(t)$  or by integrating the (classical) Langevin equation (7.29) along with velocity. Accounting for the slip in our initial state, which only produces a finite change in energy, we obtain the relation

$$\mathbf{H}_{\text{sys}}(t) = \mathbf{H}_{\text{sys}}(0) - \mathbf{H}_{\gamma}(t) + \mathbf{H}_{\xi}(t), \quad (7.45)$$

in terms of the energy lost to damping  $\mathbf{H}_\gamma(t)$  and energy generated by noise  $\mathbf{H}_\xi(t)$

$$\mathbf{H}_\gamma(t) = e^2 \int_0^t dt' \int_0^t dt'' \gamma(t', t'') \frac{1}{2} \{ \dot{\mathbf{x}}(t') \cdot \dot{\mathbf{x}}(t'') + \dot{\mathbf{x}}(t'') \cdot \dot{\mathbf{x}}(t') \}, \quad (7.46)$$

$$\mathbf{H}_\xi(t) = e \int_0^t dt' \frac{1}{2} \{ \boldsymbol{\xi}(t') \cdot \dot{\mathbf{x}}(t') + \dot{\mathbf{x}}(t') \cdot \boldsymbol{\xi}(t') \}. \quad (7.47)$$

The contribution from damping is a negative quantity as the damping kernel is a positive-definite kernel in a quadratic form. The noise is random and may drive the system erratically, but the damping may only remove energy from the system (and deliver it to the environment and interaction). Therefore it is imperative that  $\mathbf{H}_{\text{sys}}$  have a lower bound in its energy spectrum. For our model, this necessarily implies that the system + environment Hamiltonian (7.9) also has a lower bound in its energy spectrum. If this is the case then true runaway motion cannot occur. In the classical-vacuum limit, energy is continually siphoned from  $\mathbf{H}_{\text{sys}}(t)$  until all motion ceases.

Locally-damped energy is additionally simplified to

$$\dot{\mathbf{H}}_\gamma(t) = 2 e^2 \gamma_0 \dot{\mathbf{x}}(t)^2, \quad (7.48)$$

which monotonically dissipates energy in time. Nonlocal damping can produce an instantaneous increase in system energy, though the cumulative effect is always dissipative.

### 7.3 Standard Calculations of Nonrelativistic Quantum Electrodynamics

We now consider the QED Lagrangian for a nonrelativistic charged particle coupled to the electromagnetic field. By quantum electrodynamics we do not mean to imply the full theory of Dirac and electromagnetic fields, but the effective theory of point charges (represented by ordinary quantum mechanical degrees of freedom) interacting with the electromagnetic field. Also note that this theory is actually half nonrelativistic (the particle) and half relativistic (the field), and as such, some degree of additional nonrelativistic approximation is suitable. The system + environment Lagrangian is given by

$$\mathcal{L}_{\text{QED}} = \mathcal{L}_{\text{sys}} + \mathcal{L}_{\text{int}} + \mathcal{L}_{\text{env}} , \quad (7.49)$$

$$\mathcal{L}_{\text{sys}} \equiv \sum_j \frac{1}{2} m_j \dot{\mathbf{x}}_j^2 , \quad (7.50)$$

$$\begin{aligned} \mathcal{L}_{\text{int}} &= \iiint d^3 x' \{ \mathbf{J}(\mathbf{x}, \mathbf{x}') \cdot \mathbf{A}(\mathbf{x}') - \rho(\mathbf{x}, \mathbf{x}') \phi(\mathbf{x}') \} , \\ &= \sum_j e_j \{ \dot{\mathbf{x}}_j \cdot \mathbf{A}(\mathbf{x}_j) - \phi(\mathbf{x}_j) \} , \end{aligned} \quad (7.51)$$

$$\mathcal{L}_{\text{env}} = \iiint d^3 x' \frac{\epsilon_0}{2} \{ \mathbf{E}(\mathbf{x}')^2 - c^2 \mathbf{B}(\mathbf{x}')^2 \} , \quad (7.52)$$

with the interaction expressed in terms of the vector and scalar potentials, related to the electromagnetic fields in the environment Lagrangian by the standard relations

$$\mathbf{E}(\mathbf{x}) = -\nabla\phi(\mathbf{x}) - \frac{\partial}{\partial t}\mathbf{A}(\mathbf{x}) , \quad (7.53)$$

$$\mathbf{B}(\mathbf{x}) = \nabla \times \mathbf{A}(\mathbf{x}) . \quad (7.54)$$

In the Hamiltonian formalism we may express the magnetic vector potential via spatial Fourier mode  $\mathbf{k}$  and polarization  $\epsilon_k$  expansion

$$\mathbf{A}(\mathbf{x}) = \frac{1}{(2\pi)^{3/2}} \iiint d^3k \sum_{\epsilon_k} \mathbf{A}_{\mathbf{k},\epsilon_k}(\mathbf{x}), \quad (7.55)$$

$$\mathbf{A}_{\mathbf{k},\epsilon_k}(\mathbf{x}) = \frac{\boldsymbol{\epsilon}_k}{\sqrt{2\varepsilon_0\omega_k}} \left\{ e^{+i\mathbf{k}\cdot\mathbf{x}} \mathbf{a}_{\mathbf{k},\epsilon_k} + e^{-i\mathbf{k}\cdot\mathbf{x}} \mathbf{a}_{\mathbf{k},\epsilon_k}^\dagger \right\}, \quad (7.56)$$

where  $\omega_k = ck$ . To satisfy the commutation relations, the conjugate momentum field is then given by

$$\boldsymbol{\pi}(\mathbf{x}) = \frac{1}{(2\pi)^{3/2}} \iiint d^3k \sum_{\epsilon_k} \boldsymbol{\pi}_{\mathbf{k},\epsilon_k}(\mathbf{x}), \quad (7.57)$$

$$\boldsymbol{\pi}_{\mathbf{k},\epsilon_k}(\mathbf{x}) = \frac{-i\boldsymbol{\epsilon}_k}{\sqrt{2/\varepsilon_0\omega_k}} \left\{ e^{+i\mathbf{k}\cdot\mathbf{x}} \mathbf{a}_{\mathbf{k},\epsilon_k} - e^{-i\mathbf{k}\cdot\mathbf{x}} \mathbf{a}_{\mathbf{k},\epsilon_k}^\dagger \right\}. \quad (7.58)$$

Let us introduce the notation

$$\mathbf{A}_{\mathbf{k},\epsilon_k} \equiv \mathbf{A}_{\mathbf{k},\epsilon_k}(\mathbf{0}), \quad (7.59)$$

$$\boldsymbol{\pi}_{\mathbf{k},\epsilon_k} \equiv \boldsymbol{\pi}_{\mathbf{k},\epsilon_k}(\mathbf{0}). \quad (7.60)$$

These  $\mathbf{x}$ -independent field operators will more closely correspond to the “positions” and “momenta” of the reservoir oscillators, in analogy with QBM. The field operators evaluated at specific locations can then be expressed

$$\mathbf{A}_{\mathbf{k},\epsilon_k}(\mathbf{x}) = \cos(\mathbf{k}\cdot\mathbf{x}) \mathbf{A}_{\mathbf{k},\epsilon_k} - \sin(\mathbf{k}\cdot\mathbf{x}) \frac{\boldsymbol{\pi}_{\mathbf{k},\epsilon_k}}{\varepsilon_0\omega_k}, \quad (7.61)$$

$$\boldsymbol{\pi}_{\mathbf{k},\epsilon_k}(\mathbf{x}) = \cos(\mathbf{k}\cdot\mathbf{x}) \boldsymbol{\pi}_{\mathbf{k},\epsilon_k} + \sin(\mathbf{k}\cdot\mathbf{x}) \varepsilon_0\omega_k \mathbf{A}_{\mathbf{k},\epsilon_k}. \quad (7.62)$$

### 7.3.1 Electromagnetic Damping Kernels

We will now briefly introduce the integration kernels which will arise in our Langevin equation. Let us introduce a compact notation for the commutators of

vector operators

$$\llbracket \mathbf{X}, \mathbf{Y} \rrbracket \equiv \mathbf{X} \mathbf{Y}^T - \mathbf{Y} \mathbf{X}^T. \quad (7.63)$$

This object is a matrix whose entries are ordinary commutators. First we wish to inspect the phenomenologically simpler  $\mathbf{A}$ -damping associated with the dissipation kernel

$$\boldsymbol{\mu}_A[\mathbf{x}(t), \mathbf{y}(t'); t, t'] \equiv \left\langle \frac{1}{2} \llbracket \underline{\mathbf{A}}(\mathbf{x}(t), t), \underline{\mathbf{A}}(\mathbf{y}(t'), t') \rrbracket \right\rangle_{\text{env}}, \quad (7.64)$$

where  $\mathbf{x}$  and  $\mathbf{y}$  are quasi-static, e.g.  $\dot{x} \ll c$ . Note that the extrinsic time dependence is stationary, or to be more specific

$$\boldsymbol{\mu}_A[\mathbf{x}(t), \mathbf{y}(t'); t, t'] = \boldsymbol{\mu}_A[\mathbf{x}(t), \mathbf{y}(t'); t - t'], \quad (7.65)$$

but not the time dependence intrinsic to  $\mathbf{x}(t)$  and  $\mathbf{y}(t')$ . However, in the nonrelativistic and classical regimes the dissipation kernel is also spatially stationary in the sense

$$\boldsymbol{\mu}_A[\mathbf{x}(t), \mathbf{y}(t'); t, t'] = \boldsymbol{\mu}_A[\mathbf{x}(t) - \mathbf{y}(t'); t - t'], \quad (7.66)$$

given the nonrelativistic (quasi-stationary) approximation

$$e^{-i\mathbf{k} \cdot \mathbf{x}(t)} e^{+i\mathbf{k} \cdot \mathbf{y}(t')} = e^{-i\mathbf{k} \cdot [\mathbf{x}(t) - \mathbf{y}(t')] + \mathcal{O}(k^2[\mathbf{x}(t), \mathbf{y}(t')])}, \quad (7.67)$$

to be used when evaluating the field correlations. Note that we have exact spatial stationarity at coincident times, e.g.  $t = t'$ , and in the classical regime where the operators commute.

