Quantum Dynamics with Trapped Ions

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Abstract

Trapped atomic ions are one of the leading platforms for quantum simulations. Recently, they have also established themselves as a testbed for classical statistical mechanics and mesoscopic systems. The challenges in this field are twofold at the moment.

In the endeavor to simulate systems which are beyond the simulation capabilities of currently available conventional computers, trapped-ion experiments have constantly become more complex. The precise interrogation of the dynamics of the ever more complex experiments remains a challenge. It is therefore desirable to develop new schemes for the study of the dynamics of crystals of trapped ions which can be realized with state-of-the-art technology. On the other hand, there is a demand for simulation protocols that can be implemented in current experiments and, at the same time, are able to simulate physics which cannot be simulated efficiently on conventional computers. In this thesis we address both of these issues.

In the first part of the thesis, we show how the concepts of multidimensional spectroscopy can be extended to the interrogation of nonlinear dynamics of crystals of trapped ions. The scheme we present can be realized with currently available technology and reveals the nonlinear dynamics of ion crystals. It can be applied to systems with many ions as well as in the presence of thermal excitations. A further asset of the protocol we present is that different types of noise can be distinguished in the spectra.

We illustrate the feasibility of our proposal by simulating two trapped-ion experiments which can be realized in state-of-the-art setups. In particular, we show that two-dimensional spectroscopy can be used to detect signatures of a structural phase transition of an ion crystal and how it can be used to detect resonant energy exchange between normal modes. By including different noise mechanisms into the simulations, we illustrate how two-dimensional spectroscopy allows us to distinguish these different types of noise.

In the second part of this thesis we present a protocol for the simulation of open quantum system dynamics with trapped ions. Concretely, we show how the dynamics of spin-boson models can be simulated with small crystals of trapped ions. In the scheme we develop, the spin is encoded in the electronic degrees of freedom of one ion whereas the collective vibrational degrees of freedom of the crystal are used to form a tunable effective harmonic environment. Our proposal utilizes the result that a damped mode yields an effective Lorentzian spectral density. Combining several damped modes more complex spectral densities can be tailored and theoretically arbitrary spectral densities can be constructed. To avoid the experimentally demanding single site addressing, we propose to work with mixed species crystals. Then, the electronic degrees of freedom of one species can be used to encode the spin while the ions of the other species can be used to cool the vibrational degrees of freedom of the crystal and thus to engineer the environment. The strength of the dissipation can be tuned by adjusting the coupling between the spin and the cooled vibrational degrees of freedom and thus is under full control of the experimenter.

We illustrate the protocol and its flexibility by simulating an experiment which can be implemented in currently available setups. We also show that the proposed quantum simulations yield truly non-Markovian dynamics by computing two recently proposed quantitative measures of non-Markovianity. Implementations of this simulation protocol beyond this proof-of-principle experiment have the potential to enter parameter regimes which are intractable on conventional computers.
List of publications

Parts of this thesis are based on material first published in the following articles

   *Two-Dimensional Spectroscopy for the Study of Ion Coulomb Crystals*,
   See Chapter 3.

   *Simulating spin-boson models with trapped ions*,
   See Chapter 4.

The material of the following articles is not covered by this thesis.

[L3] A. Lemmer, A. Bermudez, and M. B. Plenio,
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[L4] A. Lemmer, A. Bermudez, and M. B. Plenio,
   *Driven geometric phase gates with trapped ions*,
# Contents

1. Introduction 1

2. Trapped atomic ions in radio-frequency traps 3
   2.1. Trapping ions ................................................. 3
       2.1.1. Radio-frequency traps ................................ 4
       2.1.2. Motion of trapped ions in radio-frequency traps .... 6
       2.1.3. Quantized motion of trapped ions in radio-frequency traps ...... 11
       2.1.4. Mixed species crystals .................................. 12
   2.2. Light-matter interaction ................................... 14
       2.2.1. Electric dipole Hamiltonian ............................ 15
       2.2.2. Atomic transitions and the rotating wave approximation .... 18
       2.2.3. The two-level approximation ............................ 19
       2.2.4. Laser-ion interaction in a trap .......................... 20
       2.2.5. Laser-ion interaction for a collection of ions .......... 24
   2.3. Trapped-ion qubits ........................................... 25
       2.3.1. Qubit encodings with trapped ions ...................... 26
       2.3.2. Qubit coherence times ................................... 27
   2.4. Master equations, the quantum regression theorem and adiabatic elimination 28
       2.4.1. Standard form of the Markovian master equations ....... 29
       2.4.2. Master equation in the Born-Markov approximation ...... 32
       2.4.3. The quantum regression theorem .......................... 35
       2.4.4. Time-local master equations with memory effects ....... 38
       2.4.5. Adiabatic elimination .................................... 39
   2.5. Stimulated Raman transitions and spin-dependent optical dipole forces ........ 41
       2.5.1. Two-photon stimulated Raman transitions ............... 45
       2.5.2. Optical dipole forces ..................................... 46
   2.6. Laser cooling of trapped ions ............................... 49
       2.6.1. Doppler cooling ........................................... 50
       2.6.2. EIT cooling of a single trapped ion ..................... 53
       2.6.3. Sympathetic cooling of mixed species crystals of trapped ions .... 61

3. Two-dimensional spectroscopy for the study of Coulomb crystals 65
   3.1. Basic principles of Two-Dimensional Spectroscopy ............. 66
       3.1.1. Correlation spectroscopy ................................ 68
       3.1.2. Separation of interactions ............................... 71
       3.1.3. Homogeneous and inhomogeneous broadening ............... 73
   3.2. The linear-to-zigzag structural transition .................... 74
   3.3. Nonlinear interactions in crystals of trapped ions ........... 76
       3.3.1. Third-order expansion of the Coulomb potential ........ 77
       3.3.2. Resonant interactions in the third-order Hamiltonian .... 78
       3.3.3. Fourth-order terms in the Coulomb potential ............ 80
       3.3.4. Resonant interactions in the fourth-order Hamiltonian .... 82
   3.4. Population measurement for motional modes of trapped ion crystals ........ 87
3.5. Two-dimensional spectroscopy with trapped ions – protocol and theoretical analysis
3.5.1. 2D spectroscopy protocol for the detection of nonlinear dynamics with trapped ions
3.5.2. Phase cycling and coherence transfer pathways
3.5.3. Proof of vanishing harmonic signal
3.6. Two-dimensional spectroscopy with trapped ions – simulations of experimental implementations
3.6.1. Signatures of the onset of a structural transition from 2D spectroscopy
3.6.2. Resonant energy transfer between normal modes due to third-order effects of the Coulomb potential investigated by 2D spectroscopy
3.6.3. Impact of laser phase fluctuations on phase cycling
3.7. Conclusions and outlook

4. Simulations of the dynamics of spin-boson models with trapped ions
4.1. Spin-boson model and spectral densities
4.1.1. Spin-boson model
4.1.2. Spectral densities
4.2. Path integrals in quantum mechanics
4.2.1. The quantum mechanical propagator as a path integral
4.2.2. Two-time correlation functions with path integrals
4.3. Path integrals for open quantum systems – Influence functionals
4.4. Influence functionals for harmonic oscillator environments
4.4.1. Influence functionals and spectral density for oscillator environments
4.4.2. The spectral density, two-time correlation functions and the fluctuation-dissipation theorem
4.5. Effective spectral densities of damped harmonic oscillators
4.5.1. Two-time correlation functions of damped oscillators
4.5.2. Effective spectral density of Lindblad and Ohmic damped oscillator
4.6. Experimental simulations of the spin-boson model using trapped ions
4.7. Impact of experimental sources of noise
4.7.1. Dephasing noise
4.7.2. Decoherence due to \( \sigma^z \) spin-motion coupling
4.8. Quantification of the degree of non-Markovianity of the spin-boson dynamics
4.8.1. The RHP measure of non-Markovianity
4.8.2. The BLP measure of non-Markovianity
4.9. Summary and Outlook

5. Summary and outlook

A. Derivation of EIT cooling master equation
B. Derivation of the absorption spectrum of the \( \Lambda \) system in the EIT cooling configuration
C. Linear chain mappings and the spin-boson model
D. Numerical evaluation of the measure of non-Markovianity \( \mathcal{M}_{RHP} \)
1. Introduction

A quantum computer is a device that uses quantum-mechanical phenomena to solve a computational task. Quantum computers and their possible physical implementations have received a lot of attention, because they promise to efficiently solve certain computational tasks which are believed to be intractable on conventional computers [1].

In order to realize a quantum computer, one needs a so-called universal set of gates which allows one to perform every possible computation. Interestingly, single- and two-particle gates, i.e. operations, suffice to obtain such a universal set. It is commonly agreed that these gates have to be performed with errors of less than $10^{-4}$ per operation, in order to be able to obtain a scalable implementation of a quantum computer [2, 3].

Trapped atomic ions were among the first experimental platforms to be considered for the implementation of a quantum computer after the seminal proposal by Cirac and Zoller [4], who showed that the two-particle interactions needed for a quantum computer can be realized with trapped ions. Universal sets of gates operating with error rates close to the aforementioned threshold have been demonstrated recently with trapped ions [5, 6], and even first small scale implementations of programmable quantum computers involving five quantum bits have been reported [7, 8].

Several other experimental platforms, such as superconductors, photons or quantum dots, have also been considered for the implementation of a quantum computer, and these have seen remarkable progress as well [2, 9]. However, despite significant experimental and theoretical advances the implementation of a scalable quantum computer, which outperforms current off-the-shelf computers, remains a distant goal for all platforms.

Remarkably, in order to solve problems which are intractable with classical computers, it is not necessary to build a universal quantum computer. Special-purpose devices called quantum simulators can achieve this goal and their realization is feasible with available technology. So what is a quantum simulator?

When it comes to the simulation of quantum systems, classical computers are already challenged for moderately sized problems. The direct simulation of quantum systems is often not possible due to the exponential growth of required computational resources in the system size. An idea to overcome this problem, first suggested by Feynman [10], is to use one quantum system which we can control and measure to simulate another quantum system. The advantage is obvious: The simulator is itself a quantum system and due to its quantum nature naturally provides the complexity to simulate another quantum system.

Two types of quantum simulators are distinguished [11]: digital and analog quantum simulators. In a digital quantum simulator, the state of the system to be simulated is encoded in quantum bits, the basic unit of information in quantum information processing, and the time evolution is decomposed into a series of gates. Hence, a digital quantum simulator is not much different from a quantum computer and in essence equally hard to build. An analog quantum simulator is easier to implement. This type of simulator comprises a quantum system whose Hamiltonian is tunable to a certain extent. Furthermore, one should be able to control and measure this quantum system very precisely. For a quantum simulation, one prepares the simulator in the desired initial state and tunes its Hamiltonian to that of the system to be simulated. After evolution under the desired Hamiltonian, the relevant information is retrieved from the simulator by suitable measurements.
Quantum simulation is nowadays a thriving field of research and trapped ions are among the leading platforms of the field [12]. Trapped ions have been proposed as quantum simulators for models from diverse areas of physics. The proposals include simulators of spin systems [13], models of friction [14–16], heat transport [17], and even lattice gauge theories [18]. Experimentally impressive results have been achieved: Starting from a proof-of-principle simulation of a two-spin Ising model [19], spin systems involving around ten ions [20–22] and recently up to 53 ions have been simulated [23]. Intriguing phenomena such as many body localization [24] or discrete time crystals [25] have been observed and also simulations of a lattice gauge theory have been implemented [26]. Moreover, trapped ions have also become a testbed for the simulation of classical statistical mechanics involving, for example, the observation of the Kibble-Zurek mechanism in a structural phase transition [27–29].

The ever more complex experiments demand the development of new techniques for the interrogation of the dynamics of ion crystals that facilitate to extract the relevant information efficiently. The first part of the research presented in this thesis is devoted to addressing this challenge. We consider how two-dimensional spectroscopy, which has been extremely successful in the study of complex spin systems [30] and the electronic dynamics of molecules [31], can be applied to trapped ions. We focus on the investigation of the motional dynamics of ion crystals and devise a two-dimensional spectroscopy protocol that detects nonlinear dynamics in crystals of trapped ions. Nonlinear dynamics play an important role in different realms of ion trap experiments including structural phase transitions [32] as well as some proposals for quantum simulation, see e.g. [14–16, 33].

The majority of the quantum simulation proposals and experiments focuses on closed quantum systems. The simulation of open quantum system dynamics has received a lot less attention with only a few proposals and experiments, e.g. [34, 35]. This is somewhat surprising for the simulation of open quantum systems is of high practical relevance: Virtually all quantum systems are open quantum systems. On the other hand, the simulation of open systems faces the difficulty that a tunable environment, which usually is considered to have an infinite number of degrees of freedom, has to be engineered.

The second part of the research presented in this thesis addresses this issue. We present a protocol for the simulation of open quantum systems with trapped ions. We show how the spin-boson model, an archetypical model of an open quantum system, can be simulated with crystals of trapped ions and infinite environments can be simulated using only a few ions. Our protocol is flexible and the effective environment seen by the spin can be tailored. The protocol can be extended to include more, possibly interacting, spins. This opens interesting future perspectives for quantum simulations with trapped ions. Trapped ions could then be used to simulate the physics of photosynthetic pigment protein complexes and contribute to the understanding of the physical mechanisms participating in photosynthesis.

The thesis is structured as follows. In Chapter 2, we present basic theoretical results for the description of atomic ions in radio-frequency traps. We also discuss some elementary results from the theory of open quantum systems and their application to the theoretical description of atomic ions in radio-frequency traps. In Chapter 3, we develop our protocol of two-dimensional spectroscopy with trapped ions and illustrate it by simulating two experiments. Then, in Chapter 4, we shift our focus to the simulation of open quantum systems using trapped ions. Starting from a path integral approach, we devise a procedure for the simulation of spin-boson models with trapped ions which is flexible and facilitates the simulation of the dynamics of complex quantum systems. We illustrate also this proposal with the simulation of an experiment. In Chapter 5 we summarize our results and present some future prospects for the techniques developed in this work.
2. Trapped atomic ions in radio-frequency traps

This work is focused on spectroscopy and the quantum simulation of the dynamics of open quantum systems in the context of trapped atomic ions. The aim of this thesis is to design protocols for spectroscopy and quantum simulations with crystals of trapped atomic ions, which can be implemented with currently available technology. On these grounds, it is mandatory we familiarize ourselves with the basic concepts of ion trapping and the tools available for the manipulation of ions once they are trapped.

In this chapter we review the mechanism of radio-frequency traps and the description of the interaction of trapped atomic ions in radio-frequency traps with additionally applied electromagnetic fields. For the description of these interactions we will need some theoretical tools and results which will be presented on the way. This also includes some results from the theory of open quantum systems. We close the chapter with a brief account of the theory of laser cooling.

Let us finally remark that in this work we will only consider trapped atomic ions. Experiments with molecular ions are also carried out, see e.g. [36, 37] for recent reviews, but we shall not be concerned with these here. Furthermore, we only consider ions with a single positive charge. For brevity we will use the term “trapped ions” from now on and it is understood that we refer to trapped positively-charged atomic ions.

This chapter is organized as follows. We start with the theoretical description of the classical motion of a single ion and collections of ions in a radio-frequency trap. Thereafter, we quantize the motion and briefly consider the motion of mixed-species crystals. We then discuss the interaction of the internal degrees of freedom of trapped ions with additionally applied electromagnetic fields, as for example laser of microwave radiation. In this context, we introduce the rotating wave approximation and discuss how electronic and motional degrees of trapped ions can be coupled. After discussing possible implementations of two-level systems with trapped ions in some length, we take a little detour to the theory of open quantum systems. We discuss master equations with a particular emphasis on the weak coupling master equation. In this context we also discuss the quantum regression theorem and adiabatic elimination. In the final part of the chapter we use these tools to describe the interaction of trapped ions with lasers in a stimulated Raman configuration and laser cooling.

2.1. Trapping ions

Due to their electric charge ions can be trapped by a combination of electromagnetic fields. The two most common architectures for trapping ions are the so-called Paul or radio-frequency traps [38, 39] and Penning traps [40, 41]. Paul traps, in most cases, use a combination of static and dynamical electric fields to confine the ions while Penning traps rely on static electric and magnetic fields.

Both types of traps are widely used and have their advantages and disadvantages. This work focuses on trapped atomic ions in radio-frequency traps which use a combination of static and dynamic electric fields to trap the ions because the majority of the quantum simulation experiments is conducted in such traps [12]. The necessity to use time-dependent fields in this type
2. Trapped atomic ions in radio-frequency traps

of trap can be understood by considering a result from classical electrodynamics often referred to as Earnshaw’s theorem. The theorem follows from the Laplace equation of the electrostatic potential \( \Phi(r) \) in free space

\[
\Delta \Phi(r) = 0, \tag{2.1}
\]

where \( \Delta \) is the Laplace operator. In order to stably trap a charged particle with static electric fields we would need a minimum or maximum of \( \Phi(r) \) depending on the charge of the particle. However, Earnshaw’s theorem tells us that there can only be saddle points of the electrostatic potential in free space. Accordingly, one cannot stably trap a charged particle by static electric fields alone. One way to circumvent this problem is to resort to time-dependent potentials instead.

2.1.1. Radio-frequency traps

We will discuss the mode of operation of a linear Paul trap in this section. Figure 2.1 shows a schematic picture of a linear Paul trap. It consists of four segmented rods arranged in a quadrupolar configuration. All of the rods have the same distance to the \( z \)-axis of the coordinate system in Fig. 2.1. This axis is also called the trap axis. Throughout this thesis we choose the \( z \)-axis such that it coincides with the trap axis. It is then customary to refer to the \( z \)-direction as the axial direction and to the motion along the trap axis as axial motion. \( x, y \) are then called the radial directions.

Applying a potential of the form

\[
V = U_r + V_0 \cos(\Omega_{rf} t) \tag{2.2}
\]

between diagonally opposite rods one approximately obtains a quadrupolar potential near the \( z \)-axis [39, 42]

\[
\Phi_{rf} = \frac{1}{2} [U_r + V_0 \cos(\Omega_{rf} t)] \left[ 1 + \frac{x^2 - y^2}{R^2} \right]. \tag{2.3}
\]

Here \( R \) is the distance of the \( z \)-axis to the surface of the electrodes. The drive frequency \( \Omega_{rf} \) typically lies in the radio-frequency (rf) regime, which accounts for the name “radio-frequency traps”. If the rods do not form equipotentials of \( \Phi_{rf} \), the right-hand side of Eq. (2.3) has to be multiplied by a geometric factor of order one [43].

The potential in Eq. (2.3) only provides confinement of ions in the \( x \) - and \( y \) -directions but not along \( z \). In order to obtain three-dimensional confinement a dc voltage \( U_0 \) is applied to the outer segments of the rods, as indicated in Fig. 2.1, generating a static effective potential of the form [39]

\[
\Phi_s = \kappa U_0 \left[ z^2 - \frac{1}{2} (x^2 + y^2) \right] \tag{2.4}
\]

near the center of the trap. Here \( \kappa \) is a geometric factor. Accordingly, the full potential near the center of the trap reads

\[
\Phi_{tr} = \Phi_{rf} + \Phi_s = \frac{1}{2} [U_r + V_0 \cos(\Omega_{rf} t)] \left[ 1 + \frac{x^2 - y^2}{R^2} \right] + \kappa U_0 \left[ z^2 - \frac{1}{2} (x^2 + y^2) \right]. \tag{2.5}
\]

The classical equations of motion for an ion of mass \( m \) and charge \( q \) in the potential \( \Phi_{tr} \) are found by employing Newton’s second law

\[
\ddot{r} = -\frac{q}{m} \nabla \Phi_{tr}. \tag{2.6}
\]
2.1. Trapping ions

Figure 2.1.: The figure shows a schematic of a Paul trap derived from [42]. An rf potential of the form in Eq. (2.2) is applied to the gray rods while the remaining rods are held at rf ground [42]. In order to provide confinement for the positively charged ions along the $z$-axis the other two rods are segmented. A potential that is positive with respect to the inner segments (blue) is applied to the outer (red) segments. The green spheres in the middle indicate trapped ions.

The resulting equations of motion are decoupled in the spatial coordinates. In the $z$-direction we obtain

$$\ddot{z} + \frac{2q\kappa U_0}{m}z = 0.$$  \hspace{1cm} (2.7)

Accordingly, the ion performs harmonic oscillations of frequency

$$\omega_z = \sqrt{\frac{2q\kappa U_0}{m}}.$$  \hspace{1cm} (2.8)

in the $z$-direction. Using Eq. (2.8) we can write the potential $\Phi_s$ in Eq. (2.4) as

$$\Phi_s = \frac{m}{2q} \omega_z^2 \left[ z^2 - \frac{1}{2} (x^2 + y^2) \right].$$  \hspace{1cm} (2.9)

The equations of motion in the $x$- and $y$-directions are Mathieu differential equations. After some algebra, they can be cast into the standard form of the Mathieu differential equation

$$\frac{d^2x}{d\zeta^2} + \left[ a_x + 2q_x \cos(2\zeta) \right]x = 0,$$  \hspace{1cm} (2.10)

$$\frac{d^2y}{d\zeta^2} + \left[ a_y + 2q_y \cos(2\zeta) \right]y = 0,$$  \hspace{1cm} (2.11)

where we have introduced the abbreviations

$$\zeta = \frac{\Omega_{\text{rf}} t}{2},$$

$$a_x = \frac{4q}{m\Omega_{\text{rf}}^2} \left( \frac{U_t}{R^2} - \kappa U_0 \right), \quad a_y = -\frac{4q}{m\Omega_{\text{rf}}^2} \left( \frac{U_t}{R^2} + \kappa U_0 \right),$$  \hspace{1cm} (2.12)

$$q_x = -q_y = \frac{2qV_0}{\Omega_{\text{rf}}^2 mR^2}.$$  \hspace{1cm}

The Mathieu differential equation can be solved using Floquet theory [39,42]. The stability of the solutions depends on the values of $a_\alpha$ and $q_\alpha$, $\alpha = x, y$ and the sets $\{a_\alpha, q_\alpha\}$ that yield stable solutions are called stability regions. Typically, traps are operated in the regime $|a_\alpha|, q_\alpha^2 \ll 1$ in the so-called lowest stability region that contains the point $(a_\alpha, q_\alpha) = (0, 0)$ [39,42]. For $|a_\alpha|, q_\alpha^2 \ll 1$, the ion trajectory is approximately given by

$$r_\alpha(t) = A_\alpha \cos(\omega_\alpha t) \left[ 1 - \frac{q_\alpha}{2} \cos(\Omega_{\text{rf}} t) \right],$$  \hspace{1cm} (2.13)
where the amplitude \( A_\alpha \) depends on the initial conditions and

\[
\omega_\alpha = \beta_\alpha \Omega_{rf}/2 \quad \text{with} \quad \beta_\alpha \approx \sqrt{a_\alpha + q_\alpha^2/2}.
\]  

Hence, the ion trajectories in the \( x \)- and \( y \)-directions consist of harmonic oscillations of frequency \( \omega_\alpha \ll \Omega_{rf} \) superposed with oscillations at the drive frequency \( \Omega_{rf} \). The oscillations of frequency \( \omega_\alpha \) are called secular motion. The superposed oscillations at drive frequency \( \Omega_{rf} \) have an amplitude smaller by a factor \( q_\alpha/2 \) and are therefore called micromotion. In most theoretical treatments of trapped ions, micromotion is neglected and we will do so in the following. This is justified in most experimental situations if the kinetic energy of the ions is small enough [42].

If we disregard micromotion, the ion behaves as if it were confined in an effective harmonic potential \( \Phi_r \) in the radial directions

\[
\Phi_r = \frac{m}{2q} (\omega_x^2 x^2 + \omega_y^2 y^2),
\]  

where \( \omega_x, y \) are given in Eq. (2.14) with the parameters in Eq. (2.12). Note that \( \omega_x \) and \( \omega_y \), in general, are different but can be made equal if we choose \( U_t = 0 \). Then, we obtain \( \omega_x = \omega_y = \omega_r \).

If additionally \( a_\alpha \ll q_\alpha \), we can write the degenerate radial frequency \( \omega_r \) as

\[
\omega_r \approx \frac{|q_\alpha|}{2\sqrt{2}} \Omega_{rf} = \frac{qV_0}{\sqrt{2}\Omega_{rf}mR^2}.
\]  

We see that the radial potential is dependent on the ion’s mass. This is important if one considers several ions of different masses confined in the same trap.

In summary, for appropriate choices of the trap parameters, the full potential \( \Phi_{rf} \) of a linear Paul trap can be described by an effective harmonic potential, also called pseudopotential, in all three spatial directions. Combining Eqs. (2.9) and (2.15), we obtain this effective trap potential as

\[
\Phi_{eff} = \frac{m}{2q} \sum_{\alpha=x,y,z} \omega_\alpha^2 r_\alpha^2,
\]  

where the \( \omega_\alpha \) are given in Eqs. (2.8) and (2.14). In later chapters we always assume trapping conditions such that the trap potential can be described by the pseudopotential \( \Phi_{eff} \) of Eq. (2.17) above.

Let us finally note that traps designs other than the macroscopic structure shown in Fig. 2.1 have been explored recently. Many groups are also using miniaturized planar ion traps, where the electrode structure is embedded on a chip [44, 45]. In these surface electrode traps the ions also experience nearly harmonic potentials such that the potential of these traps can still be described by Eq. (2.17). However, the trap potentials in the surface electrode traps are typically shallower than those in the macroscopic Paul traps [45].

### 2.1.2. Motion of trapped ions in radio-frequency traps

After we have understood the basic working principle of an ion trap and know the equations of motion for a single trapped ion, we move on to the description of the dynamics of a collection of trapped ions held in the same radio-frequency trap. Due to their charge, the ions repel each other and accordingly their potential energy consists of two contributions now: the potential energy \( V_T \) due to the trap confinement and the potential energy \( V_C \) due to the Coulomb repulsion. Using the trap potential \( \Phi_{eff} \) in Eq. (2.17) we obtain the potential energy \( V = V_T + V_C \) of \( N \) trapped ions of mass \( m \) as

\[
V = \frac{m}{2} \sum_{\alpha=x,y,z} \sum_{i=1}^{N} \omega_\alpha^2 r_\alpha^2 + \sum_{i,j=1}^{N} \frac{1}{8\pi\varepsilon_0} \frac{e^2}{|r_i - r_j|}.
\]
Here $\mathbf{r}_i$ denotes the position of ion $i$, $\varepsilon_0$ is the vacuum permittivity and $e$ is the elementary charge. Adding the kinetic energy $T$ of the ions we obtain the system’s Lagrangian

$$L = T - V = \frac{m}{2} \sum_{i, \alpha} \dot{r}_{i\alpha}^2 - \frac{m}{2} \sum_{i, \alpha} \omega_{i\alpha}^2 \dot{r}_{i\alpha}^2 - \sum_{i \neq j} \frac{e^2}{8 \pi \varepsilon_0 |\mathbf{r}_i - \mathbf{r}_j|}. \quad (2.19)$$

When the ions are brought to sufficiently low temperatures, they form a spatially ordered structure known as Coulomb crystal [46]. The ions are then located at well-defined equilibrium positions and perform only small excursions around these. If we adjust the trapping potentials such that the confinement in two of the directions is much stronger than in the third direction, the ions will arrange on a string along the axis of weakest confinement. A typical choice is $\omega_x, \omega_y \gg \omega_z$, in which case the ions arrange along the $z$-axis. Accordingly, the radial equilibrium positions $x_{0i}^0$ and $y_{0i}^0$ are given by $x_{0i}^0 = y_{0i}^0 = 0$ while the axial equilibrium positions are found by solving

$$\left. \frac{\partial V}{\partial z_i} \right|_{z_i^0} = 0. \quad (2.20)$$

Defining the characteristic length scale

$$l_z = \sqrt[3]{\frac{e^2}{4 \pi \varepsilon_0 m \omega_z^2}} \quad (2.21)$$

we can write Eq. (2.20) in terms of the dimensionless equilibrium positions

$$u_{0i}^0 = z_{0i}^0 / l_z, \quad (2.22)$$

which yields

$$\left. \frac{\partial V}{\partial z_i} \right|_{z_i^0} = u_{0i}^0 - \sum_{j \neq i} \frac{u_{0i}^0 - u_{0j}^0}{|u_{0i}^0 - u_{0j}^0|^3} = 0. \quad (2.23)$$

The advantage of the above representation is that the equation is independent of the specific ion mass and trapping frequency. The values of the $z_{0i}^0$ for a specific setup are easily obtained from Eq. (2.22).