Next we define the damping kernel as to be positive definite and extrinsically stationary.

$$\boldsymbol{\mu}[\mathbf{x}(t), \mathbf{y}(t'); t, t'] \equiv -\frac{\partial}{\partial t'} \boldsymbol{\mu}[\mathbf{x}(t), \mathbf{y}(t'); t, t'], \quad (7.68)$$

$$\tilde{\boldsymbol{\mu}}[\mathbf{x}(t), \mathbf{y}(t'); \omega] \equiv i\omega \tilde{\boldsymbol{\gamma}}[\mathbf{x}(t), \mathbf{y}(t'); \omega], \quad (7.69)$$

where with the partial derivative and Fourier transform we neglect any intrinsic time dependence in  $\mathbf{x}(t)$  and  $\mathbf{y}(t')$ .

In the Fourier domain of the extrinsic time variables, we calculate the classical or quasi-static  $\mathbf{A}$ -coupling damping kernel to be

$$\tilde{\boldsymbol{\gamma}}_A[\mathbf{r}; \omega] = 2\gamma_0 \left\{ \tilde{S}_1\left(\left|\frac{\mathbf{r}\omega}{c}\right|\right) - \frac{1}{2} \tilde{S}_0\left(\left|\frac{\mathbf{r}\omega}{c}\right|\right) \hat{\mathbf{r}} \hat{\mathbf{r}}^T \right\}, \quad (7.70)$$

$$\gamma_0 = \frac{1}{12\pi\epsilon_0 c^3}, \quad (7.71)$$

in terms of the entire functions

$$\tilde{S}_1(z) \equiv \frac{3}{2} \frac{(z^2 - 1) \sin(z) + z \cos(z)}{z^3}, \quad (7.72)$$

$$\tilde{S}_0(z) \equiv 3 \frac{(z^2 - 3) \sin(z) + 3z \cos(z)}{z^3}. \quad (7.73)$$

This is without considering any cutoff frequency  $\Lambda$  in the structure constants or field modes. In the coincidence limit (or in the nonrelativistic limit of a point particle) one recovers the usual Ohmic damping and when far separated all cross correlations vanish.

$$\lim_{r \rightarrow 0} \tilde{\boldsymbol{\gamma}}_A[\mathbf{r}; \omega] = 2\gamma_0, \quad (7.74)$$

$$\lim_{r \rightarrow \infty} \tilde{\boldsymbol{\gamma}}_A[\mathbf{r}; \omega] = 0, \quad (7.75)$$

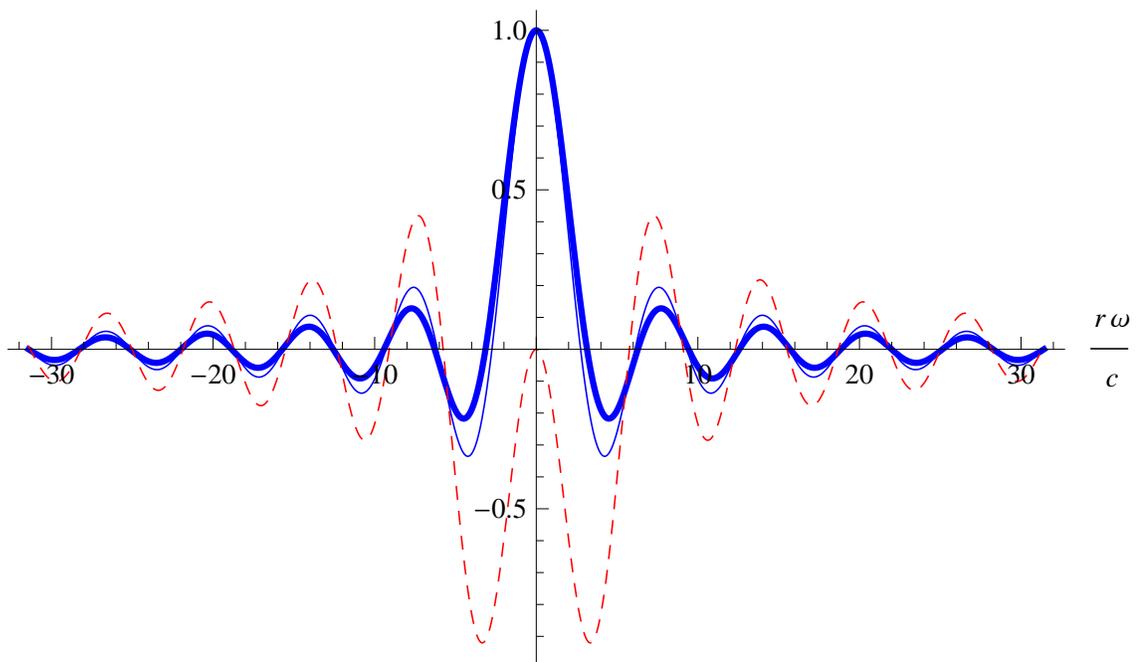


Figure 7.1: Comparison of sinc (bold),  $\tilde{S}_1$ , and  $\tilde{S}_0$  (dashed). Sinc and  $\tilde{S}_1$  are extremely qualitatively similar, both being unity at zero whereas  $S_0$  vanishes at zero.

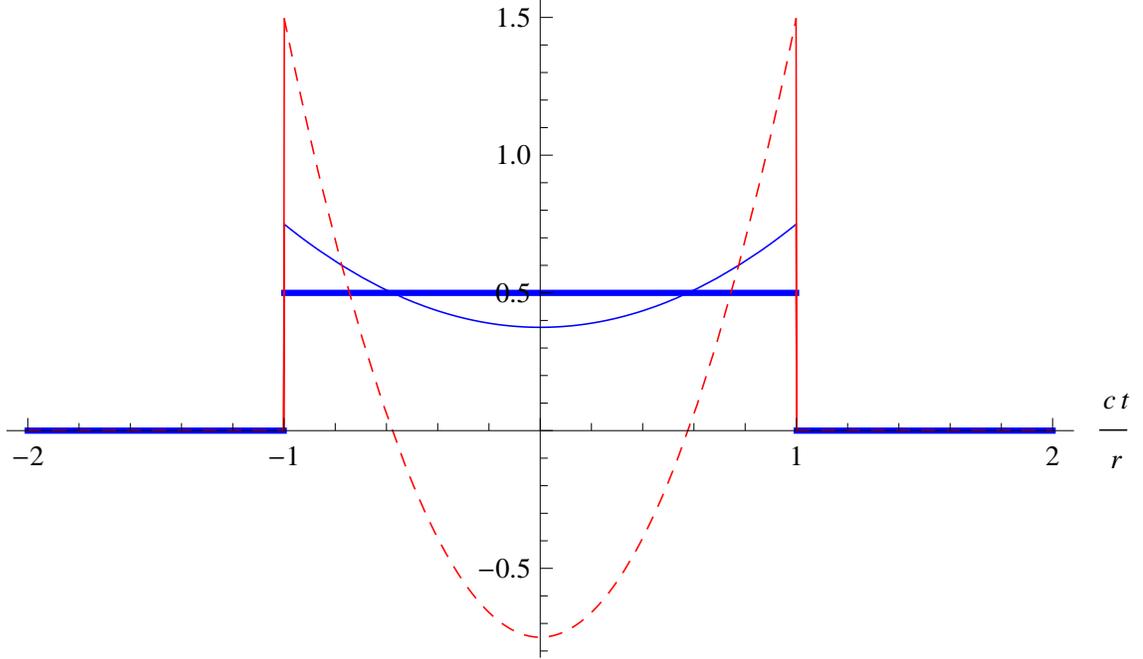


Figure 7.2: The same functions as in Fig. 7.1, but in the time domain: the rectilinear distribution (bold),  $S_1$ , and  $S_2$  (dashed).

In Fig. 7.1 we compare these special functions to  $\text{sinc}(z)$ , which is the simpler result from analogous coupling to a scalar field. In Fig. 7.2 we compare these functions in the time domain.

For  $\boldsymbol{\pi}$ -coupling to the field, we must consider the dissipation kernel associated with the correlation

$$\boldsymbol{\mu}_\pi[\mathbf{x}(t), \mathbf{y}(t'); t, t'] \equiv \left\langle \frac{1}{2} \llbracket \boldsymbol{\pi}(\mathbf{x}(t), t), \boldsymbol{\pi}(\mathbf{y}(t'), t') \rrbracket \right\rangle_{\text{env}}, \quad (7.76)$$

which is related to the  $\mathbf{A}$ -coupling via

$$\gamma_\pi[\mathbf{r}; t, t'] = -\frac{\partial^2}{\partial t'^2} \gamma_A[\mathbf{r}; t, t'], \quad (7.77)$$

$$\tilde{\gamma}_\pi[\mathbf{r}; \omega] = \omega^2 \tilde{\gamma}_A[\mathbf{r}; \omega], \quad (7.78)$$

or supra-Ohmic and thus not manifestly phenomenological. As an integration kernel  $\gamma_\pi$  is relatively pathological and must be integrated by parts twice to obtain the well-behaved kernel  $\gamma_A$ .