For the cases of $N = 2$ and $N = 3$ ions, Eq. (2.23) can be solved analytically leading to [47]

$$N = 2 : \quad z_1^0 = - \frac{1}{\sqrt{4}} l_z, \quad z_2^0 = - z_1^0 \quad (2.24)$$

$$N = 3 : \quad z_1^0 = - \frac{1}{\sqrt{5}} l_z, \quad z_2^0 = 0, \quad z_3^0 = \frac{1}{\sqrt{5}} l_z. \quad (2.25)$$

For larger values of $N$, the $u_{0i}^0$ have to be calculated numerically. Values of the $u_{0i}^0$ for up to ten ions can be found in [47].

Let us now assume that the ions are sufficiently cold and form a Coulomb crystal. Denoting the small amplitude excursions around the equilibrium positions $r_{0i\alpha}^0$ by $q_{i\alpha}(t)$ we may express the ions’ spatial coordinates as

$$r_{i\alpha}(t) = r_{i\alpha}^0 + q_{i\alpha}(t). \quad (2.26)$$

Due to the small amplitude of the $q_{i\alpha}(t)$ one can approximate the full potential $V$ in Eq. (2.18) to second order in the small displacements

$$V \approx V^{(2)} = \frac{1}{2} \sum_{i, j, \alpha, \beta} \left. \frac{\partial^2 V}{\partial r_{i\alpha} \partial r_{j\beta}} \right|_{r_{0i\alpha}^0} q_{i\alpha} q_{j\beta}. \quad (2.27)$$
Here we omitted the constant zeroth order energy shift, which is immaterial for the dynamics. Note that the first order contribution vanishes because we are expanding around equilibrium. For the linear string configuration, the motion in different spatial directions decouples in the second order approximation, so we can write the potential energy as

$$V^{(2)} = \frac{m\omega_x^2}{2} \sum_{i,j} V_{ij}^{\alpha} q_i q_j \alpha.$$ \hfill (2.28)

In the axial direction the Hessian of the potential evaluated at the ions’ equilibrium positions reads

$$V_{ij}^z = \begin{cases} 1 + 2 \sum_{p \neq j} \frac{1}{|u_{ij} - u_{ij}|} & \text{if } i = j \\ -2 \frac{1}{|u_{ij} - u_{ij}|} & \text{if } i \neq j \end{cases}.$$ \hfill (2.29)

In the radial directions, the Hessians can be written in terms of the trap anisotropies

$$\xi_{\alpha} = \left( \frac{\omega_z}{\omega_{\alpha}} \right)^2, \quad \alpha = x, y.$$ \hfill (2.30)

We have

$$V_{ij}^{\alpha} = \left( \frac{1}{\xi_{\alpha}} + \frac{1}{2} \right) \delta_{ij} - \frac{1}{2} V_{ij}^z, \quad \alpha = x, y,$$ \hfill (2.31)

where $\delta_{ij}$ is the Kronecker delta. Note that small values of the trap anisotropies $\xi_{\alpha}$ imply that the confinement in the radial directions is much stronger than in the axial direction. Let us also remark that the second order expansion of the potential energy is a very good approximation in most experimental situations [39].

We can now write down the Lagrangian of the system in the harmonic approximation. The generalized coordinates are now the $q_i \alpha$. Using Eq. (2.27) we find

$$L = \frac{m}{2} \sum_{i,\alpha} \dot{q}_i^2 \alpha - \frac{m\omega_x^2}{2} \sum_{i,j,\alpha} V_{ij}^{\alpha} q_i q_j \alpha.$$ \hfill (2.32)

The equations of motion for the $q_j \alpha$ are found by evaluating the associated Euler-Lagrange equations

$$m \ddot{q}_i \alpha + m \omega_x^2 \sum_j V_{ij}^{\alpha} q_j \alpha = 0.$$ \hfill (2.33)

Here we used that the $V_{ij}^{\alpha}$ are symmetric. Let us assume that we are expanding around a stable equilibrium. This implies that the $V^{\alpha}$ are positive. From Eq. (2.33) we obtain a set of $N$ coupled differential equations with constant coefficients for each spatial direction. A fundamental set of solutions for such a system is found by using an ansatz of the form $q_i \alpha = b_i e^{i \omega t}$. Since we are expanding around a stable equilibrium, we expect oscillatory solutions of the form [48]

$$q_i \alpha = C b_i \alpha e^{-i\omega t},$$ \hfill (2.34)

where $C$ is an overall scaling factor. Substituting this ansatz into Eq. (2.33) yields

$$-\omega_x^2 b_i \alpha + \omega_x^2 \sum_j V_{ij}^{\alpha} b_j \alpha = 0.$$ \hfill (2.35)

Using the vector $b^{\alpha} = (b_1^{\alpha}, \ldots, b_N^{\alpha})^T$, we can write the above equation as a matrix-vector multiplication

$$\omega_x^2 V^{\alpha} b^{\alpha} = \omega_x^2 b^{\alpha}.$$ \hfill (2.36)
This is essentially the eigenvalue equation of $V^\alpha$. Since the $V^\alpha$ are symmetric and positive, they have $N$ eigenvalues $\lambda_{n,\alpha} > 0$ with orthogonal eigenvectors $b^\alpha_n$. The frequency associated to each $b^\alpha_n$ by Eq. (2.36) is $\omega_{n,\alpha} = \sqrt{\lambda_{n,\alpha}}$. The general solution for $q_{i\alpha}$ is a superposition of all solutions in the form of Eq. (2.34) that fulfill Eq. (2.36) \[ q_{i\alpha} = \sum_{n=1}^{N} C_n b^\alpha_{in} e^{-i \sqrt{\lambda_{n,\alpha}} \omega_z t}, \]

(2.37)

where $b^\alpha_{in}$ is the $i$th component of eigenvector $b^\alpha_n$ in direction $\alpha$, and the scale factors $C_n$ are complex numbers determined from the initial conditions. Note that finally we only consider the real part of the solution.

From Eq. (2.37) we see that the general solution for each of the coordinates is a superposition of oscillations with frequencies $\sqrt{\lambda_{n,\alpha}} \omega_z$ whose relative amplitude is determined by the components $b^\alpha_{in}$ of the eigenvectors of $V^\alpha$. This is a well-known result from the theory of small oscillations [48]. The system is described by a set of normal modes with frequencies $\sqrt{\lambda_{n,\alpha}} \omega_z$ and the $b^\alpha_{in}$ are the normalized amplitudes of mode $n$ in direction $\alpha$ at ion $i$.

A different route to arrive at this result is to perform a normal-mode analysis. In every spatial direction the Lagrangian in Eq. (2.32) is a quadratic form which can be diagonalized by an orthogonal transformation $B^\alpha$ [48]. The transformations $B^\alpha$ are given by the eigenvectors $b^\alpha_n$ of $V^\alpha$

\[ B^\alpha = (b^\alpha_1, \ldots, b^\alpha_N). \]

(2.38)

The $b^\alpha_n$ are mutually orthogonal and form a complete basis such that

\[ \sum_j (B^\alpha_i)^T B^\alpha_j = \sum_j B^\alpha_i B^\alpha_j = \delta_{ik}, \quad \sum_j B^\alpha_i (B^\alpha)^T_j = \sum_j B^\alpha_i b^\alpha_j = \delta_{ik} \]

(2.39)

and

\[ (B^\alpha)^T V^\alpha B^\alpha = \text{diag}(\lambda_1, \ldots, \lambda_N). \]

(2.40)

Let us now introduce the new coordinates $Q_{n,\alpha}$ which are related to the $q_{i\alpha}$ by

\[ q_{i\alpha} = \sum_{n=1}^{N} B^\alpha_{in} Q_{n,\alpha}. \]

(2.41)

Inserting the definition above into the Lagrangian and using the properties of the $B^\alpha$ in Eq. (2.39) yields

\[ L = \frac{m}{2} \sum_{n,\alpha} (\dot{Q}_{n,\alpha}^2 - \omega_{n,\alpha}^2 Q_{n,\alpha}^2), \]

(2.42)

where we defined

\[ \omega_{n,\alpha} = \sqrt{\lambda_{n,\alpha}} \omega_z. \]

(2.43)

The system is now described in terms of $N$ uncoupled harmonic oscillators in each spatial direction. These are called the normal modes. Accordingly, the $Q_{n,\alpha}$ are called the normal coordinates and the $\omega_{n,\alpha}$ the normal mode frequencies. The normal mode frequencies are determined from the eigenvalues of the $V^\alpha$ through Eq. (2.43). The entries $B^\alpha_{in}$ of the matrices $B^\alpha$ correspond to the normalized amplitude of normal mode $n$ at ion $j$ [48].
2. Trapped atomic ions in radio-frequency traps

Normal-mode structure for linear crystals

It is now worth considering Eqs. (2.29) and (2.31) again. We see that $V^x$ and $V^y$ only differ from $V^z$ by a term that is proportional to the identity. Accordingly, all Hessians $V^\alpha$ are diagonalized by the same orthogonal matrix $B$, i.e.

$$B^\alpha = B, \quad \alpha = x, y, z.$$  \hspace{1cm} (2.44)

This means that the structure of the normal modes is the same for all spatial directions.

Let us first consider the axial direction. According to Eq. (2.43) the axial normal mode frequencies are given by

$$\omega_{n,z} = \sqrt{\lambda_{n,z} \omega_z}.$$  \hspace{1cm} (2.45)

We now assume that the $\lambda_{n,z}$ are ordered such that their values increase with increasing $n$. It can be shown [47] that the smallest eigenvalue of $V^z_{ij}$ is always $\lambda_{1,z} = 1$ with eigenvector

$$b_1 = \frac{1}{\sqrt{N}} (1, \ldots, 1)^T.$$  \hspace{1cm} (2.46)

Accordingly, all ions move in phase in this mode, and we can identify it as the center-of-mass mode. Since $\lambda_1 = 1$ is the smallest eigenvalue, the center-of-mass mode is the energetically lowest lying mode in the axial direction and its frequency coincides with the trap frequency. Further, it can be shown [47] that the second eigenvalue is $\lambda_{2,z} = 3$ with eigenvector

$$b_2 = \frac{1}{\sqrt{\sum_i u_i^2}} (u_1^0, \ldots, u_N^0)^T,$$  \hspace{1cm} (2.47)

where the $u_i^0$ are the scaled equilibrium positions of Eq. (2.23). Accordingly, the second mode is called stretch or breathing mode. Higher eigenvalues and the corresponding eigenvectors have to be computed numerically. For up to $N = 10$ ions, the energetically highest lying mode in the axial direction, corresponding to the eigenvalue $\lambda_{z,N}$, is the mode where neighboring ions perform out-of-phase oscillations. Although we are not aware of a general proof, it seems likely that this is the case for arbitrary $N$ [32].

In the radial directions, the normal mode frequencies read

$$\omega_{n,\alpha} = \sqrt{\lambda_{n,\alpha} \omega_\alpha}, \quad \alpha = x, y,$$  \hspace{1cm} (2.48)

where due to Eq. (2.31), we can write the $\lambda_{n,\alpha}$ as

$$\lambda_{n,\alpha} = \frac{1}{\varepsilon_\alpha} + \frac{1}{2} - \frac{\lambda_{n,z}}{2}, \quad \alpha = x, y.$$  \hspace{1cm} (2.49)

Accordingly, the eigenvalues in the radial directions decrease with increasing $n$. Note, however, that the eigenvalues in the axial and radial directions with equal index $n$ have the same associated eigenvector, and thus the modes with equal index $n$ have the same structure. It follows from our considerations of the axial modes that the energetically highest lying mode in the radial directions is the center-of-mass mode. The mode corresponding to the axial breathing mode is dubbed the rocking or tilt mode in the radial directions and is the energetically second highest mode. Finally, the energetically lowest lying mode is the mode where neighboring ions move out of phase and is called the zigzag mode in the radial directions. The mode structure for a linear crystal of $N = 3$ ions is illustrated in Fig. 2.2.
2.1. Trapping ions

Figure 2.2.: Normal modes for a linear crystal of three trapped $^{40}\text{Ca}^+$ ions. Part a) shows the normal mode frequencies for $\{\omega_x, \omega_y, \omega_z\} = 2\pi \{4,5,2\} \text{MHz}$. The ordering of the $y$-modes is the same as that of the $x$-modes. Parts b) and c) show the normal mode structure in the axial and radial directions, respectively, and the names by which the modes are commonly referred to. Note that the name “Egyptian” mode for the third mode in the axial directions is only used for $N = 3$ ions.