### 7.3.2 Coulomb-Gauge Hamiltonian

In the Coulomb gauge we choose  $\nabla \cdot \mathbf{A} = 0$ . The scalar potential  $\phi$  becomes the non-dynamical Coulomb potential and the vector potential  $\mathbf{A}$  is now purely transverse, i.e.  $\mathbf{k} \cdot \boldsymbol{\epsilon}_k = 0$ . The system + environment Hamiltonian may then be expressed

$$\mathbf{H}_{\text{QED}}^A = \sum_j \left\{ \frac{[\mathbf{p}_j - e_j \mathbf{A}(\mathbf{x}_j)]^2}{2m_j} + e_j \phi(\mathbf{x}_j) \right\} + \mathbf{H}_{\text{field}}, \quad (7.79)$$

$$\mathbf{H}_{\text{field}} \equiv \iiint d^3x' \frac{1}{2} \left\{ \varepsilon_0^{-1} \boldsymbol{\pi}(\mathbf{x}')^2 + \varepsilon_0 c^2 [\nabla \times \mathbf{A}(\mathbf{x}')]^2 \right\}. \quad (7.80)$$

By Parseval's theorem, we may also express the field Hamiltonian

$$\mathbf{H}_{\text{field}} \equiv \iiint d^3k \sum_{\boldsymbol{\epsilon}_k} \frac{1}{2} \left\{ \varepsilon_0^{-1} \boldsymbol{\pi}_{\mathbf{k}, \boldsymbol{\epsilon}_k}^2 + \varepsilon_0 \omega_k^2 \mathbf{A}_{\mathbf{k}, \boldsymbol{\epsilon}_k}^2 \right\}, \quad (7.81)$$

or as a reservoir of oscillators, given the expansions (7.55) and (7.57).

The nonrelativistic Heisenberg equations of motion for the system as driven by the field are then given by

$$\dot{\mathbf{x}}_j = \frac{\mathbf{p}_j}{m_j} - \frac{e_j}{m_j} \mathbf{A}(\mathbf{x}_j), \quad (7.82)$$

$$\dot{\mathbf{p}}_j = -e_j \nabla \phi(\mathbf{x}_j). \quad (7.83)$$

Additional contributions to  $\dot{\mathbf{p}}$ , that one might imagine from  $[[\mathbf{A}(\mathbf{x}), \mathbf{p}]]$ , vanish due to our gauge constraint. We have not invoked any nonrelativistic approximations here beyond the  $\mathbf{p}^2/2m$  system Hamiltonian.

The Heisenberg equations of motion for the environment as driven by the system are given by

$$\dot{\mathbf{A}}_{\mathbf{k},\epsilon_k} = \frac{1}{\varepsilon_0} \boldsymbol{\pi}_{\mathbf{k},\epsilon_k} + \frac{1}{\varepsilon_0 \omega_k} \sum_j \frac{e_j}{2} \{ \sin(\mathbf{k} \cdot \mathbf{x}_j), \dot{\mathbf{x}}_j \}, \quad (7.84)$$

$$\dot{\boldsymbol{\pi}}_{\mathbf{k},\epsilon_k} = -\varepsilon_0 \omega_k^2 \mathbf{A}_{\mathbf{k},\epsilon_k} + \sum_j \frac{e_j}{2} \{ \cos(\mathbf{k} \cdot \mathbf{x}_j), \dot{\mathbf{x}}_j \}. \quad (7.85)$$

Their driven solution can be expressed in the manifestly-Hermitian form

$$\mathbf{A}(\mathbf{x}_i, t) = \boldsymbol{\xi}_i^A(t) - \sum_j e_j \{ (\boldsymbol{\mu}_{ij}^A * \dot{\mathbf{x}}_j)(t) + (\dot{\mathbf{x}}_j * \boldsymbol{\mu}_{ji}^A)(t) \}, \quad (7.86)$$

in terms of the  $\mathbf{A}$ -coupling dissipation kernel discussed in Sec. 7.3.1 and where we have abbreviated the previous notation from  $\boldsymbol{\mu}_{x_i x_j}$  to  $\boldsymbol{\mu}_{ij}$ . The convolutions here are therefore evaluated as

$$(\boldsymbol{\mu}_{ij}^A * \dot{\mathbf{x}}_j)(t) = \int_0^t dt' \boldsymbol{\mu}^A[\mathbf{x}_i(t), \mathbf{x}_j(t'); t-t'] \dot{\mathbf{x}}_j(t'). \quad (7.87)$$

Without yet resolving the damping kernel, our quantum Langevin equation is given by

$$m_i \ddot{\mathbf{x}}_i(t) = -e_i \nabla \phi(\mathbf{x}_i) - e_i \dot{\boldsymbol{\xi}}_i^A(t) + \frac{d}{dt} \sum_j e_i e_j \{ (\boldsymbol{\mu}_{ij}^A * \dot{\mathbf{x}}_j)(t) + (\dot{\mathbf{x}}_j * \boldsymbol{\mu}_{ji}^A)(t) \}. \quad (7.88)$$

Note that we have sourced the field in Eq. (7.86) with velocity, as was done in [44], and as is required to obtain the standard result. It is peculiar to note that, as velocity is not a system variable, this Langevin equation cannot immediately be given a standard interpretation with  $\boldsymbol{\xi}_i^A(t)$  representing quantum-thermal noise. As  $\mathbf{v}$  and  $\boldsymbol{\pi}$  do not commute one cannot construct a factorized initial state of the form

$$\rho_{\text{sys+env}} = \rho_{\text{sys}}(\mathbf{x}, m\mathbf{v}) \otimes \rho_{\text{env}}(\mathbf{A}, \boldsymbol{\pi}), \quad (7.89)$$

and thus one cannot sample  $\xi_i^A(t)$  from a statistically-independent thermal distribution using the canonical coordinates of the Hamiltonian. However, the Hamiltonian of the following section will provide a satisfactory perspective in this regard, and the resultant dynamics appear to be equivalent to for a single particle in the dipole approximation. [More generally this is not so transparent.] In Sec. 7.5 we will opt for the more (canonically and thermodynamically) natural choice of momentum as the source in our effective theory.

### 7.3.3 Position-Coupling Hamiltonian

Equivalently, one may represent the vector interaction

$$e \dot{\mathbf{x}} \cdot \mathbf{A}(\mathbf{x}) = e \mathbf{x} \cdot \dot{\mathbf{A}}(\mathbf{x}) - e \frac{d}{dt}[\mathbf{x} \cdot \mathbf{A}(\mathbf{x})], \quad (7.90)$$

and neglect the total derivative in the action, as was done in [15]. This choice of gauge results in different notion of momentum for the system and environment.

Applying interaction (7.90) one obtains the system + environment Hamiltonian

$$\begin{aligned} \mathbf{H}_{\text{QED}}^\pi = & \sum_j \left\{ \frac{\mathbf{p}_j^2}{2m_j} + e_j \phi(\mathbf{x}_j) \right\} \\ & + \iiint d^3x' \left\{ \frac{1}{2\varepsilon_0} [\boldsymbol{\pi}(\mathbf{x}') - \rho(\mathbf{x}, \mathbf{x}') \mathbf{x}]^2 + \frac{\varepsilon_0 c^2}{2} [\nabla \times \mathbf{A}(\mathbf{x}')]^2 \right\}, \end{aligned} \quad (7.91)$$

which takes analogous form to the QBM Hamiltonian (7.9). Note that the (nonrelativistically) bilinear interaction here is

$$\mathbf{H}_{\text{int}} = - \sum_j e_j \mathbf{x}_j \cdot \boldsymbol{\pi}(\mathbf{x}_j), \quad (7.92)$$

and as in QBM the remaining  $\rho^2 \mathbf{x}^2$  term is an appropriate system-potential renormalization so that  $\phi$  is the renormalized potential.

The nonrelativistic Heisenberg equations of motion for the system as driven by the field are then given by

$$\dot{\mathbf{x}}_j = \frac{\mathbf{P}_j}{m_j}, \quad (7.93)$$

$$\dot{\mathbf{p}}_j = -e_j \nabla \phi_{\text{bare}}(\mathbf{x}_j) + e_j \boldsymbol{\pi}(\mathbf{x}_j), \quad (7.94)$$

and the Heisenberg equations of motion for the environment as driven by the system are given by

$$\dot{\mathbf{A}}_{\mathbf{k}, \epsilon_k} = \frac{1}{\epsilon_0} \boldsymbol{\pi}_{\mathbf{k}, \epsilon_k} - \sum_j e_j \cos(\mathbf{k} \cdot \mathbf{x}_j) \mathbf{x}_j, \quad (7.95)$$

$$\dot{\boldsymbol{\pi}}_{\mathbf{k}, \epsilon_k} = -\epsilon_0 \omega_k^2 \mathbf{A}_{\mathbf{k}, \epsilon_k} + \epsilon_0 \omega_k \sum_j e_j \sin(\mathbf{k} \cdot \mathbf{x}_j) \mathbf{x}_j. \quad (7.96)$$

Their driven solution can be expressed in the manifestly-Hermitian form

$$\boldsymbol{\pi}(\mathbf{x}_i, t) = \boldsymbol{\xi}_i^\pi(t) - \sum_j e_j \{ (\boldsymbol{\mu}_{ij}^\pi * \mathbf{x}_j)(t) + (\mathbf{x}_j * \boldsymbol{\mu}_{ji}^\pi)(t) \}, \quad (7.97)$$

in terms of the  $\boldsymbol{\pi}$ -coupling dissipation kernel discussed in Sec. 7.3.1. Without yet resolving the damping kernel, our quantum Langevin equation is given by

$$m_i \ddot{\mathbf{x}}_i(t) = -e_i \nabla \phi_{\text{bare}}(\mathbf{x}_i) + e_i \boldsymbol{\xi}_i^\pi(t) - \sum_j e_i e_j \{ (\boldsymbol{\mu}_{ij}^\pi * \mathbf{x}_j)(t) + (\mathbf{x}_j * \boldsymbol{\mu}_{ji}^\pi)(t) \}. \quad (7.98)$$