2.1.3. Quantized motion of trapped ions in radio-frequency traps

The description of the motion of ions in radio-frequency traps in the previous section was purely classical. In this section we will quantize the motion. In standard quantum mechanics, one usually considers the Hamiltonian rather than the Lagrangian. Introducing the generalized momenta

$$p_{j\alpha} = \frac{\partial L}{\partial \dot{q}_{j\alpha}} = m\dot{q}_{j\alpha},$$

we can transform the Lagrangian in Eq. (2.32) to the corresponding Hamiltonian

$$H = \sum_{i,\alpha} \frac{p_{i\alpha}^2}{2m} + \frac{m\omega^2}{2} \sum_{i,j,\alpha} V_{ij}^\alpha q_{i\alpha} q_{j\alpha}.$$  (2.51)

Using the orthogonal matrix $B$ from Eq. (2.44) we can define the normal coordinates $Q_{n,\alpha}$ and momenta $P_{n,\alpha}$ analogous to Eq. (2.41)

$$q_{j\alpha} = \sum_n B_{jn} Q_{n,\alpha}, \quad p_{j\alpha} = \sum_n B_{jn} P_{n,\alpha}.$$  (2.52)

Inserting the above definitions in Eq. (2.51) and using the orthogonality properties of the matrix $B$ in Eq. (2.39), the Hamiltonian takes the form

$$H = \sum_{n,\alpha} \left( \frac{p_{n,\alpha}^2}{2m} + \frac{m}{2} \omega_{n,\alpha}^2 Q_{n,\alpha}^2 \right),$$  (2.53)

which is the Hamiltonian of $N$ independent harmonic oscillators in each spatial directions. We now quantize each of the oscillators by replacing the generalized coordinates and momenta by the corresponding operators $\hat{Q}_{n,\alpha}$ and $\hat{P}_{n,\alpha}$ that obey the canonical commutation relation [49]

$$[\hat{Q}_{n,\alpha}, \hat{P}_{n',\alpha'}] = i\hbar \delta_{\alpha,\alpha'} \delta_{n,n'},$$  (2.54)

where $\hbar$ is the reduced Planck constant. Let us now introduce the usual annihilation and creation operators $\hat{a}_{n,\alpha}$ and $\hat{a}_{n,\alpha}^\dagger$. The destruction operator $\hat{a}_{n,\alpha}$ annihilates an excitation of mode $n$ in direction $\alpha$ and the creation operator $\hat{a}_{n,\alpha}^\dagger$ creates an excitation in the corresponding mode. Using
these operators we can write the generalized coordinate and momentum operators as \[49\]
\[
\hat{Q}_{n,\alpha} = \sqrt{\frac{\hbar}{2m\omega_{n,\alpha}}} (\hat{a}_{n,\alpha} + \hat{a}_{n,\alpha}^\dagger),
\]
\[
\hat{P}_{n,\alpha} = i\sqrt{\frac{\hbar m\omega_{n,\alpha}}{2}} (\hat{a}_{n,\alpha} - \hat{a}_{n,\alpha}^\dagger).
\] (2.55)

The annihilation and creation operators satisfy the following commutation relations
\[
[\hat{a}_{n,\alpha}, \hat{a}_{n',\alpha'}] = [\hat{a}_{n,\alpha}^\dagger, \hat{a}_{n',\alpha'}^\dagger] = 0, \quad [\hat{a}_{n,\alpha}, \hat{a}_{n',\alpha'}^\dagger] = \delta_{\alpha,\alpha'}\delta_{n,n'}.
\] (2.56)

Substituting \(\hat{Q}_{n,\alpha}\) and \(\hat{P}_{n,\alpha}\) by the annihilation and creation operators according to Eq. (2.55), we can cast the Hamiltonian in Eq. (2.53) into the standard form
\[
\hat{H} = \sum_{n,\alpha} \hbar \omega_{n,\alpha} (\hat{a}_{n,\alpha}^\dagger \hat{a}_{n,\alpha} + \frac{1}{2} \mathbb{1}),
\] (2.57)

where \(\mathbb{1}\) is the identity operator. Note that from now on we omit the ground state energy of the Hamiltonian because it has no influence on the oscillator dynamics.

Combining Eqs. (2.52) and (2.55), we write the single ion position and momentum operators as
\[
\hat{q}_{j\alpha} = \sum_n B_{jn} \sqrt{\frac{\hbar}{2m\omega_{n,\alpha}}} (\hat{a}_{n,\alpha} + \hat{a}_{n,\alpha}^\dagger),
\] (2.58)
\[
\hat{p}_{j\alpha} = \sum_n iB_{jn} \sqrt{\frac{\hbar m\omega_{n,\alpha}}{2}} (\hat{a}_{n,\alpha}^\dagger - \hat{a}_{n,\alpha}).
\] (2.59)

### 2.1.4. Mixed species crystals

As a last point in this section we discuss the motion of mixed-species ion crystals in a trap. Mixed-species ion crystals consist either of ions of different elements or ions of different isotopes of the same element. The diagonalization of the Lagrangian in Eq. (2.32) is somewhat more complicated in this case because the ions have different masses. In the following we will again focus on linear crystals and restrict our considerations to the axial direction.

For a collection of \(N\) ions with different masses \(m_i\), the axial contribution of the potential in Eq. (2.18) takes the form
\[
V = \sum_{i=1}^{N} \frac{1}{2} a_0 z_i^2 + \sum_{i \neq j}^{N} \frac{1}{8\pi\varepsilon_0} \frac{e^2}{|z_i - z_j|}.
\] (2.60)

The radial equilibrium positions remain at \(x_0^i = y_0^i = 0\). The axial equilibrium positions of the ions are determined by Eq. (2.20) which we can cast into the form in Eq. (2.23), if we redefine the scaled equilibrium positions as
\[
u_0^i = z_0^i / \tilde{l}_z,
\] (2.61)
with the characteristic length scale
\[
\tilde{l}_z = \sqrt{\frac{e^2}{4\pi\varepsilon_0 a_0}}.
\] (2.62)
2.1. Trapping ions

Note that \( \tilde{I}_z \) reduces to \( I_z \) in Eq. (2.21) for \( a_0 = m\omega_z^2 \). Hence, also the axial equilibrium positions are the same for the mixed-species crystals and the harmonic approximation of the Lagrangian in Eq. (2.19) reads

\[
L = \sum_i \frac{m_i}{2} \ddot{q}_{iz}^2 - \frac{a_0}{2} \sum_{i,j} V^z_{ij} \dot{q}_{iz} \dot{q}_{jz},
\]  

(2.63)

where \( V^z \) is defined in Eq. (2.29) above. The problem of finding the normal modes of the system is now hampered by the fact that the masses are not homogeneous. The equation that determines the normal mode frequencies and amplitudes corresponding to Eq. (2.36) now reads [50]

\[
a_0 \sum_j V^z_{ij} \tilde{b}_{jn} = \omega_n^2 m_i b_{in},
\]  

(2.64)

which is a generalized eigenvalue problem. The eigenvectors \( b_n \) fulfilling Eq. (2.64) are then orthogonal with respect to a metric tensor \( T_{ij} = m_i \delta_{ij} \) [50]. In order to bring the system to a diagonal form we perform a change of coordinates. We write all masses \( m_i \) in terms of some reference mass \( m_0 \)

\[
m_i = \mu_i m_0,
\]  

(2.65)

and define the new set of coordinates

\[
\tilde{q}_{iz} = \sqrt{\mu_i} q_{iz}.
\]  

(2.66)

In terms of the new coordinates \( \tilde{q}_{iz} \) the Lagrangian in Eq. (2.63) becomes

\[
L = \frac{m_0}{2} \left( \sum_i \tilde{q}_{iz}^2 - \dot{\omega}_n^2 \sum_{i,j} V^z_{ij} \tilde{q}_{iz} \tilde{q}_{jz} \right),
\]  

(2.67)

Here, we have introduced \( \dot{\omega}_n^2 = a_0 / m_0 \) and

\[
\tilde{V}^z_{ij} = V^z_{ij} / \sqrt{\mu_i \mu_j}.
\]  

(2.68)

The matrix \( \tilde{V}^z_{ij} \) is symmetric and positive because \( V^z_{ij} \) is symmetric and positive. It thus has \( N \) eigenvalues \( \tilde{\lambda}_{n,z} > 0 \) with mutually orthogonal eigenvectors \( \tilde{b}_n \). Hence, we can diagonalize the Lagrangian in Eq. (2.67) by an orthogonal transformation

\[
\tilde{B} = (\tilde{b}_1, \ldots, \tilde{b}_N),
\]  

(2.69)

as we did with the Lagrangian in Eq. (2.32) above. Note that \( \tilde{B} \) has the properties in Eq. (2.39) and satisfies Eq. (2.40) for \( \tilde{V}^z \). Analogous to Eq. (2.41) we define the normal coordinates \( \tilde{Q}_{n,z} \)

\[
\tilde{q}_{iz} = \sum_n \tilde{b}_{jn} \tilde{Q}_{n,z}.
\]  

(2.70)

Inserting the normal coordinates into Eq. (2.67) and using the properties of \( \tilde{B} \) we obtain the Lagrangian

\[
L = \frac{m_0}{2} \sum_n (\tilde{Q}_{n,z}^2 - \dot{\omega}_{n,z}^2 \tilde{Q}_{n,z}^2),
\]  

(2.71)

where

\[
\dot{\omega}_{n,z} = \sqrt{\tilde{\lambda}_{n,z}} \omega_z.
\]  

(2.72)

Again, we find the Lagrangian of \( N \) independent harmonic oscillators with frequencies \( \dot{\omega}_{n,z} \).

We can now quantize the motion following the procedure presented in the previous section. Introducing the generalized momenta

\[
\tilde{p}_{iz} = m_0 \dot{\tilde{q}}_{iz},
\]  

(2.73)
we obtain the Hamiltonian corresponding to the Lagrangian in Eq. (2.67)

\[ H = \left( \sum_i \frac{\tilde{p}_{iz}^2}{2m_0} + \tilde{\omega}_z^2 \sum_{i,j} \tilde{V}_{ij} \tilde{q}_{iz} \tilde{q}_{jz} \right) \]  
\[ (2.74) \]

Next we introduce the normal momenta and coordinates

\[ \tilde{p}_{jz} = \sum_n \tilde{B}_{jn} \tilde{P}_{nz}, \quad \tilde{q}_{jz} = \sum_n \tilde{B}_{jn} \tilde{Q}_{nz} \]  
\[ (2.75) \]

to obtain the diagonal form of the Hamiltonian. Replacing the normal-mode momenta and coordinates by the corresponding operators and introducing the annihilation and creation operators as in Eq. (2.55) yields the standard Hamiltonian of a collection of harmonic oscillators

\[ \hat{H} = \sum_n \hbar \tilde{\omega}_{nz} \hat{a}^\dagger_{nz} \hat{a}_{nz}, \]  
\[ (2.76) \]

where we already omitted the ground state energy and \( \hat{a}_{nz} \) annihilates an excitation in mode \( n \) while \( \hat{a}^\dagger_{nz} \) creates an excitation in mode \( n \).

Finally, the single ion position and momentum operators \( \hat{q}_{jz} \) and \( \hat{p}_{jz} \) can be written in terms of the annihilation and creation operators of the normal modes as

\[ \hat{q}_{jz} = \sum_n \sqrt{\frac{\hbar}{2m_j \tilde{\omega}_{nz}}} (\hat{a}_{nz} + \hat{a}^\dagger_{nz}), \]  
\[ (2.77) \]

\[ \hat{p}_{jz} = \sum_n i \sqrt{\frac{\hbar m_j \tilde{\omega}_{nz}}{2}} (\hat{a}^\dagger_{nz} - \hat{a}_{nz}). \]  
\[ (2.78) \]

Note, however, that \( \hat{q}_{jz} \) and \( \hat{p}_{jz} \) are given in the skew coordinates defined by Eq. (2.66) and hence do not describe the physical single ion position and momentum. Using Eqs. (2.66) and (2.77) we readily obtain the operator for the physical coordinates

\[ \hat{q}_{jz} = \frac{1}{\sqrt{m_j}} \sum_n \tilde{B}_{jn} \sqrt{\frac{\hbar}{2\tilde{\omega}_{nz}}} (\hat{a}_{nz} + \hat{a}^\dagger_{nz}). \]  
\[ (2.79) \]

The conjugate momenta \( p_{jz} \) are obtained from the Lagrangian in Eq. (2.63) and read \( p_{jz} = m_j \dot{q}_{jz} \). Combining Eqs. (2.65), (2.66) and (2.73) we find \( p_{jz} = \sqrt{\mu_j} \tilde{p}_{jz} \). Inserting the result of Eq. (2.78) into this expression yields

\[ \hat{p}_{jz} = \sqrt{m_j} \sum_n i \tilde{B}_{jn} \sqrt{\frac{\hbar \tilde{\omega}_{nz}}{2}} (\hat{a}^\dagger_{nz} - \hat{a}_{nz}). \]  
\[ (2.80) \]

Note that, in order to ease notation, we omit operator hats from now on.

### 2.2. Light-matter interaction

In this section, we review the interaction of trapped ions with electromagnetic fields applied in addition to the trapping fields. With properly chosen fields the internal levels of the ions can be coherently manipulated and it is possible to couple the internal degrees of freedom of the ions to the motional degrees of freedom. These interactions are the standard tools for the manipulation of trapped ions in current experiments and we discuss both types of interaction here. We consider the atom-light interaction in the so-called semi-classical approximation, where the ions are treated quantum mechanically while the radiation is treated classically.
2.2. Light-matter interaction

2.2.1. Electric dipole Hamiltonian

In classical electrodynamics the electric and magnetic fields $E(r,t)$ and $B(r,t)$ can be derived from a vector potential $A(r,t)$ and a scalar potential $U(r,t)$ through

\[
E(r,t) = -\frac{\partial}{\partial t}A(r,t) - \nabla U(r,t),
\]

\[
B(r,t) = \nabla \times A(r,t).
\]

However, the vector and scalar potentials are not uniquely defined. In fact, there is an infinite set of pairs $\{A(r,t), U(r,t)\}$ from which the same fields $E(r,t)$ and $B(r,t)$ can be derived. Two pairs of potentials $\{A(r,t), U(r,t)\}$ and $\{A'(r,t), U'(r,t)\}$ yielding the same fields are connected by a gauge transformation

\[
A'(r,t) = A(r,t) - \nabla \chi(r,t),
\]

\[
U'(r,t) = U(r,t) - \frac{\partial}{\partial t} \chi(r,t),
\]

where $\chi(r,t)$ is an arbitrary scalar function.