### 7.3.4 The Single-Particle Nonrelativistic Theory

Now we will consider the well-known result of a single nonrelativistic particle, for which the dipole approximation is valid. Integrating the convolution in Langevin equation (7.88) by parts once and discarding the slip we obtain the Langevin equa-

tion

$$m\ddot{\mathbf{x}}(t) = -e\nabla\phi(\mathbf{x}) - 2e^2(\gamma^\pi * \dot{\mathbf{x}})(t) + e\dot{\boldsymbol{\xi}}^\pi(t), \quad (7.99)$$

which is runaway free for  $m > 0$  and reasonable  $\phi(\mathbf{x})$ , as proven in Sec. 7.2.5. However  $m$  will not turn out to be the phenomenological mass of the system. Discarding the transient slip, we can integrate the convolution in Langevin equation (7.88) by parts twice to obtain Langevin equation (7.98); they are equivalent. From Langevin equation (7.98), we can integrate by parts once to obtain

$$m\ddot{\mathbf{x}}(t) = -e\nabla\phi(\mathbf{x}) - e\dot{\boldsymbol{\xi}}^A(t) + 2e^2\frac{d}{dt}\{(\gamma^A * \ddot{\mathbf{x}})(t) + \gamma^A(t)\dot{\mathbf{x}}(0) - \gamma^A(0)\dot{\mathbf{x}}(t)\}. \quad (7.100)$$

Discarding the transient slips and reordering like terms we have the time-nonlocal Abraham-Lorentz equation with noise.

$$m_{\text{ren}}\ddot{\mathbf{x}}(t) = -e\nabla\phi(\mathbf{x}) + 2e^2(\gamma^A * \ddot{\mathbf{x}})(t) - e\dot{\boldsymbol{\xi}}^A(t), \quad (7.101)$$

$$m_{\text{ren}} = m_{\text{bare}} + 2e^2\gamma^A(0) \quad (7.102)$$

where we have staved off the remaining local damping approximation as to keep  $\gamma(0)$  finite, however  $(\gamma^A * \ddot{\mathbf{x}})(t) \approx \gamma_0 \ddot{\mathbf{x}}(t)$ . Note the relationship between the bare and renormalized masses in Eq. (7.102). If the phenomenological mass is to be finite, and the bare mass positive, then the theory requires a finite cutoff so that  $\gamma^A(0) \propto \gamma_0 \Lambda$  is finite and also not too large. The time-local theory admits runaway solutions because the mass renormalization is allowed to diverge, making the bare mass negative. This theory requires a U.V. cutoff bounded above on the order of  $\varepsilon_0 c^3 m_{\text{ren}} / e^2$ .

## 7.4 The Equivalence of Repairs

Ford & O'Connell [70] have already shown that all one requires to repair the non-relativistic theory is to enforce a cutoff that is not too large, so that the bare mass of the system remains positive. Here we wish to briefly demonstrate that other physically motivated repairs are equivalent to enforcing a cutoff.

It has long been known that runaway solutions can be avoided if the classical particle is made sufficiently large in extent (e.g., see [117]). It was also realized that this repair kept the mechanical mass of the system positive. With this fact in mind, note that instead of inserting a cutoff regulator into our integration kernel, we may instead choose to not evaluate the field operators at precisely the same location, effectively enforcing a smallest resolution  $\delta r$  as depicted in Fig 7.3. Note from Eq. (7.70) that this is equivalent to a specific cutoff regulator with U.V. cutoff  $\Lambda = c/\delta r$ . Therefore, by extending the classical particle, one is essentially introducing an extraordinarily complicated cutoff regulator, with a cutoff on the order of  $c/\text{length}$ .

Another repair which has been occasionally proposed is to delay the effect of the backreaction. Consider for instance, the substitution in the Abraham-Lorentz equation

$$\gamma_0 \dot{\mathbf{v}}(t) \rightarrow \gamma_0 \frac{\mathbf{v}(t) - \mathbf{v}(t - \delta t)}{\delta t}. \quad (7.103)$$

If we take the Laplace transform, then we find this equivalent to the convolution

$$\gamma_0 \dot{\mathbf{v}}(t) \rightarrow (\gamma * \dot{\mathbf{v}})(t), \quad (7.104)$$

$$\tilde{\gamma}(\omega) = \gamma_0 \frac{1 - e^{-i\omega\delta t}}{i\omega\delta t}, \quad (7.105)$$

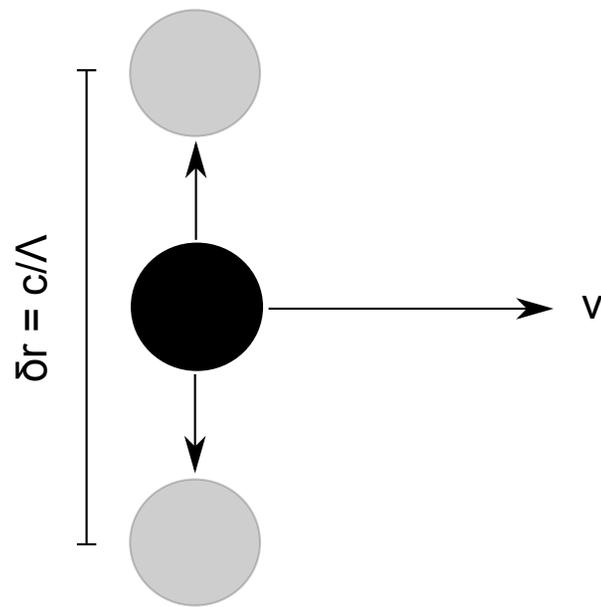


Figure 7.3: Point-separation regulation.

and so one is simply introducing the cutoff  $\Lambda = 1/\delta t$ .

Finally, we should mention that  $\gamma(t, t')$  does need to be positive definite and so  $\tilde{\gamma}(\omega)$  needs to be positive. The above two (simple) regulators are not always positive, and they require a bit more work to get into a positive definite form. For the point-separation regulator one really needs to consider a continuous smearing regulator or charge distribution.

## 7.5 Effective Theory of Nonrelativistic QED

Ford & O'Connell (FO) have shown that the Abraham-Lorentz equation is perturbatively dissipative by iterating the local limit of Eq. (7.101) and perturbatively matching it to their equation

$$m\ddot{\mathbf{x}} = \mathbf{F}_{\text{ext}}(\mathbf{x}) + \frac{2e^2\gamma_0}{m} \frac{d}{dt} \mathbf{F}_{\text{ext}}(\mathbf{x}), \quad (7.106)$$

here for the classical vacuum. Eq. (7.106) is manifestly causal and can be proven to be runaway free. As the Abraham-Lorentz equation must be viewed as the perturbative truncation of a series expansion in  $1/c$ , we find the FO result to be perfectly satisfactory in analyzing the stability of the non-relativistic Langevin equation.

However the FO equation does share behaviors with the Abraham-Lorentz (and Abraham-Dirac-Lorentz) equation that some find peculiar. In particular, dissipation does not occur during uniform acceleration as the Larmor formula might suggest, but only in the ramp up to uniform acceleration and the ramp down from uniform acceleration. To investigate the physical basis of these behaviors, we would like to motivate a more direct microscopic derivation of the FO equation from an

effective Hamiltonian theory.

First, we choose the  $\mathbf{A}$ -coupling Hamiltonian (7.79), as the vector potential has a phenomenological Ohmic-like damping kernel. The alternative  $\boldsymbol{\pi}$ -coupling is relatively pathological and must be carefully manipulated into a form like  $\mathbf{A}$ -coupling to give phenomenological results. [See Sec. 7.3.1 for specific calculations.] Secondly, unlike the standard derivation we gave in Sec. 7.3.2, here we will restrict ourselves to the canonical system and environment partitioning so that we have an immediate and obvious notion of thermal noise. From these two choices, the following results are essentially determined.

We consider the following effective Hamiltonian in the Coulomb gauge and dipole approximation

$$\mathbf{H}_{\text{EQED}} = \frac{\mathbf{p}^2}{2m} - \frac{e}{m} \mathbf{p} \cdot \mathbf{A} + e \phi(\mathbf{x}) + \mathbf{H}_{\text{field}}, \quad (7.107)$$

where we have dropped the  $\mathbf{A}^2$  term for several reasons. Primarily we neglect  $\mathbf{A}^2$  because it is a higher-order term which describes photon-photon interactions that would only produce effects in the open system at an order which we have already neglected. Secondly, if we were to keep the  $\mathbf{A}^2$  then we would be confronted with the strange task of handling it appropriately. Note that as a field operator  $\mathbf{A}^2$  is as large as  $\mathbf{H}_{\text{field}}$ . If not treated in some perturbative manner,  $\mathbf{A}^2$  would have to be initially equilibrated as to produce thermal fluctuations in the open system. This pathology appears to be an artifact of our choice of gauge and the nonrelativistic limit, as in the corresponding relativistic Hamiltonian

$$\mathbf{H}_{\text{rel}} = \sqrt{(mc^2)^2 + (\mathbf{p} - e \mathbf{A})^2} + e \phi(\mathbf{x}), \quad (7.108)$$

no such large interaction terms occur as to require such peculiar equilibration in the initial state of the system and environment. It is also likely that including these self-interactions in the electromagnetic-field dynamics would further complicate the renormalization of this effective theory.

Next we note that in this formalism  $\mathbf{p}$  is the system variable and not  $\dot{\mathbf{x}}$ , therefore we naturally choose to source the field equations of motion with the system momentum and not the velocity. As explained in Sec. 7.3.2, as velocity is not a system variable, if the velocity source is chosen then the resulting Langevin equation cannot immediately be given a standard interpretation with  $\boldsymbol{\xi}(t)$  representing quantum-thermal noise. Given that  $\dot{\mathbf{x}}$  and  $\boldsymbol{\pi}$  do not commute, one cannot construct a factorized initial state of the form

$$\rho_{\text{sys+env}} = \rho_{\text{sys}}(\mathbf{x}, m\dot{\mathbf{x}}) \otimes \rho_{\text{env}}(\mathbf{A}, \boldsymbol{\pi}), \quad (7.109)$$

and thus one cannot sample  $\boldsymbol{\xi}(t)$  from a statistically-independent thermal distribution using the canonical coordinates of the Hamiltonian.

Given our canonical choice of source, we obtain the following driven solution of the field in the (nonrelativistic) dipole approximation

$$\mathbf{A}(t) = \underbrace{\boldsymbol{\xi}(t)}_{\text{noise}} - \underbrace{2e\gamma_0 \dot{\mathbf{p}}(t)}_{\text{damping}} + \underbrace{2e\gamma(0) \mathbf{p}(t)}_{\text{renormalization}}, \quad (7.110)$$

and post-slip. Note that Eq. (7.110) gives a perturbatively different (yet perturbatively consistent) account of backreaction as compared the result of velocity driving in Eq. (7.86). As argued above, given Hamiltonian (7.107), momentum driving is the natural choice if one desires thermal noise and its associated dissipation. In fact,

from the perspective here, the choice of velocity driving entails that the environment is being allowed to act as its own source and drive itself, to a perturbative degree. Given this perturbative feedback loop in the environment dynamics, it is therefore not surprising that there would be perturbative runaway behavior as a result. From this perspective, it is also the case that the “backreaction” in Eq. (7.86) actually contains some perturbative amount of hidden noise and the “noise” therein actually contains some perturbative amount of hidden backreaction. So when one takes the classical vacuum limit of (7.86), one is actually enforcing an artificial, but perturbative, constraint upon the backreaction of the open system. Again, it is therefore no surprise that this would result in a perturbative runaway behavior.

Given the choice of momentum driving, the resultant open-system equations of motion are then given by

$$\dot{\mathbf{x}}(t) = \frac{\mathbf{p}(t)}{m_{\text{ren}}} + 2 e^2 \gamma_0 \frac{\dot{\mathbf{p}}(t)}{m_{\text{bare}}} - \frac{e}{m_{\text{bare}}} \boldsymbol{\xi}(t), \quad (7.111)$$

$$\dot{\mathbf{p}}(t) = \mathbf{F}_{\text{ext}}(\mathbf{x}), \quad (7.112)$$

in terms of the renormalized mass

$$\frac{1}{m_{\text{ren}}} = \frac{1}{m_{\text{bare}}} + 2 e^2 \gamma(0), \quad (7.113)$$

and consistent with this order of perturbative analysis we may express our Langevin equations

$$\dot{\mathbf{x}}(t) = \frac{\mathbf{p}(t)}{m_{\text{ren}}} + 2 e^2 \gamma_0 \frac{\dot{\mathbf{p}}(t)}{m_{\text{ren}}} - \frac{e}{m_{\text{ren}}} \boldsymbol{\xi}(t), \quad (7.114)$$

$$\dot{\mathbf{p}}(t) = \mathbf{F}_{\text{ext}}(\mathbf{x}). \quad (7.115)$$

In this perspective, the renormalization of the  $\mathbf{p}^2/2m$  mass and the  $e/m(\mathbf{p} \cdot \mathbf{A})$  mass enter at different orders. At this order we only have determined the first renormalization, which we may then perturbatively extend to the second, under the critical assumption that everything will naturally resolve at higher orders.

Note carefully the dramatic difference in the mass renormalization here in Eq. (7.113) as contrast to the standard mass renormalization in Eq. (7.102). *To obtain a finite dressed mass, the standard calculation may require a large negative bare mass. However, here that never happens.* Instead, one might require a vanishing positive bare mass.

Finally note that when combined, relations (7.114) and (7.115) exactly reproduce the structureless FO equation (7.106). Thus we have constructed a microscopic model which directly generates a well-behaved Langevin equation. Our reward for this task is that in doing so we have discovered a very different perspective for mass renormalization, which appears far more well behaved.

### 7.5.1 Proof of Stability

Although it is already known that Eq. (7.106) is runaway free, and it is obviously causal, we would like to give a more canonical derivation of this fact from within the confines of our formalism. We will now show that the dynamics of the open system are dissipative and stable in a manner analogous to the QBM proof given in Sec. 7.5.1. Let us denote the system Hamiltonian

$$\mathbf{H}_{\text{sys}} \equiv \frac{\mathbf{p}^2}{2m} + U(\mathbf{x}). \quad (7.116)$$

One may then calculate an energy constraint from either the Heisenberg equations of motion for  $\mathbf{H}_{\text{sys}}(t)$  or by integrating the (classical) Langevin equation (7.114) along with the second (7.115). Accounting for the slip in our initial state, which only produces a finite change in energy, we obtain the relation

$$\mathbf{H}_{\text{sys}}(t) = \mathbf{H}_{\text{sys}}(0) - \mathbf{H}_{\gamma}(t) + \mathbf{H}_{\xi}(t), \quad (7.117)$$

in terms of the energy lost to damping  $\mathbf{H}_{\gamma}(t)$  and energy generated by noise  $\mathbf{H}_{\xi}(t)$

$$\mathbf{H}_{\gamma}(t) = 2 \frac{e^2 \gamma_0}{m} \int_0^t dt' \dot{\mathbf{p}}(t')^2, \quad (7.118)$$

$$\mathbf{H}_{\xi}(t) = \frac{e}{m} \int_0^t dt' \frac{1}{2} \{ \boldsymbol{\xi}(t') \cdot \dot{\mathbf{p}}(t') + \dot{\mathbf{p}}(t') \cdot \boldsymbol{\xi}(t') \}. \quad (7.119)$$

The contribution from damping is a negative quantity and it monotonically increases in magnitude. The noise is random and may drive the system erratically, but the damping may only remove energy from the system (and deliver it to the environment and interaction). In the classical-vacuum limit, energy is continually siphoned from  $\mathbf{H}_{\text{sys}}(t)$  so long as external forces are applied.

It is important to note that the system here is given by the canonical definition, and so  $\mathbf{H}_{\text{sys}}$  does not correspond to the mechanical energy of the particle. From Eq. (7.114) the velocity and momentum differ by the vector potential, which itself includes both backreaction and noise. If the system momentum  $\mathbf{p}$  relaxes under dissipative motion, then so must  $\dot{\mathbf{p}}$  and by extension the backreaction  $\gamma_0 \dot{\mathbf{p}}$ . Given noise, and with the slip accounted for, the system velocity may only fluctuate around the stable average

$$\langle m \mathbf{v} \rangle_{\boldsymbol{\xi}} = \langle \mathbf{p} + 2 e^2 \gamma_0 \dot{\mathbf{p}} \rangle_{\boldsymbol{\xi}}, \quad (7.120)$$

and so the system velocity must also be stable. If no external forces are being applied, then the canonical and mechanical momenta will limit towards each other (on average, given noise) in the late-time limit.

Note that these energy constraints apply to the open system and only relate the system (particle) energy to backreaction. Although the terms backreaction and radiation reaction are sometimes used interchangeably, we prefer the former term because the connection between dissipative backreaction and energy transfer into radiation is not immediate. For the full particle plus field system, it is the Hamiltonian (7.107) which is conserved, and energy may be stored and transferred between particle, field, and interaction terms in complex ways. The field itself may also be partitioned into near and far field degrees of freedom. Energy dissipated due to backreaction does not immediately or necessarily monotonically convert into energy in the radiation (the far fields), and thus there is no reason to expect the radiated power to instantaneously match the dissipated power. It would actually be peculiar for such a conversion to occur exactly, outside of some kind of steady-state process. The important implication of the energy constraint above is that it shows that *backreaction* never causes a net *increase* of the energy of the particle at the expense of the field (as happens for runaway solutions).

## 7.6 Discussion

We have derived new stochastic equations of motion (7.114)-(7.115) for a non-relativistic charged particle in the electromagnetic field. The resulting dynamics as

combined into (7.106) are equivalent to the previous results of Ford & O’Connell, however our underlying microscopic derivation approaches the problem from a very different perspective. As a result we are allowed a vantage point from whence renormalization never causes the bare mass of the system to be negative, which is the ultimate cause of instability in the standard result.

Our derivation differs from the standard treatment in that first we choose our coupling gauge,  $\dot{\mathbf{x}} \cdot \mathbf{A}$  coupling versus  $\mathbf{x} \cdot \dot{\mathbf{A}}$  coupling in the Lagrangian, based upon phenomenological and mathematical considerations of the field correlations: the former is well behaved whereas the latter is relatively pathological. From the choice of  $\dot{\mathbf{x}} \cdot \mathbf{A}$  coupling in the Lagrangian, one then has  $\mathbf{p} \cdot \mathbf{A}$  coupling in the Hamiltonian. Our second and more important step is to then realize that  $\mathbf{p}$  is the proper system variable for nonequilibrium thermodynamics, and not  $\dot{\mathbf{x}}$ . The generation of thermal noise in the Langevin equation dictates that the field be driven by momentum and not velocity. Sourcing the field with momentum yields a perturbatively different (yet perturbatively consistent) account of backreaction, but a dramatically different account of mass renormalization. From thereon we consistently treat our Hamiltonian as an effective and nonrelativistic Hamiltonian, and we are able to derive naturally causal and stable Langevin equations.

Comparing our equations of motion to the standard equations, while in the same coupling gauge, we are able to see that the standard velocity-driving “noise” (even when taken to vanish) is not thermal and actually contains a small portion of backreaction. Additionally, the environment is allowed to (perturbatively) act as a source for its own dynamics. When this false noise is made to vanish, the

physical correspondence of this situation is not to the classical vacuum environment, but a constrained environment dynamic whereupon the environment is made to endlessly self interact and drive itself in a recursive manner. This feedback loop in the environment ultimately leads to runaway solutions in the system. Our equations of motion contain truly thermal noise and accompanying truly resistive dissipation. As such, runaway solutions cannot occur for us.