Let us consider a plane electromagnetic wave of frequency $\omega$ and wave vector $k$. The electric and magnetic fields then have the form

\[
E(r,t) = E_0 \cos(\omega t - kr + \phi),
\]

\[
B(r,t) = \frac{e_k \times E_0}{c} \cos(\omega t - kr + \phi).
\]

Here, $E_0$ is the electric field vector, $\phi$ a phase and $c$ the speed of light. The vector $e_k$ is a unit vector along the direction of propagation $k$ of the wave. Working in the so-called Coulomb gauge, where $\nabla A(r,t) = 0$, we can write the plane wave in terms of the potentials [51]

\[
\tilde{A}(r,t) = -\frac{E_0}{\omega} \sin(\omega t - kr + \phi),
\]

\[
U_\perp(r,t) = 0.
\]

It can readily be checked that the potentials above yield the correct form for the fields using the condition $k \cdot E_0 = 0$.

We now consider the simplest case of an atom interacting with an electromagnetic field, i.e. the interaction of a hydrogen atom with an externally applied electromagnetic field. More specifically, we consider the interaction of the externally applied field and the electron of the hydrogen atom. We follow the discussions in [49, 51]. The Hamiltonian of the electron is that of a charged particle interacting with an electromagnetic field

\[
H = \frac{1}{2m_e}(\mathbf{p} + e\tilde{A}(r,t))^2 - eU(r,t) + \frac{e}{m_e} \mathbf{S} \cdot \mathbf{B}(r,t),
\]

where $m_e$ is the electron’s mass, $\mathbf{p}$ and $\mathbf{r}$ its momentum and position operators respectively, and $e$ the elementary charge. The vector and scalar potentials in Eq. (2.89) describe both the externally applied field and the potential of the nucleus. The last term on the right-hand side describes the interaction of the electron’s magnetic moment, i.e. the electron’s spin, $\mathbf{S}$ with the magnetic field component of the plane wave. In the Coulomb gauge, the applied field is described by the potentials in Eqs. (2.87) and (2.88). For the static electric field of the nucleus, the vector potential is zero and the scalar potential is the usual Coulomb potential

\[
U_C(r) = \frac{1}{4\pi\varepsilon_0} \frac{e}{|\mathbf{r} - \mathbf{r}_0|}.
\]
2. Trapped atomic ions in radio-frequency traps

where $\varepsilon_0$ is the vacuum permittivity and $\mathbf{r}_0$ the position of the nucleus. Using that $[\hat{\mathbf{A}}(\mathbf{r}, t), p] = 0$ in the Coulomb gauge, we can cast the Hamiltonian in Eq. (2.89) into the form

$$ H = H_{\text{at}} + H_{\text{int}}. $$

(2.91)

Here

$$ H_{\text{at}} = \frac{p^2}{2m_e} + V_C(\mathbf{r}) $$

(2.92)

is the atomic Hamiltonian with $V_C(\mathbf{r}) = eU_C(\mathbf{r})$ the energy due to the Coulomb interaction of the electron with the nucleus, and

$$ H_{\text{int}} = e\frac{m_e}{e} \mathbf{p} \cdot \hat{\mathbf{A}}(\mathbf{r}, t) + \frac{e^2}{m_e} \mathbf{S} \cdot \mathbf{B}(\mathbf{r}, t) + \frac{e^2 \hat{\mathbf{A}}(\mathbf{r}, t)^2}{2m_e} $$

(2.93)

describes the interaction of the electron with the applied field. The third term in $H_{\text{int}}$ can be neglected unless we are dealing with very intense fields [49] such that

$$ H_{\text{int}} \approx H_{\text{II}}(t) + H_{\text{I}}(t) = e\frac{m_e}{e} \mathbf{p} \cdot \hat{\mathbf{A}}(\mathbf{r}, t) + e\frac{m_e}{e} \mathbf{S} \cdot \mathbf{B}(\mathbf{r}, t). $$

(2.94)

Let us now inspect the interaction Hamiltonian in Eq. (2.94) in more detail. First, we consider the relative magnitude of the matrix elements of the operators $H_{\text{I}}$ and $H_{\text{II}}$, which we write as $H_{\text{I}}/H_{\text{II}}$. The elements of $\mathbf{S}$ are of the order of the reduced Planck constant $\hbar$ and those of $\mathbf{B}$ of order $|E_0|k/\omega = E_0k/\omega$. This leads us to the estimate

$$ \frac{H_{\text{II}}(t)}{H_{\text{I}}(t)} \approx \frac{\hbar k}{p}, $$

(2.95)

where $p = |\mathbf{p}|$ is the modulus of the electron’s momentum. The electron is bound to the atom and therefore confined to a length scale of the order of the Bohr radius $a_0$. With the position-momentum uncertainty relation we can estimate $\hbar/p \leq a_0$. Writing $k = 2\pi/\lambda$, where $\lambda$ is the wavelength of the applied radiation, we obtain

$$ \frac{H_{\text{II}}(t)}{H_{\text{I}}(t)} \approx \frac{a_0}{\lambda}. $$

(2.96)

The typical wavelengths $\lambda$ in quantum optics experiments are of the order of hundreds of nanometers while the atomic dimensions for hydrogen are of the order of $a_0 = 0.053\,\text{nm}$. In this case we have

$$ \frac{a_0}{\lambda} \ll 1. $$

(2.97)

Therefore, we also neglect $H_{\text{II}}(t)$ and approximate the interaction Hamiltonian as

$$ H_{\text{I}}(t) = e\frac{m_e}{e} \mathbf{p} \cdot \hat{\mathbf{A}}(\mathbf{r}, t). $$

(2.98)

Now, we can write the electron’s position as $\mathbf{r} = \mathbf{r}_0 + \delta \mathbf{r}$, where $\mathbf{r}_0$ is the position of the nucleus and $\delta \mathbf{r}$ is the distance of the electron to the nucleus, which is of the order of the Bohr radius $a_0$. If we then write the sine in $\hat{\mathbf{A}}(\mathbf{r}, t)$, given in Eq. (2.87), in terms of two complex exponentials, we have

$$ e^{\pm i\mathbf{k} \cdot \mathbf{r}} = e^{\pm i\mathbf{k} \cdot \mathbf{r}_0} e^{\pm i\mathbf{k} \cdot \delta \mathbf{r}} = e^{\pm i\mathbf{k} \cdot \mathbf{r}_0} (1 \pm i\mathbf{k} \cdot \delta \mathbf{r} - \frac{1}{2} (\mathbf{k} \cdot \delta \mathbf{r})^2 + \ldots). $$

(2.99)

The terms $\mathbf{k} \cdot \delta \mathbf{r}$ are of the order $a_0/\lambda \ll 1$ which we neglected in going from Eq. (2.94) to Eq. (2.98). Hence, for a consistent description we replace the electron’s coordinate $\mathbf{r}$ by
the coordinate of the nucleus \( r_0 \) in the applied field. The interaction Hamiltonian then takes the form

\[
H_{\text{int}} = \frac{e}{m_e} \mathbf{p} \cdot \tilde{\mathbf{A}}(r_0, t).
\]

(2.100)

The approximation we have performed here is called the electric dipole approximation [49] and the interaction Hamiltonian \( H_{\text{int}} \) we obtained is called the electric dipole Hamiltonian. Physically, we can understand this approximation in the following way. As we stated above, the typical wavelengths \( \lambda \) in quantum optics experiments are of the order of hundreds of nanometers while the atomic dimensions are of the order of the Bohr Radius \( a_0 = 0.053 \text{nm} \). Due to this large difference in length scales, the applied field appears practically constant over the spatial extent of the atom, such that we can replace the electron’s coordinate by that of the nucleus [51].

A very useful form of the electric dipole Hamiltonian is obtained in the so-called Goppert-Mayer gauge, which is obtained from the Coulomb gauge by the gauge transformation

\[
\chi(r, t) = -(r - r_0) \cdot \tilde{\mathbf{A}}(r_0, t).
\]

(2.101)

According to Eqs. (2.83) and (2.84), the transformed potentials read

\[
A'(r, t) = \tilde{\mathbf{A}}(r, t) - \tilde{\mathbf{A}}(r_0, t),
\]

(2.102)

\[
U'(r, t) = U_C(r) + (r - r_0) \cdot \frac{\partial}{\partial t} \tilde{\mathbf{A}}(r_0, t).
\]

(2.103)

With the transformed potentials, the Hamiltonian in Eq. (2.89) becomes

\[
H = \frac{1}{2m_e} (\mathbf{p} + eA'(r, t))^2 + V_C(r) + \frac{e}{m_e} \mathbf{S} \cdot \mathbf{B}(r, t) + e(r - r_0) \cdot \mathbf{E}(r_0, t),
\]

(2.104)

where we used \( \mathbf{E}(r_0, t) = -\frac{\partial}{\partial t} \tilde{\mathbf{A}}(r_0, t) \), see Eqs. Eq. (2.81) and (2.87), (2.88). We now introduce the electric dipole operator

\[
\mathbf{d} = -e(r - r_0)
\]

(2.105)

such that we can write the Hamiltonian in Eq. (2.104) as

\[
H = \frac{1}{2m_e} (\mathbf{p} + eA'(r, t))^2 + V_C(r) + \frac{e}{m_e} \mathbf{S} \cdot \mathbf{B}(r, t) - \mathbf{d} \cdot \mathbf{E}(r_0, t).
\]

(2.106)

In the electric dipole approximation, we have \( A'(r, t) \rightarrow A'(r_0, t) \) and we neglect the term \( \sim \mathbf{S} \cdot \mathbf{B}(r, t) \). Then, the above Hamiltonian simplifies to

\[
H = \frac{1}{2m_e} \mathbf{p}^2 + V_C(r) - \mathbf{d} \cdot \mathbf{E}(r_0, t) = H_{\text{at}} + H_d.
\]

(2.107)

Here we have introduced the electric dipole Hamiltonian

\[
H_d = -\mathbf{d} \cdot \mathbf{E}(r_0, t).
\]

(2.108)

Let us close this section with the remark that the above calculation was performed for hydrogen but the electric dipole Hamiltonian can be derived for the outer electron of any atom, thus also involving atoms with more electrons [51].
2. Trapped atomic ions in radio-frequency traps

2.2.2. Atomic transitions and the rotating wave approximation

By applying an electromagnetic field to an atom or trapped ion we can drive transitions between internal states of the atom or ion. Transitions that occur due to the coupling of two states through the electric dipole Hamiltonian in Eq. (2.108) are called (electric) dipole transitions. Let us consider an atom whose internal states of energy \( \varepsilon_F \) we denote by \( |F\rangle \) leading to the atomic Hamiltonian

\[
H_{at} = \sum_F \varepsilon_F |F\rangle \langle F|.
\]

(2.109)

Let us further consider that the atom is irradiated by a plane electromagnetic wave of the form given in Eq. (2.85) and that the interaction is described by the electric dipole Hamiltonian in Eq. (2.108). In the atomic basis the electric dipole Hamiltonian reads

\[
H_d = \sum_{F,F'} -\langle F' | \mathbf{d} \cdot \mathbf{E}(r_0,t) | F \rangle \langle F | F' \rangle \langle F | = \sum_{F,F'} e^{i (F' | \mathbf{r} \cdot \mathbf{E}_0 | F)} \cos(\omega t + \phi) | F' \rangle \langle F|,
\]

(2.110)

where we have assumed \( r_0 = 0 \). The atomic states \( |F\rangle \) have a definite parity \( \Pi |F\rangle = \pi_F |F\rangle \), where \( \Pi \) is the parity operator and \( \pi_F = +1, -1 \) [52]. Since the electric dipole Hamiltonian is odd under parity, there can only be transitions between states of different parity. This can easily be seen by considering

\[
\langle F' | \mathbf{r} \cdot \mathbf{E}_0 | F \rangle = \langle F' | \Pi | \Pi \mathbf{r} \cdot \mathbf{E}_0 \Pi | F \rangle = -\pi_F \pi_F' \langle F' | \mathbf{r} \cdot \mathbf{E}_0 | F \rangle,
\]

(2.111)

from which it follows that \( \langle F' | \mathbf{r} \cdot \mathbf{E}_0 | F \rangle = -\langle F' | \mathbf{r} \cdot \mathbf{E}_0 | F \rangle = 0 \) for \( \pi_F = \pi_F' \). Defining the Rabi frequencies

\[
\Omega_{F'F} = e^{i (F' | \mathbf{r} \cdot \mathbf{E}_0 | F)} \frac{\hbar}{\hbar}
\]

(2.112)

we can write the dipole Hamiltonian as

\[
H_d = \hbar \sum_{F \neq F'} \Omega_{F'F} \cos(\omega t + \phi) | F' \rangle \langle F|.
\]

(2.113)

Thus, the Rabi frequencies determine the strength of the coupling between two states. Transitions which have a vanishing matrix element of the electric dipole Hamiltonian are called electric dipole forbidden. Yet, a vanishing electric dipole Rabi frequency does not mean that two states cannot be coupled by electromagnetic radiation. Two such states can be connected by a magnetic dipole transition, where the magnetic dipole moment of the transition couples to the magnetic field component of the radiation, or an electric quadrupole transition, where the electronic quadrupole moment couples to the gradient of the electric field. The magnetic dipole and electric quadrupole Hamiltonians are obtained from the interaction Hamiltonian in Eq. (2.94) considering the contributions that are first order in \( a_0/\lambda \) [49]. The matrix elements of the two Hamiltonians are of the same order of magnitude but are much smaller than those of the electric dipole Hamiltonian [49].