In conclusion, there are several clear avenues for future analysis. First and primarily, the relativistic regime should be considered with a similar formalism as presented herein, or at least one that appeals to thermodynamic considerations. The fully-relativistic problem is far more challenging, both due to the nonlinearity of the system Hamiltonian and due to the fact that the dipole approximation cannot be made. Perhaps some easier progress can be made by bridging the Dirac-Bhabha family of backreactions to an appropriate canonical formalism, as is more natural for nonequilibrium thermodynamics. Secondly, multiple charged particles should be considered. Here in the nonrelativistic regime, the magnetostatic interactions should be derivable from first principles. It would be interesting to compare such results to those of the Darwin Lagrangian.

## Chapter 8

### Discussion

#### 8.1 Summary of Key Findings

To summarize our findings, in Chapter 2 we reported our weak-coupling solutions to the general problem of quantum open systems. This entailed a translation of canonical perturbation theory from closed systems to open systems. Subtleties involved revealed a loss of accuracy inherent in the solutions of all weak-coupling master equations, which we detailed further in Sec. 4.2. We were also able to apply a more general interpretation of the completely-positive semi-group theorem to the non-Markovian regime. This led to a natural measure of decoherence strength, which we detailed further in Sec. 4.7. We gave a somewhat thorough study of the environment correlation function, influence kernels and their respective relation. One consequence of this was the discovery of a fluctuation-dissipation inequality (essentially the Heisenberg uncertainty principle for environments), which we detailed further in Sec. 4.6. Finally we gave a three-way derivation of the (equilibrium) fluctuation-dissipation relation: from the microscopic theory, from observable invariance in the environment, and from detailed balance (or observable independence in the system).

From this perturbative formalism we derived a number of additional results in Chapter 4. In Sec. 4.5 we detailed the reduced equilibrium states of open systems,

given environments initially in equilibrium, wherein we demonstrated a three-way correspondence among the global equilibrium state, the master equation and the Schrödinger equation. In Sec. 4.3 we detailed how to handle the preparation of properly correlated initial states without abandoning the linear formalism, effectively killing two birds with one stone. In Sec. 4.4 we studied the widely used *rotating-wave approximation* (RWA) in its application to open-system dynamics. It was seen that (away from resonance) the pre-trace RWA interaction Hamiltonian generated the correct perturbative relaxation rates, whereas the post-trace RWA Lindblad equation generated all perturbative timescales correctly.

In Chapter 3 we reported our exact solutions to the problem of quantum Brownian motion. Our contribution to this great lineage of work was multifold. Perhaps most importantly, we pointed out a critical subtlety which caused all previous derivation of the diffusion coefficients to be incorrectly specified for nonlocal damping. We were also able to extend QBM to multipartite systems of arbitrary system-system and system-environment couplings. Finally, we derived many useful relations which allowed us to calculate exact solutions via contour integration. An exact calculation in the supra-Ohmic regime revealed the conventional classification of spectral-density functions to be inadequately regulated.

In Chapter 5 we essentially perturbed off of the QBM solutions of Chapter 3, to derive the dynamics of strong-coupling perturbation. We were able to rigorously prove this calculation to be compatible with all aspects of regular QBM, however we have yet to properly apply this formalism to more general problems.

In Chapter 6 we applied the formalisms of Chapters 2 & 3 to the consideration

of multipartite quantum systems immersed in a single quantum field. Such problems are inherently non-Markovian due to the intricate space-time correlations of the dynamical, relativistic field. From our perturbative formalism we knew perturbative master equations (including Born-Markov, RWA Lindblad, etc.) were incapable of calculating entanglement to second order. In contrast to previous claims, we found that all initial states of two-level atoms undergo finite-time disentanglement. We additionally discovered them to be rather different from previously studied harmonic atoms, in that harmonic atoms grow more entangled with proximity whereas two-level atoms grow more disentangled.

Finally, in Chapter 7 we revisited the infamous Abraham-Lorentz force. Mathematical intuitions from the previous chapters are what lead us to reconsider this problem from a (canonical) open-systems perspective. It was shown that the standard calculation of the Abraham-Lorentz force either relies upon (1) field correlations which are more pathological than what is necessary or (2) a perspective which does not readily noise which is truly thermal. Our equations of motion use the more well-behaved field correlations and admit truly thermal noise (within the context in which they are derived) and were rigorously proven to be stable and causal, even in the limit of local damping. Instead of the  $\ddot{\mathbf{x}}$  Abraham-Lorentz force, we obtained a  $\ddot{\mathbf{p}}$  damping backreaction.

## 8.2 Future Directions

It is difficult to summarize all possible outgrowths of study from this body of work, but we shall briefly discuss a handful of important future studies that should be considered. Perhaps most importantly, our discovery of accuracy loss in the solutions to all weak-coupling master equations has dealt an extremely unfortunate blow to an entire field of study within physics. Second-order master equations, as we have largely relied upon in this work, are extremely ubiquitous due to them being moderately easy to calculate for arbitrary systems and environments. Fourth-order master equations, which one needs for full second-order solutions, are extremely unwieldy, and to our knowledge, have only only been calculated in two very particular cases: a single oscillator or a single two-level atom bilinearly coupled to bosonic bath. If one could resolve some simplified expressions for the fourth-order coefficients, as has been done countless times for the second-order master equation coefficients, this would be a great boon to the field.

Next there likely remains a great deal of further interest in the back-reaction of charged particles in the electromagnetic field. The non-relativistic calculation came out very cleanly, largely because the theory becomes effectively linearized in the non-relativistic limit. Still in the non-relativistic limit, it might be worthwhile to investigate more general classical distributions of charge and what effect that has upon the environment's dispersion and resulting back-reaction. It would also be worthwhile to restrict ourselves to the classical point-particle case and push into fully relativistic theory, perhaps with a combined canonical and covariant treatment

of the problem. Finally we must also consider more accurate calculation of the nonlocal damping and its effects.

One program that we are presently working on is the application of our dynamically-generated and properly-correlated initial states to QBM. We have determined a way of swapping states which remains linear, thus keeping the problem (formally) exactly solvable. It would be a meritable accomplishment to calculate non-perturbative and full-time solutions which are also free of initial-time pathologies, though nothing (otherwise) interesting is expected to happen.

Finally, we need to apply our newly discovered strong-coupling formalism to some appropriate problems. This should likely start with the explicit calculation of the free-particle two-time thermal covariance, perhaps restricted to the case of local damping and zero temperature. Ideally it would be desirable to apply both strong-coupling and weak-coupling to the same problem, however the class of problems which are most easily tractable in each formalism does not appear to overlap very much.

## Appendix A

### Canonical-Like Perturbation Theory with Time Dependence

#### A.1 Introduction

In this appendix we present some mathematical formalisms to obtain the perturbative solutions of certain time-dependent Hamiltonian or Liouvillian dynamics, which is in the spirit of canonical perturbation theory as used for time-independent Hamiltonian or Liouville operators. Specifically we consider time-periodic and asymptotically-stationary dynamics, as these relate to the categories of problems we encountered in Chapter 2.

Canonical perturbation theory works exceptionally well for time-independent dynamics because it fits to the exact form of solution, including all time dependence therein. This is in contrast to the time-dependent perturbation theory of Dirac, which is not nearly as accurate in its most general application. In a well-controlled perturbative expansion, one wants to know to what order their solution is good and also that errors do not become large when other parameters (e.g. time) are varied greatly. The motivating idea behind this work is that one can perform well-controlled perturbation theory in time-dependent problems, by respecting some degree of time-dependent structure which must be present in the solutions. Therefore we do not aim to present a general method for integrating arbitrary time-dependent dynamics, but specialized integrators aimed at particular kinds of time-dependent dynamics,

for which some known mathematical structure must be present.

We explicitly apply this concept for two classes of time-dependent problems: time-periodic and asymptotically stationary (time-independent) dynamics. The former can be utilized to inspect the validity of the so-called rotating-wave approximation.

## A.2 Homogeneous Dynamics

In canonical perturbation theory, as used for time-independent Hamiltonians in quantum mechanics, we have a linear system of ordinary differential equations with time-independent coefficients

$$\dot{\mathbf{X}}(t) = \mathbf{A} \mathbf{X}(t), \quad (\text{A.1})$$

here written in the form of a system of first-order ODE's. The time-translation generator  $\mathbf{A}$  is expanded perturbatively as

$$\mathbf{A} = \sum_{k=0} \mathbf{A}_k, \quad (\text{A.2})$$

such that  $\mathbf{A}_k = \mathcal{O}(g^k)$  in some perturbative parameter  $g$ . and for which we know how to solve the zeroth-order problem exactly. I.e. to lowest order we know the eigen-system

$$\mathbf{A} \mathbf{u}_i = \lambda_i \mathbf{u}_i, \quad (\text{A.3})$$

$$\mathbf{u}_i^* \mathbf{A} = \lambda_i \mathbf{u}_i^*, \quad (\text{A.4})$$

with the orthonormal relation

$$\mathbf{u}_i^* \mathbf{u}_j = \delta_{ij}. \quad (\text{A.5})$$

In closed-system quantum mechanics the left and right eigen-vectors are adjoint, whereas in open-system quantum mechanics the time evolution is not unitary and so the left and right eigen vectors can be very different (see Sec. 2.2.2.2). For open systems, the eigen-vectors noted here are not vectors in Hilbert space but *super-vectors* in the space of operators which act upon Hilbert space vectors.

In this work we focus primarily upon non-degenerate perturbation theory and consider the treatment of degeneracy (and near degeneracy) to be resolved by a redefinition of expansion (A.2), where the degenerate subspace-subspace dynamics is redefined back into  $\mathbf{A}_0$  and solved for exactly. The remainder of our results will assume this procedure has been carried out if necessary.

As we know the exact solution is of the form

$$\mathbf{X}(t) = \mathbf{G}(t) \mathbf{X}(0), \quad (\text{A.6})$$

$$\mathbf{G}(t) = e^{t\mathbf{A}}, \quad (\text{A.7})$$

it then suffices find a perturbative expansion of the eigen-system of  $\mathbf{A}$ , as we can express the transition matrix via the spectral decomposition

$$e^{t\mathbf{A}} = \sum_i e^{\lambda_i t} \mathbf{u}_i \mathbf{u}_i^*. \quad (\text{A.8})$$

Solutions in this form are always qualitatively correct and non-secular in time, regardless of the fact that they are perturbative.

### A.3 Periodic Dynamics

Now suppose we have a linear system of parametric (ordinary) differential equations of the form

$$\dot{\mathbf{X}}(t) = \mathbf{A}(t) \mathbf{X}(t), \quad (\text{A.9})$$

where  $\mathbf{A}(t)$  has period  $\mathcal{T}$ . The exact solution can be expressed

$$\mathbf{X}(t) = \mathbf{G}(t) \mathbf{X}(0), \quad (\text{A.10})$$

$$\mathbf{G}(t) = \lim_{d\tau \rightarrow 0} \prod_{\tau=0}^t e^{d\tau \mathbf{A}(\tau)}. \quad (\text{A.11})$$

Even when discretized,  $d\tau > 0$ , solutions of this form preserve the Lie group structure, e.g. unitarity. Furthermore, Floquet's theorem states that the solution may be represented

$$\mathbf{G}(t) = \mathbf{P}(t) e^{t\mathbf{F}}, \quad (\text{A.12})$$

where  $\mathbf{P}(t)$  is an operator, also with period  $\mathcal{T}$ , which transforms  $\mathbf{X}(t)$  into a rotating basis wherein the dynamics are driven by the time-independent  $\mathbf{F}$ . Therefore the eigen-values of  $\mathbf{F}$  determine stability (exponential decay and growth) just as in ordinary homogeneous evolution. Also note that given the Lie-group nature of our solutions,  $\mathbf{P}(t)$  can be expressed as an exponential matrix, or product of exponential matrices, though it is not necessarily time-translation invariant, and in closed-system quantum mechanics  $\mathbf{P}(t)$  is unitary. Therefore relations (A.11)-(A.12) comprise the structure from which we will apply perturbation theory.

Next assume we have a perturbative expansion of the time-translation gener-

ator

$$\mathbf{A}(t) = \sum_{k=0} \mathbf{A}_k(t), \quad (\text{A.13})$$

where  $\mathbf{A}_k(t) = \mathcal{O}(g^k)$  in some perturbative parameter  $g$ , and such that we have exact zeroth-order solutions

$$\mathbf{G}_0(t) = \mathbf{P}_0(t) e^{t\mathbf{F}_0} \mathbf{X}(0), \quad (\text{A.14})$$

$$\dot{\mathbf{G}}_0(t) = \mathbf{A}_0(t) \mathbf{G}_0(t). \quad (\text{A.15})$$

We will proceed to find perturbative solutions in Floquet normal form, Eq. (A.12), which also respect the Lie group structure in Eq. (A.11). The only assumption we will make is that the period  $\mathcal{T}$  is respected to all orders in  $\mathbf{A}(t)$ . This will be a generalization of the *Floquet-Magnus series* found in Ref. [33].

The first step is to shift all time dependence into the higher-order perturbative terms of  $\mathbf{A}(t)$ . We do this by making a change of basis  $\mathbf{G}(t) = \mathbf{P}_0(t) \underline{\mathbf{G}}(t)$  whereupon we arrive at the equation of motion

$$\dot{\underline{\mathbf{G}}}(t) = \left[ \mathbf{F}_0 + \sum_{k=1} \underline{\mathbf{A}}_k(t) \right] \underline{\mathbf{G}}(t), \quad (\text{A.16})$$

$$\underline{\mathbf{A}}_k(t) \equiv \mathbf{P}_0^{-1}(t) \mathbf{A}_k(t) \mathbf{P}_0(t). \quad (\text{A.17})$$

Now we may express the Floquet solution perturbatively, with  $\mathbf{P}_0^{-1}(t) \mathbf{P}(t)$  given by a Magnus series [109], as

$$\underline{\mathbf{G}}(t) = e^{\underline{\Phi}(t)} e^{t\mathbf{F}}, \quad (\text{A.18})$$

$$\underline{\Phi}(t) = \sum_{k=1} \underline{\Phi}_k(t), \quad (\text{A.19})$$

$$\mathbf{F} = \sum_{k=0} \mathbf{F}_k. \quad (\text{A.20})$$

The two unknown series will be solved, order-by-order, with two constraints: (1) the dynamical constraint of the equations of motion and (2) the periodicity constraint of the Floquet normal form. Inserting these expressions into the equation of motion (A.16) yields the expression

$$\left[ \frac{d}{dt} e^{+\Phi(t)} \right] e^{-\Phi(t)} + e^{+\Phi(t)} \mathbf{F} e^{-\Phi(t)} = \mathbf{F}_0 + \sum_{k=1} \underline{\mathbf{A}}_k(t). \quad (\text{A.21})$$

To inspect this equation perturbatively we expand the exponential operators with Taylor series and regroup terms order-by-order

$$e^{+\Phi(t)} = \mathbf{1} + \Phi(t) + \frac{1}{2} \Phi^2(t) + \mathcal{O}(g^3), \quad (\text{A.22})$$

$$= \mathbf{1} + \Phi_1(t) + \left( \Phi_2(t) + \frac{1}{2} \Phi_1^2(t) \right) + \mathcal{O}(g^3). \quad (\text{A.23})$$

The zeroth-order constraint is trivially satisfied.

$$\mathbf{0} + \mathbf{F}_0 = \mathbf{F}_0 + \mathbf{0}. \quad (\text{A.24})$$

The first-order constraint yields the dynamical relation

$$\dot{\Phi}_1(t) = \text{Ad}[\mathbf{F}_0] \Phi_1(t) + \underline{\mathbf{A}}_1(t) - \mathbf{F}_1, \quad (\text{A.25})$$

in terms of the commutator super-operator

$$\text{Ad}[\mathbf{A}] \mathbf{B} = \mathbf{A} \mathbf{B} - \mathbf{B} \mathbf{A}. \quad (\text{A.26})$$

Higher-order constraints will yield analogous recursive dynamics of the form

$$\dot{\Phi}_k(t) = \text{Ad}[\mathbf{F}_0] \Phi_k(t) + \mathbf{C}_k(t) - \mathbf{F}_k, \quad (\text{A.27})$$

where  $\mathbf{C}_k(t)$  depends upon  $\underline{\mathbf{A}}_k(t)$  and products of lower-order terms. This equation of motion has two unknowns  $\Phi_k(t)$  and  $\mathbf{F}_k$ . The dynamical equation is periodic,

but the solution will only be periodic if it additionally has matching boundary conditions at the beginning and end of one period. By design,  $\Phi(0) = \mathbf{0}$  to all orders and therefore we can fix  $\Phi(t)$  to have the correct periodicity by choosing  $\mathbf{F}_k$  such that  $\Phi_k(\mathcal{T}) = \Phi_k(0) = \mathbf{0}$ .

For now we will ignore the fact that the commutator super-operator is inherently degenerate and proceed with naive solutions. These naive solutions will be shown to be more generally correct when interpreted properly. Given that  $\Phi(0)$  must vanish to all orders, solutions then take the form

$$\Phi_k(t) = \int_0^t d\tau e^{+(t-\tau)\text{Ad}[\mathbf{F}_0]} \{ \mathbf{C}_k(\tau) - \mathbf{F}_k \}, \quad (\text{A.28})$$

in terms of the exponential super-operator

$$e^{+t\text{Ad}[\mathbf{F}]} \Phi = e^{+t\mathbf{F}} \Phi e^{-t\mathbf{F}}. \quad (\text{A.29})$$

Next we find a simple constraint for the unknown  $\mathbf{F}_k$  by invoking the periodicity of  $\Phi(t)$ , specifically we will use  $\Phi(\mathcal{T}) = \Phi(0) = \mathbf{0}$ . One can then show that

$$\mathbf{F}_k = \frac{\text{Ad}[\mathbf{F}_0]}{\mathbf{1} - e^{-\mathcal{T}\text{Ad}[\mathbf{F}_0]}} \int_0^{\mathcal{T}} d\tau e^{-\tau\text{Ad}[\mathbf{F}_0]} \mathbf{C}_k(\tau). \quad (\text{A.30})$$

Given that  $\mathbf{C}_k(t)$  has period  $\mathcal{T}$ , Eq. (A.30) takes the form of the operator-Laplace transform

$$\mathbf{F}_k = \text{Ad}[\mathbf{F}_0] \hat{\mathbf{C}}_k(\text{Ad}[\mathbf{F}_0]), \quad (\text{A.31})$$

if we interpret the operator-Laplace transform as operating from the left and with the limit  $\text{Ad}[\mathbf{F}_0] \rightarrow 0$  taken when acting upon the null space of the operator. Eq. (A.28) can also be identified as a operator-Laplace convolution.