Let us close this section by introducing a very useful approximation called the rotating wave approximation. To this end, we consider an atom initially in eigenstate \( |k\rangle \) of \( H_d \) which is subject to laser radiation. We assume that the atom-field interaction is described by the Hamiltonian \( H_d \) in Eq. (2.113). We now want to calculate the amplitude to make a transition to a different eigenstate \( |j\rangle \) that has a non-vanishing electric dipole coupling to state \( |k\rangle \) in a time \( t \). In first-order time-dependent perturbation theory this amplitude is given by

\[
\langle k \rightarrow j \rangle^{(1)}(t) = -\frac{i}{\hbar} \int_0^t dt_1 \langle j | H_d(t_1) | k \rangle e^{i \omega t_1},
\]

(2.114)
where we have introduced $\omega_{jk} = (\epsilon_j - \epsilon_k)/\hbar$. Expanding the cosine in terms of complex exponentials we obtain

$$
\varepsilon_{k\rightarrow j}^{(1)}(t) = -\frac{i}{2} \frac{\Omega_{jk}}{\omega - \omega_{jk}} \int_0^t dt' \left( e^{i(\omega + \omega_{jk})t} e^{i\delta} + e^{-i(\omega - \omega_{jk})t} e^{-i\delta} \right) = \frac{\Omega_{jk}}{2} \left( \frac{e^{-i(\omega - \omega_{jk})t} - 1}{\omega - \omega_{jk}} - \frac{e^{i(\omega + \omega_{jk})t} - 1}{\omega + \omega_{jk}} e^{i\delta} \right).$$

(2.115)

Let us now assume that $\Omega_{jk} \ll \omega_{jk}$. Note that actually $\Omega_{jk} \in \mathbb{C}$ and possibly both $\Omega_{jk}$ and $\omega_{jk}$ are negative. When writing $\Omega_{jk} \ll \omega_{jk}$ we actually mean $|\Omega_{jk}| \ll |\omega_{jk}|$. We follow this convention, which is common in quantum optics, throughout this thesis and omit the modulus operators in the following. In case $\Omega_{jk} \ll \omega_{jk}$, the above amplitude is negligible unless $\omega \approx \omega_{jk}$. Thus, not surprisingly, transitions can only be driven if the energy of the applied radiation matches the energy difference between the two states. We now assume $\omega, \omega_{jk} > 0$ and $\omega \approx \omega_{jk}$ so that

$$
\omega = \omega_{jk} + \delta_{jk}, \quad \text{where} \quad \delta_{jk} \ll \omega_{jk}.
$$

(2.116)

Then the second term in the last line of Eq. (2.115) will always be only a small correction to the first one such that we can write

$$
\varepsilon_{k\rightarrow j}^{(1)}(t) \approx -\frac{i}{2} \frac{\Omega_{jk}}{\omega - \omega_{jk}} \left( e^{-i(\omega - \omega_{jk})t} - 1 \right) e^{i\delta} = \frac{\Omega_{jk}}{2} \left( e^{-i\delta_{jk}t} - 1 \right) e^{i\delta},
$$

(2.117)

which yields the transition probability

$$
P_{k\rightarrow j}^{(1)} = |\varepsilon_{k\rightarrow j}^{(1)}(t)|^2 = \frac{|\Omega_{jk}|^2}{\delta_{jk}^2} \sin^2 \left( \frac{\delta_{jk}t}{2} \right).
$$

(2.118)

This procedure of neglecting the “fast rotating” term with the time dependence $\omega + \omega_{jk} \gg \omega - \omega_{jk}$ is called the rotating wave approximation. In case $\delta_{jk} \gg \Omega_{jk}$, the probability in Eq. (2.118) is also negligible. Hence, more generally one can neglect time-dependent terms of the form $\Omega e^{i\omega t}$ in a Hamiltonian when $\omega \gg \Omega$. This is a more general statement of the rotating wave approximation, which is a well-known and frequently-used procedure in quantum optics [51].

Let us now assume $\omega > 0$ and $\omega_{jk} > 0$ for all $j$. For $\Omega_{jk} \ll \omega_{jk}$ the $e^{i\omega t}$ term of the cosine in the Hamiltonian in Eq. (2.113) always produces only a small correction to the $e^{-i\omega t}$ term. Hence, for $\Omega_{jk} \ll \omega_{jk}$ we can always apply a rotating wave approximation to this term. With this knowledge, we neglect this contribution already in the Hamiltonian. In fact, this is what is commonly done in calculations in quantum optics [53,54]. The Hamiltonian of Eq. (2.113) then in a rotating wave approximation becomes

$$
H_{\text{d}} = \hbar \sum_{j \neq k} \frac{\Omega_{jk}}{2} e^{-i(\omega t + \phi)} |j\rangle \langle k| + \text{H.c.},
$$

(2.119)

where the abbreviation H.c. stands for “Hermitian conjugate”.

### 2.2.3. The two-level approximation

Let us now consider an atom irradiated by an electromagnetic wave. And let us assume that the detuning of the field is much larger than the Rabi frequency, i.e. $\delta_{jk} \gg \Omega_{jk}$, for all transitions except for one between two states which we denote $|e\rangle$ and $|g\rangle$. Then, by Eq. (2.118) only transitions between states $|e\rangle$ and $|g\rangle$ will take place and we can neglect all other transitions. Of
course, the atom needs to be in one of the two states or a superposition thereof, initially. In this case we can reduce the atomic Hamiltonian in Eq. (2.109) for the internal atomic levels to

\[ H_{\text{at}} = \sum_{F=e,g} \epsilon_F |F\rangle\langle F| = \epsilon_e |e\rangle\langle e| + \epsilon_g |g\rangle\langle g|. \]  

(2.120)

To cast the above Hamiltonian into its standard form we use that \(|e\rangle\) and \(|g\rangle\) form a basis of the two-dimensional space of the internal levels, i.e. \(|e\rangle\langle e| + |g\rangle\langle g| = \mathbb{I}_2\), and that adding a contribution proportional to the unit operator \(\mathbb{I}_2\) to the Hamiltonian only shifts the zero of the energy. Adding \(-\frac{\epsilon_e + \epsilon_g}{2} \mathbb{I}_2\) to the Hamiltonian in Eq. (2.120) yields

\[ H_{\text{at}} = \frac{\hbar \omega_0}{2} \sigma^z, \]  

(2.121)

where we have introduced the Pauli operator

\[ \sigma^z = |e\rangle\langle e| - |g\rangle\langle g|. \]  

(2.122)

and the transition frequency

\[ \omega_0 = \frac{\epsilon_e - \epsilon_g}{\hbar}. \]  

(2.123)

The dipole Hamiltonian in Eq. (2.113) for the two-level system in a rotating wave approximation becomes

\[ H_d = \frac{\hbar \Omega}{2} e^{-i(\omega t + \phi)} \sigma^+ + \frac{\hbar \Omega^*}{2} e^{i(\omega t + \phi)} \sigma^- = \frac{\hbar \Omega}{2} e^{-i(\omega t + \phi)} \sigma^+ + \text{H.c.}, \]  

(2.124)

where we omitted the indices of the Rabi frequency \(\Omega = \Omega_{eg}\) and introduced the Pauli operators

\[ \sigma^+ = |e\rangle\langle g|, \quad \sigma^- = |g\rangle\langle e|. \]  

(2.125)

Note that \(\sigma^+ = (\sigma^-)^\dagger\). Let us, for completeness, introduce the Pauli operators \(\sigma^x\) and \(\sigma^y\)

\[ \sigma^x = \sigma^+ + \sigma^-, \quad \sigma^y = -i \sigma^+ + i \sigma^- . \]  

(2.126)

If we choose the representation

\[ |e\rangle \doteq \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \quad |g\rangle \doteq \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \]  

(2.127)

then the Pauli operators have the representation

\[ \sigma^x \doteq \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma^y \doteq \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma^z \doteq \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \]  

(2.128)

2.2.4. Laser-ion interaction in a trap

In this section we will discuss the interaction of trapped ions with externally applied electromagnetic fields. In order to keep the presentation as clear as possible we start by considering a single ion and then extend the analysis to a collection of \(N\) ions that form a Coulomb crystal.

The total Hamiltonian of an ion that interacts with an additionally applied electromagnetic field splits into three parts

\[ H = H_{\text{at}} + H_{\text{m}} + H_{\text{int}}. \]  

(2.129)

The Hamiltonian \(H_{\text{at}}\) describes the internal degrees of freedom, \(H_{\text{m}}\) the motional degrees of freedom and \(H_{\text{int}}\) the interaction with the externally applied field. We assume that the internal
levels can be described as a two-level system such that the internal Hamiltonian is of the form in Eq. (2.121)

\[
H_{\text{int}} = \frac{\hbar \omega_0}{2} \sigma^z.
\] (2.130)

The motional Hamiltonian is given by Eq. (2.57) and for a single ion reads

\[
H = \sum_{\alpha=1,2} \hbar \omega_\alpha a_\alpha^\dagger a_\alpha,
\] (2.131)

where we have once more omitted the ground-state energies. The interaction of the ion with the applied field is of the form given in Eq. (2.124) but now includes the ion’s position \( \mathbf{r} \). In a rotating wave approximation it reads [42]

\[
H_{\text{int}} = \frac{\hbar \Omega}{2} \sigma^+ e^{i(\mathbf{k} \cdot \mathbf{r} + \phi)} + \text{H.c.}
\] (2.132)

The effects of the interaction between the applied electromagnetic field and the ion are most conveniently studied in an interaction picture with respect to the Hamiltonian

\[
H_0 = H_{\text{int}} + H_{\text{int}}.
\] (2.133)

Yet, before we move to the interaction picture we remind ourselves that we can write the ion’s coordinates as \( \mathbf{r}_\alpha(t) = \mathbf{r}_\alpha^0 + \mathbf{q}_\alpha(t) \). For a single ion there is only one motional mode in each spatial direction, which is quantized according to Eq. (2.55). The small oscillations \( \mathbf{q}_\alpha(t) \) can then be written as \( \mathbf{q}_\alpha(t) = q_\alpha \mathbf{e}_\alpha \), where \( \mathbf{e}_\alpha \) is a unit vector in direction \( \alpha \) and

\[
q_\alpha = \sqrt{\frac{\hbar}{2m \omega_\alpha}} (a_\alpha + a_\alpha^\dagger).
\] (2.134)

Inserting this identity into the interaction Hamiltonian in Eq. (2.132) yields

\[
H_{\text{int}} = \hbar \frac{\Omega}{2} \sigma^+ e^{i(\mathbf{k} \cdot \mathbf{r} + \phi)} e^{i \sum_\alpha k_\alpha \sqrt{\frac{\hbar}{2m \omega_\alpha}} (a_\alpha + a_\alpha^\dagger)} e^{-i \omega_\alpha t} + \text{H.c.}
\] (2.135)

where \( k_\alpha = \mathbf{k} \cdot \mathbf{e}_\alpha \) is the component of the wave vector in direction \( \alpha \). For the transformation of the Hamiltonian \( H_{\text{int}} \) to the interaction picture, we use the operator identity [53]

\[
e^{\alpha A} B e^{-\alpha A} = \sum_{n=0}^\infty \frac{\alpha^n}{n!} [A, B]_n,
\] (2.136)

where \( A, B \) are operators, \( \alpha \in \mathbb{C}, [\cdot, \cdot] \) denotes the commutator and \( [A, B]_n = [A, [A, B]_{n-1}] \) with \( [A, B]_0 = B \). Applying the above identity we obtain the following transformations for the spin and mode operators

\[
e^{iH_0^\dagger} \sigma e^{-iH_0^\dagger} = \sigma e^{-i\omega_0 t}, \quad e^{iH_0^\dagger} \sigma^+ e^{-iH_0^\dagger} = \sigma^+ e^{i\omega_0 t},
\] (2.137)

\[
e^{iH_0^\dagger} a_\alpha e^{-iH_0^\dagger} = a_\alpha e^{-i\eta_\alpha t}, \quad e^{iH_0^\dagger} a_\alpha^\dagger e^{-iH_0^\dagger} = a_\alpha^\dagger e^{i\eta_\alpha t}.
\] (2.138)

With the help of the identities in Eqs. (2.137) and (2.138), we obtain the interaction Hamiltonian in an interaction picture with respect to \( H_0 \) as

\[
\hat{H}_{\text{int}} = e^{iH_0^\dagger} H_{\text{int}} e^{-iH_0^\dagger} = \frac{\hbar \Omega}{2} \sigma^+ e^{i(\mathbf{k} \cdot \mathbf{r} + \phi)} e^{i \sum_\alpha \eta_\alpha (a_\alpha e^{-i\eta_\alpha t} + a_\alpha^\dagger e^{i\eta_\alpha t})} e^{-i(\omega_\alpha t)} + \text{H.c.},
\] (2.139)

where we have introduced the Lamb-Dicke factors

\[
\eta_\alpha = k_\alpha \sqrt{\frac{\hbar}{2m \omega_\alpha}}.
\] (2.140)
Let us now inspect the Hamiltonian in Eq. (2.139) in more detail. We follow the treatment in [42]. For simplicity we assume that the applied radiation propagates along one of the spatial coordinates, say \( z \), such that \( k_x = k_y = 0 \). For convenience, we write \( k_z = k \), \( \eta_z = \eta \) and \( a_z^\dagger = a^\dagger \). \( \tilde{H}_{\text{int}} \) then becomes

\[
\tilde{H}_{\text{int}} = \hbar \frac{\Omega}{2} \sigma^+ e^{i(k_z^0 + \phi)} e^{i\eta (a e^{-i\omega t} + a^\dagger e^{i\omega t})} e^{-i\delta t} + \text{H.c.},
\]

(2.141)

where we have also introduced the detuning \( \delta = \omega - \omega_0 \) of the applied radiation. Expanding the exponential including the annihilation and creation operators in terms of \( \eta \) we see that the Hamiltonian contains terms of the form \( \sigma^+ a^l (a^\dagger)^m \), where \( l, m \in \mathbb{N} \), which carry a time-dependence \( e^{i\omega_0 (m-l)t} = e^{i\omega_0 \bar{n}t} \). Note that the Hamiltonian also contains the Hermitian conjugate of these terms. If the detuning \( \delta \) fulfills \( \delta \approx s \omega_c \), then these contributions become resonant and connect states of the form \( |g\rangle |n\rangle \) and \( |e\rangle |n'\rangle \), where \( n' = n + s \). The Rabi frequencies for transitions between these states are given by [42]

\[
\Omega_{n,n'} = \Omega_{n,n'} = \Omega \langle n' | e^{i\eta (a + a^\dagger)} | n \rangle = \Omega e^{-\eta^2/2} \eta |n| \sqrt{n_{<1} \bar{L}_{n,\eta}(\eta^2)}. \tag{2.142}
\]

Here, \( n_{<} \) and \( n_{>} \) stand for the smaller and greater of \( n \) and \( n' \) and \( \bar{L}_{n,\eta}(x) \) is the generalized Laguerre polynomial

\[
\bar{L}_{n,\eta}(x) = \sum_{m=0}^{n} (-1)^m \frac{n + \gamma}{n - m} \frac{x^m}{m!}.
\tag{2.143}
\]

Transitions connecting states with occupation numbers \( n \) and \( n' = n + s \) with a simultaneous excitation or deexcitation of the internal levels are called the \( s \)th red sideband for \( s < 0 \) and the \( |s\rangle \)th blue sideband for \( s > 0 \). In our considerations, as in most quantum information and quantum simulation experiments, only small values of \( n \) and \( s \) are relevant.

The interaction Hamiltonian in Eq. (2.141) and the Rabi frequencies in Eq. (2.142) can be considerably simplified in the so-called \textit{Lamb-Dicke regime}. In the Lamb-Dicke regime the spatial extent of the atomic wave function is much smaller than the wavelength of the applied radiation. This means that at all times we have [42]

\[
\sqrt{\langle k^2 z^2 \rangle} = \eta \sqrt{\langle (a + a^\dagger)^2 \rangle} \ll 1.
\tag{2.144}
\]

If the above Lamb-Dicke criterion is fulfilled, we have \( \eta \ll 1 \). Note, however, that the converse is not necessarily true [39]. If the harmonic oscillator describing the motion is in a thermal state with mean occupation number \( \bar{n} \), the Lamb-Dicke criterion of Eq. (2.144) can also be written as

\[
\eta \sqrt{2\bar{n} + 1} \ll 1.
\tag{2.145}
\]

In the Lamb-Dicke regime, we can approximate the interaction Hamiltonian in Eq. (2.141) to first order in \( \eta \) [42] and obtain

\[
\tilde{H}_{\text{int}} \approx H_{\text{LD}} = \hbar \frac{\Omega}{2} \sigma^+ e^{i(k_z^0 + \phi)} \left[ \mathbb{1} + i\eta (a e^{-i\omega t} + a^\dagger e^{i\omega t}) \right] e^{-i\delta t} + \text{H.c.}.
\tag{2.146}
\]

This simplified Hamiltonian only contains three resonances which are called the carrier, first red sideband and first blue sideband transitions. For \( \delta = 0 \) the first term is resonant and if \( \eta \Omega / \omega_c \ll 1 \) we can neglect the other contributions in a rotating wave approximation. In this case the Hamiltonian takes the form

\[
\tilde{H}_{\text{int}} \approx H_{C} = \hbar \frac{\Omega}{2} e^{i(k_z^0 + \phi)} \sigma^+ + \text{H.c.}.
\tag{2.147}
\]
This Hamiltonian induces transitions between states of the form \(|g⟩|n⟩\) and \(|e⟩|n⟩\) with Rabi frequency \(Ω\). These transitions are called carrier transitions.

For \(δ = −ω\), the second term in the Hamiltonian of Eq. (2.146) is resonant. The resonant part of the Hamiltonian now reads

\[
H_{\text{bsb}} = \hbar \eta \frac{Ω}{2} e^{(i k_F^0 + Φ)} σ^+ a + \text{H.c.}
\]  

(2.148)

The Hamiltonian connects states of the form \(|g⟩|n⟩\) and \(|e⟩|n − 1⟩\) with a Rabi frequency \(\sqrt{2} Ω\) and is called the first blue sideband Hamiltonian. Note that in order to obtain \(H_{\text{bsb}}\) we need to be able to neglect the other contributions in Eq. (2.146). The carrier transition can be neglected in a rotating wave approximation if \(Ω/ω \ll 1\). If this condition is met, then automatically the third contribution in \(H_{LD}\) will also be negligible.

The last resonance is obtained for \(δ = ω\). In this case the resonant part of \(H_{LD}\) in Eq. (2.146) is

\[
H_{\text{bsh}} = \hbar \eta \frac{Ω}{2} e^{(i k_F^0 + Φ)} σ^− a^+ + \text{H.c.}
\]  

(2.149)

This Hamiltonian connects states of the form \(|g⟩|n⟩\) and \(|e⟩|n + 1⟩\) with a Rabi frequency \(\sqrt{n + 1} Ω\). It is called the first red sideband Hamiltonian. Again, in order to obtain \(H_{\text{bsh}}\) we need to be able to neglect the other contributions in Eq. (2.146) with the carrier transition being the dominant contribution. Also, in this case, we can neglect the carrier contribution in a rotating wave approximation if \(Ω/ω \ll 1\), which also renders the red sideband contribution negligible.

It is worth having a brief look at the Lamb-Dicke factor for different types of transitions. In the field of trapped ions, one mostly deals with either optical or microwave frequency transitions, sometimes radio-frequency transition are also considered but we shall neglect these for the moment. Consider, for instance, \(^{25}\text{Mg}^+\) which features a hyperfine structure with electronic ground states with total angular momentum \(F = 2\) and \(F = 3\). For an applied static magnetic field of a few Gauss, the magnetic sublevels are sufficiently split such that one can define a two-level system in the hyperfine ground state manifold \([19, 55]\). One possible choice is the set of states \(|g⟩ \equiv |F = 3, m_F = 3⟩\) and \(|e⟩ \equiv |F = 2, m_F = 2⟩\) with a transition frequency \(Ω_0/2π \approx 1.8\) GHz.

The Lamb-Dicke factor for this transition and a mode frequency \(Ω_m/2π = 1\) MHz is

\[
η_{\text{mw}} \approx 5.4 \cdot 10^{-7}.
\]  

(2.150)

Thus, for a transition in the microwave regime and a plane wave the Lamb-Dicke factor practically vanishes. An optical transition, however, can yield a small but still appreciable Lamb-Dicke factor. \(^{40}\text{Ca}^+\) has an optical transition near 729 nm. For a trapping frequency \(Ω_m/2π = 1\) MHz we obtain a Lamb-Dicke factor

\[
η_L \approx 0.1.
\]  

(2.151)

Accordingly, in many cases optical fields are used to induce spin-motion coupling also for hyperfine and Zeeman qubits. We will discuss a method to do this in Section 2.5. Yet, this does not mean that optical fields are the only option to achieve spin-motion coupling for microwave transitions. In fact, spin-motion coupling can be realized using the magnetic field gradient of the near field of oscillating microwave currents in surface traps \([56, 57]\) or by applying a static magnetic field gradient in addition to microwave radiation \([58, 59]\).

Let us close this section with two remarks. In our derivation of the sideband transition Hamiltonians we have neglected the linewidth of the atomic transitions. In order to implement sideband transitions one needs that the states \(|g⟩\) and \(|e⟩\) have a sufficiently small linewidth such that the sidebands can be resolved. Note, however, that by assuming the states are stable we have implicitly assumed that they have a sharp energy.
Second, higher order sidebands than the first may be driven as well. By continuing the expansion of the interaction Hamiltonian in Eq. (2.141) in powers of \( \eta \) operator terms such as \( \sigma^+ \sigma^- \) or \( \sigma^+ (a^\dagger)^2 \) will appear to second order in \( \eta \). In the Lamb-Dicke regime where \( \eta \ll 1 \) these terms can be made resonant but will in general be weak.

### 2.2.5. Laser-ion interaction for a collection of ions

Let us now move on to consider a collection of \( N \) ions interacting with an additionally applied electromagnetic field. We assume that the ions are of the same species and are sufficiently cold that they form a Coulomb crystal. The internal Hamiltonian now reads

\[
H_{\text{int}} = \sum_{j=1}^{N} \frac{\hbar \Omega_j}{2} \sigma^+_{j \uparrow} e^{i(k \cdot r_j - \omega t + \phi)} + \text{H.c.} \tag{2.154}
\]

Note that the Rabi frequencies \( \Omega_j \) can be different, in general. We also have applied a rotating wave approximation using \( \Omega_j \ll \omega_0 \). As in the previous case, we move to an interaction picture with respect to

\[
H_0 = H_{\text{int}} + H_{\text{m}} \tag{2.155}
\]

to study the interaction of the ions with the applied electromagnetic field. \( H_{\text{int}} \) and \( H_{\text{m}} \) are now given by Eqs. (2.152) and (2.153), respectively. The small excursions \( q_j(t) = r_j(t) - r^0_j \) of the ions around equilibrium according to Eq. (2.28) are given by

\[
q_{j \alpha} = \sum_n B_{jn} \sqrt{\frac{\hbar}{2m \omega_{n,\alpha}}} (a_{n,\alpha} + a_{n,\alpha}^\dagger). \tag{2.156}
\]

Inserting \( r_j(t) = r^0_j + q_j(t) \) into the interaction Hamiltonian in Eq. (2.154) yields

\[
H_{\text{int}} = \sum_{j=1}^{N} \frac{\hbar \Omega_j}{2} \sigma^+_{j \uparrow} e^{i(k \cdot r^0_j + \phi)} e^{i \sum_n B_{jn} \sqrt{\frac{\hbar}{2m \omega_{n,\alpha}}} (a_{n,\alpha} + a_{n,\alpha}^\dagger)} e^{-i \omega t} + \text{H.c.} \tag{2.157}
\]

Moving to an interaction picture with respect to \( H_0 \) in Eq. (2.155) the spin operators \( \sigma^\pm \) and mode operators \( a_{n,\alpha}, a_{n,\alpha}^\dagger \) transform as in Eqs. (2.137) and (2.138) and we obtain

\[
\tilde{H}_{\text{int}} = e^{i \hbar d} H_{\text{int}} e^{-i \hbar d} = \sum_{j=1}^{N} \frac{\hbar \Omega_j}{2} \sigma^+_{j \uparrow} e^{i(k \cdot r^0_j + \phi)} e^{i \sum_n B_{jn} \sqrt{\frac{\hbar}{2m \omega_{n,\alpha}}} (a_{n,\alpha} - i \omega_{n,\alpha} a_{n,\alpha}^\dagger)} e^{-i (\omega t - \omega_0) t} + \text{H.c.}, \tag{2.158}
\]
where we have introduced the Lamb-Dicke factors

\[ \eta_{n,\alpha} = k_\alpha \sqrt{\frac{\hbar}{2m\omega_{n,\alpha}}}. \]  

(2.159)

We now assume that we are in the Lamb-Dicke regime and expand the exponential containing the motional ladder operators to first order in the \( \eta_{n,\alpha} \)

\[
\hat{H}_{\text{int}} \approx \sum_{j=1}^{N} \frac{\hbar}{2} B_{jm} \eta_{n,\alpha} e^{i(kv_j^0 + \phi)} \left[ 1 + i \sum_{n,\alpha} B_{jm} \eta_{n,\alpha} (a_{n,\alpha} e^{-i\omega_{n,\alpha} t} + a_{n,\alpha}^\dagger e^{i\omega_{n,\alpha} t}) \right] e^{-i\delta t} + \text{H.c.} \]  

(2.160)

Here we introduced the detuning of the applied radiation from the carrier transition \( \delta = \omega - \omega_0 \).