As  $\mathbf{C}(t)$  has period  $\mathcal{T} = \frac{2\pi}{\Omega}$  it should be well represented by the Fourier series

$$\mathbf{C}_k(t) = \sum_n e^{+in\Omega t} \mathbf{c}_{k;n}, \quad (\text{A.32})$$

which results in the solutions

$$\mathbf{F}_k = \sum_n \frac{\text{Ad}[\mathbf{F}_0]}{\text{Ad}[\mathbf{F}_0] - in\Omega} \mathbf{c}_{k;n}, \quad (\text{A.33})$$

$$\Phi_k(t) = \sum_n \frac{1 - e^{+in\Omega t}}{\text{Ad}[\mathbf{F}_0] - in\Omega} \mathbf{c}_{k;n}. \quad (\text{A.34})$$

And in general, given the any functional decomposition of the form

$$\mathbf{C}_k(t) = \sum_n c_n(t) \mathbf{c}_{k;n}, \quad (\text{A.35})$$

we can express the solutions

$$\mathbf{F}_k = \sum_n \text{Ad}[\mathbf{F}_0] \hat{c}_n(\text{Ad}[\mathbf{F}_0]) \mathbf{c}_{k;n}, \quad (\text{A.36})$$

$$\hat{\Phi}_k(s) = \frac{1}{s} \sum_n \frac{s \hat{c}_n(s) - \text{Ad}[\mathbf{F}_0] \hat{c}_n(\text{Ad}[\mathbf{F}_0])}{s - \text{Ad}[\mathbf{F}_0]} \mathbf{c}_{k;n}, \quad (\text{A.37})$$

where  $\hat{f}(s) = \int_0^\infty dt e^{-st} f(t)$ .

### A.3.0.1 Degenerate Dynamics

Even assuming  $\mathbf{F}_0$  has no degeneracy, the operation  $\text{Ad}[\mathbf{F}_0] \mathbf{O} = [\mathbf{F}_0, \mathbf{O}]$  is still degenerate, specifically for the diagonal entries in the eigen-basis of  $\mathbf{F}_0$ . Therefore these differential equations naturally decouple into the commutative and non-commutative terms. Let us denote this projective decomposition  $\mathbf{Z} = \mathbf{Z}^\parallel + \mathbf{Z}^\perp$  where  $\mathbf{Z}^\parallel$  lies in the null space of  $\text{Ad}[\mathbf{F}_0]$  and  $\mathbf{Z}^\perp$  constitutes the remainder. The decoupled

dynamical relations are then

$$\dot{\Phi}_k^\perp(t) = \text{Ad}[\mathbf{F}_0] \Phi_k^\perp(t) + \mathbf{C}_k^\perp(t) - \mathbf{F}_k^\perp, \quad (\text{A.38})$$

$$\dot{\Phi}_k^\parallel(t) = \mathbf{C}_k^\parallel(t) - \mathbf{F}_k^\parallel. \quad (\text{A.39})$$

The corresponding commuting solutions are much simpler. In this case one recovers the results of Ref. [33] as they effectively considered  $\mathbf{F}_0 = 0$ . One can also consider this to encompass a rigorous derivation of the rotating-wave approximation. The solutions  $\mathbf{F}^\parallel$  and  $\Phi^\parallel(t)$  can be most easily expressed in terms of the time average and variance of  $\mathbf{C}^\parallel(t)$ .

$$\mathbf{F}_k^\parallel = \langle \mathbf{C}_k^\parallel \rangle, \quad (\text{A.40})$$

$$\Phi_k^\parallel(t) = \int_0^t d\tau \Delta \mathbf{C}_k^\parallel(t). \quad (\text{A.41})$$

Note that if  $\mathbf{F}_0$  itself is degenerate, and not simply  $\text{Ad}[\mathbf{F}_0]$ , then that portion of this degenerate evolution is not perturbative in the sense of  $g \ll \|\mathbf{F}_0\|$  but  $g \ll \mathcal{T}^{-1}$ .

## A.4 Asymptotically-Homogeneous and Commutative Dynamics

Now suppose we have a linear system of parametric differential equations of the form

$$\dot{\mathbf{X}}(t) = \mathbf{A}(t) \mathbf{X}(t), \quad (\text{A.42})$$

such that the time-translation generator  $\mathbf{A}(t)$  commutes with itself at late time and for vanishing perturbation. The specific example we have in mind is that  $\mathbf{A}(t)$  is asymptotically constant in time. One might consider a Hamiltonian system which

is perturbed for a finite time or an open system with dynamics which relax to stationary values.

Next assume we have a perturbative expansion of our time-translation generator

$$\mathbf{A}(t) = \sum_{k=0} \mathbf{A}_k(t), \quad (\text{A.43})$$

where  $\mathbf{A}_k(t) = \mathcal{O}(g^k)$  in some perturbative parameter  $g$ , and  $\mathbf{A}_0(t)$  commutes with itself for all times. Again, exact solutions can be expressed as in Eq. (A.11), but we also know that we have commutative propagation at late time and also for vanishing perturbation. E.g.

$$\lim_{t > \tau \gg 0} \mathbf{G}(t, \tau) = e^{\int_{\tau}^t d\tau \mathbf{A}(\tau)}, \quad (\text{A.44})$$

$$\mathbf{G}_0(t, 0) = e^{\int_0^t d\tau \mathbf{A}_0(\tau)}, \quad (\text{A.45})$$

where the two-time propagator is defined such that  $\mathbf{X}(t) = \mathbf{G}(t, \tau) \mathbf{X}(\tau)$ . Therefore we look for an initial-time propagator,  $\mathbf{G}(t) = \mathbf{G}(t, 0)$ , of the form

$$\mathbf{G}(t) = e^{+\Phi(t)} \mathbf{G}'(t), \quad (\text{A.46})$$

$$\Phi(t) \equiv \int_0^t d\tau \mathbf{A}(\tau). \quad (\text{A.47})$$

*Fer expansion* [56] provides a perturbation theory well-tailored to the above problem.

We start by inspecting the iterated equation of motion

$$\dot{\mathbf{G}}'(t) = \mathbf{A}'(t) \mathbf{G}'(t), \quad (\text{A.48})$$

$$\mathbf{A}'(t) \equiv e^{-\Phi(t)} \mathbf{A}(t) e^{+\Phi(t)} - \int_0^1 d\eta e^{-\eta \Phi(t)} \mathbf{A}(t) e^{+\eta \Phi(t)}, \quad (\text{A.49})$$

and we note that these dynamics must be at minimum  $\mathcal{O}(g)$  in addition to vanishing in the late-time regime. Asymptotically vanishing is a special case of asymptotic commutativity and therefore if our first iteration was valid then we can solve this new equation by iterating again.

$$\mathbf{G}'(t) = e^{+\Phi'(t)} \mathbf{G}''(t), \quad (\text{A.50})$$

$$\Phi'(t) \equiv \int_0^t d\tau \mathbf{A}'(\tau), \quad (\text{A.51})$$

and in general

$$\mathbf{G}^{[n]}(t) = e^{+\Phi^{[n]}(t)} \mathbf{G}^{[n+1]}(t), \quad (\text{A.52})$$

$$\Phi^{[n]}(t) \equiv \int_0^t d\tau \mathbf{A}^{[n]}(\tau), \quad (\text{A.53})$$

$$\mathbf{A}^{[n+1]}(t) \equiv e^{-\Phi^{[n]}(t)} \mathbf{A}^{[n]}(t) e^{+\Phi^{[n]}(t)} - \int_0^1 d\eta e^{-\eta \Phi^{[n]}(t)} \mathbf{A}^{[n]}(t) e^{+\eta \Phi^{[n]}(t)}. \quad (\text{A.54})$$

The Fer expansion is then expressed in its entirety

$$\mathbf{G}(t) = e^{+\Phi(t)} e^{+\Phi'(t)} e^{+\Phi''(t)} \dots \quad (\text{A.55})$$

All of the late-time and  $\mathcal{O}(g^0)$  dynamics are contained within the first factor, while successive factors contain both short-time and higher-order dynamics. To prove that  $\Phi^{[n]}(t)$  occur in increasing orders of  $g$ , we first note that the iterated time-translation generator can be expressed

$$\mathbf{A}'(t) = \left\{ e^{-\text{Ad}[\Phi(t)]} - \int_0^1 d\eta e^{-\eta \text{Ad}[\Phi(t)]} \right\} \mathbf{A}(t), \quad (\text{A.56})$$

in terms of the commutator super-operation,  $\text{Ad}[\mathbf{A}] \mathbf{B} = \mathbf{A} \mathbf{B} - \mathbf{B} \mathbf{A}$ . The integral can then be evaluated as

$$\mathbf{A}'(t) = \frac{1}{\text{Ad}[\Phi(t)]} \left\{ (\mathbf{1} + \text{Ad}[\Phi(t)]) e^{-\text{Ad}[\Phi(t)]} - \mathbf{1} \right\} \mathbf{A}(t), \quad (\text{A.57})$$

and finally we can see that upon series expansion the commutator-dependent pre-factor is at minimum  $\mathcal{O}(\text{Ad}[\Phi])$ . Because of perturbative commutativity in the time-translation generator,  $\mathbf{A}'(t) = \mathcal{O}(g)$  despite the fact that  $\mathbf{A}(t)$  and  $\Phi(t)$  are both  $\mathcal{O}(g^0)$ .  $\Phi'(t)$  is therefore  $\mathcal{O}(g)$  and given the above relation we have  $\mathbf{A}''(t) = \mathcal{O}(g^2)$ ,  $\mathbf{A}'''(t) = \mathcal{O}(g^4)$ , ... By induction one can see that for  $n > 0$ ,  $\mathbf{A}^{[n]}(t)$  and  $\Phi^{[n]}(t)$  are both of the *tetration* or iterated exponential order  $(n-1)2$  in  $g$ . Tetration is defined as

$${}^n q = \left. q^{q^{\cdot^{\cdot^q}}} \right\} n, \quad (\text{A.58})$$

and is a very rapid pace of convergence: much faster than if we had relied upon the Magnus series of  $\mathbf{G}'(t)$ , but equivalent at first order in  $g$ .

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