Let us now again consider the case where the radiation is directed along one of the spatial directions \( \alpha \). We now assume that we are in the Lamb-Dicke regime and expand the exponential containing the detunings \( \eta_{n,\alpha} \).

\[
\hat{H}_{\text{int}} \approx \sum_{j=1}^{N} iB_{jm} \eta_{n,\alpha} \frac{\Omega_j}{2} e^{i(kv_j^0 + \phi)} \sigma_j^+ a_{n,\alpha} e^{-i\delta_{n,\alpha} t} + \text{H.c.}, \]  

(2.161)

where we have introduced the detunings

\[
\delta_{n,\alpha} = \delta + \omega_{n,\alpha} = \omega - (\omega_0 - \omega_{n,\alpha}). \]  

(2.162)

It is not immediately clear if the Hamiltonian in Eq. (2.161) is amenable to further simplification. If the modes are well-separated in frequency, it is possible that \( \eta_{n,\alpha} \Omega / \delta_{n,\alpha} \ll 1 \) for all \( n \neq m \) such that we can neglect all terms except the one for mode \( m \). Then, the Hamiltonian in Eq. (2.161) becomes the red sideband Hamiltonian for a single mode

\[
\hat{H}_{\text{int}} \approx \hbar \sum_{j} iB_{jm} \eta_{m,\alpha} \frac{\Omega_j}{2} e^{i(kv_j^0 + \phi)} \sigma_j^+ a_{m,\alpha} e^{-i\delta_{m,\alpha} t} + \text{H.c.} \]  

(2.163)

To obtain such a coupling is easier for axial modes. Let us denote a typical distance between ions by \( d \). The ratio between Coulomb and trapping potentials can be estimated to be of the order of \( e^2/(4\pi\varepsilon_0 d^3 m\omega_0^2) \) [33]. Taking the typical separation \( d \approx l_z \) with \( l_z \) from Eq. (2.21) we see that this ratio is of order one for axial modes and the modes are well separated in energy. For the radial modes of an ion string, however, the ratio between Coulomb and trapping potentials is smaller by a factor \( \xi_{\alpha} = \omega_r^2 / \omega_0^2, \alpha = x, y, z \) compared to the axial direction. Also the bandwidth of the phonon branches is smaller, as can be seen from Eqs. (2.43) and (2.48), (2.49). The axial normal mode frequencies are in the range \( \omega_{n,z} \in [\omega_0, \omega_{n,z} \sqrt{\lambda_{N,z}} \omega_0] \) while the radial frequencies are in the range \( \omega_{n,\alpha} \in [\omega_0 \sqrt{1 + \xi_{\alpha} \frac{1 - \chi_{N,z}}{2}}, \omega_\alpha] \). For \( \xi_{\alpha} \ll 1 \), we may expand the square root in the lower limit of the interval, yielding the estimate \( \omega_{\alpha} \in [\omega_\alpha (1 - \frac{\xi_{\alpha} \chi_{N,z}}{4}), \omega_\alpha] \). Thus, we see that for \( \xi_{\alpha} \ll 1 \) the radial modes have a much smaller bandwidth than the axial phonons. This can also be appreciated in Fig. 2.2 above. In this case it can happen that the coupling to several modes is non-negligible such that we cannot simplify the Hamiltonian in Eq. (2.161) further.

### 2.3. Trapped-ion qubits

In many applications the internal structure of trapped ions is described as a two-level system. In the previous section, we have seen how this reduction can be achieved. A quantum mechanical
two-level system is often also referred to as a qubit. The term originates from the field of quantum information processing, where the qubit is the basic unit of information [1].

The reduction of the many internal states of trapped ions to a two-level system is also common in applications other than quantum information processing. The reduction to a two-level system is also used, for example, in frequency standards where one measures a specific atomic transition frequency very precisely [60] or in quantum simulations where trapped ions are often used to simulate spin-1/2 systems [12, 55].

In this section, we discuss possible implementations of qubits with trapped ions and their coherence properties.

2.3.1. Qubit encodings with trapped ions

A popular choice for trapping are alkaline earth metal ions such as Be\(^+\), Mg\(^+\) and Ca\(^+\). After ionization, these ions only possess a single valence electron such that the ground state is of the form \(n^2S_{1/2}\). Here \(n\) is the principal quantum number and we use the notation \(2s+1l_j\), where \(2s+1\) is the spin multiplicity, \(l\) the orbital angular momentum and \(j=l+s\) the total electronic angular momentum.

Since the other, energetically lower lying, electronic shells are fully occupied, the electronic structure of ionized alkaline earth metals is similar to that of hydrogen. However, there is no \(l\) degeneracy because the probability density of finding the valence electron close to the nucleus depends on \(l\). The Coulomb energy due to the interaction with the nucleus and the other electrons therefore varies for different \(l\) and lifts the degeneracy [52]. Now, there are different possibilities to encode a qubit in the energy levels of the valence electron of a trapped ion. These are called optical, hyperfine and Zeeman qubit and we discuss them in the following.

Optical qubits

In optical qubits the two level system is realized by an electronic ground state and a metastable excited state with a transition frequency in the optical regime. One realization of an optical qubit are the \(4^2S_{1/2}\) and the \(4^2D_{5/2}\) states of \(^{40}\text{Ca}^+\) with a transition near 729\,nm. The \(^2D_{5/2}\) state has a lifetime \(T_1 \approx 1.2\,s\) [61]. The state of the two level system is usually read out by using so-called electron shelving [42]. In this method, one uses a third level \(|r\rangle\) such that the transition \(|g\rangle \leftrightarrow |r\rangle\) can be driven independently of the transition \(|g\rangle \leftrightarrow |e\rangle\). The state \(|r\rangle\) should have a short lifetime and decay back directly to the state \(|g\rangle\). Such a transition is called a closed or cycling transition. Now, if the ion is initially in a superposition of states \(|g\rangle\) and \(|e\rangle\) and one applies radiation to drive the transition \(|g\rangle \leftrightarrow |r\rangle\), the ion is projected either to state \(|g\rangle\) or to state \(|e\rangle\) on the first scattering event. If the electron is projected to state \(|g\rangle\), many photons can be scattered from the transition \(|g\rangle \leftrightarrow |r\rangle\), which can then be registered by photomultiplier tubes or CCD cameras [42], while ideally no fluorescence is detected if the system is projected to state \(|e\rangle\). In this manner, one can distinguish the qubit states efficiently. Note that such a cycling transition is also needed for Doppler laser cooling which we will discuss in Section 2.6.1. A generic level structure of an optical qubit is shown in Fig. 2.3 a).

Hyperfine qubits

Hyperfine qubits can be encoded with ions that have a non-vanishing nuclear spin. If the ions possess a nuclear spin \(I\), there are two ground-state hyperfine manifolds with \(F = I + 1/2\) where \(m_F = -(I + 1/2), \ldots, I + 1/2\) and \(F = I − 1/2\) where \(m_F = -(I − 1/2), \ldots, I − 1/2\). The degeneracy of the hyperfine states can be lifted by applying a static magnetic field. The transition frequencies of hyperfine qubits usually lie in the microwave regime \(\omega_0/2\pi \approx 1 − 10\,\text{GHz}\) [61].
2.3. Trapped-ion qubits

a) Figure 2.3.: Level structure for trapped-ion qubits: Panel a) schematically shows an optical qubit where the excited qubit state $|e\rangle$ has a finite lifetime $\tau$ and is connected to the ground state $|g\rangle$ by an optical transition. $|g\rangle$ can be coupled to a fast decaying level $|r\rangle$ for laser cooling and state detection. Decay from state $|r\rangle$ is indicated by the curly lines. Panel b) shows a hyperfine qubit with transition frequency in the microwave regime, where the qubit levels may be coupled by stimulated Raman transitions (arrows to dashed line) or directly by a microwave. Here, $|e\rangle$ can be coupled to a fast decaying level $|r\rangle$ for readout and cooling. Figure derived from [61].

Again one needs a third level for the qubit readout. Typically, this level is connected to one of the qubit states by an optical transition. The generic structure of a hyperfine qubit is shown in Fig. 2.3 b). One example of an ion that allows us to encode a hyperfine qubit is $^{25}\text{Mg}^+ [55]$. It has a nuclear spin $I = 5/2$ such that there are two hyperfine manifolds with $F = 2$ and $F = 3$. One choice of a qubit is $|g\rangle = |F = 3, m_F = 3\rangle$ and $|e\rangle = |F = 2, m_F = 2\rangle$ with a transition frequency of about 1.8 GHz at a magnetic field of a few Gauss and the third level in the $^{3}_2P_{1/2}$ or $^{3}_2P_{3/2}$ manifolds, which are separated in energy from the ground state manifold by about 280 nm.

Zeeman qubits

For atomic species without a nuclear spin there is no hyperfine structure and the $^{n}_2S_{1/2}$ ground state manifold consists of the two $m_j = \pm 1/2$ levels. The degeneracy of the two levels can be lifted by applying a magnetic field leading to the so-called Zeeman qubit. One option to form a Zeeman qubit are the $^{n}_2S_{1/2}$ ground states of $^{40}\text{Ca}^+$ that has no nuclear spin. This type of qubit is often used in Penning traps, which rely on strong static magnetic fields on the order of a few Tesla to confine the ions [62, 63]. Depending on the applied magnetic field, the transition frequencies range from a few MHz [61] to GHz [62, 63].

2.3.2. Qubit coherence times

Let us now turn to the coherence properties of the trapped ion qubits. If we consider an optical qubit with stable state $|g\rangle$ and a metastable state $|e\rangle$, the coherence time $T_2^*$ between superpositions of the two states is limited by the lifetime $T_1$ of the upper state $T_2^* \leq 2T_1 [64]$. Hence, it is not surprising that for optical qubits, the finite lifetime of the metastable state can play a role on experimental time scales [61].

For hyperfine qubits one can make a rough estimate of the decay rate of the qubit levels leading to $\Gamma \approx 10^{-12}\text{s}^{-1} [39]$. Accordingly, the lifetime of hyperfine qubits is practically infinite. The same is true for Zeeman qubits.

In most experimental situations, however, the limiting factor of the coherence times are mag-
netic field fluctuations. They lead to coherence times on the order of milliseconds for all types of qubits \[61\]. Other influences that limit the coherence times are, for example, fluctuating laser intensities, frequencies and phases. In a magnetic field \( B \) with small fluctuations around a desired value \( B_0 \) the shift of the qubit transition frequency usually can be written as \[39\]

\[
\Delta \omega_0 = \left. \frac{\partial \omega_0}{\partial B} \right|_{B_0} (B - B_0) + \frac{1}{2} \left. \frac{\partial^2 \omega_0}{\partial B^2} \right|_{B_0} (B - B_0)^2. \tag{2.164}
\]

In most cases, the first order contributes larger shifts and is thus mostly responsible for dephasing. Hyperfine qubits can be made more robust with respect to fluctuations in the magnetic fields by using transitions between states of the form \( |F, m_F = 0 \rangle \) and \( |F', m_F' = 0 \rangle \), which are insensitive to fluctuations in the magnetic field to first order for \( B = 0 \) \[65\]. For this type of qubit coherence times exceeding 10 minutes have been observed \[66\]. However, the degeneracy of magnetic sublevels at zero magnetic field, hampers the addressability of individual states and it is more advantageous to work with finite fields.

It turns out that there are first-order field insensitive transitions, i.e. transitions where the first term in Eq. (2.164) vanishes, for finite magnetic fields. With such transitions coherence times of \( T_2^* \approx 10 \text{s} \) in \(^9\text{Be}^+\) \[65\] and \( T_2^* \approx 50 \text{s} \) in \(^{43}\text{Ca}^+\) \[67\] have been observed. Magnetic field insensitive transitions are commonly used for atomic frequency standards, see e.g. \[68\], and are therefore dubbed (atomic) clock transitions.

### 2.4. Master equations, the quantum regression theorem and adiabatic elimination

In this section we will introduce two theoretical tools that are frequently used in the following parts of this thesis and in quantum optics in general, namely, (Markovian) master equations and adiabatic elimination.

Master equations are employed to describe the dynamics of open quantum systems. An open quantum system is a system whose time evolution cannot be described by the interactions within the system alone. In order to describe the dynamics of such a system accurately, we have to take into account the effects of the surroundings on the system’s dynamics. Under certain conditions it is possible to derive closed dynamical equations for an open quantum system. Here, we will discuss one such situation, which is the limit of weak coupling between the system and its environment, where a Markovian master equation can be derived. We will also discuss a result called the quantum regression theorem. This is a method to calculate correlation functions of operators of an open quantum system whose time evolution is described by a Markovian master equation. Finally, we will briefly allude to non-Markovian master equations which describe a wider class of open quantum systems.

Adiabatic elimination, on the other hand, is a procedure to find an effective dynamics for a system by eliminating one part of the system from the dynamics. This can greatly simplify the description of the dynamics of a system and in that sense it is not much different from master equations.

This section is organized as follows. We start with the description of open quantum systems and present the derivation of a Markovian master equation. Thereafter, we introduce the quantum regression theorem and then throw a glance at the more general case of non-Markovian master equations. We close the section with a brief account of adiabatic elimination and illustrate the procedure with an example.
2.4. Master equations, the quantum regression theorem and adiabatic elimination

2.4.1. Standard form of the Markovian master equations

For a closed quantum system the time evolution of the system is governed by the Schrödinger equation

\[ i\hbar \frac{d}{dt} |\psi(t)\rangle = H(t) |\psi(t)\rangle, \]

(2.165)

where \( |\psi(t)\rangle \) is the state of the system at a given time and \( H(t) \) the system’s Hamiltonian. If we have incomplete knowledge about the system’s state, which is usually the case, it is more convenient to work with the density matrix \( \rho(t) \) of the system [49]. If, for example, we only know that the system is in a certain state \( |\psi_i\rangle \) with probability \( p_i \geq 0 \), \( \sum_i p_i = 1 \), a more convenient description of the state of the system is the density matrix

\[ \rho = \sum_i p_i |\psi_i\rangle \langle \psi_i|. \]

(2.166)

The density matrix has the following properties [49]: It is Hermitian \( \rho = \rho^\dagger \), has unit trace \( \text{Tr} \rho = 1 \) and is positive semidefinite \( \rho \geq 0 \).

Using the adjoint of the Schrödinger equation (2.165) one obtains the equation of motion for the density matrix

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

(2.167)

which is also known as the Liouville–von-Neumann equation.

Let us now turn to open quantum systems. For an open quantum system the effects of the environment on the system’s dynamics cannot be neglected leading to a time evolution different from Eq. (2.167). In order to derive an equation of motion for the open quantum system, one usually starts by considering an enlarged system consisting of the system of interest and its environment. The Hamiltonian of the complete system splits into three parts

\[ H = H_S + H_E + H_I, \]

(2.168)

where \( H_S \) is the Hamiltonian of the free evolution of the system of interest and \( H_E \) the free Hamiltonian of the environment. The third part \( H_I \) describes the interaction between the system of interest and its environment.

Our goal now is to obtain an equation of motion for the state of the system of interest, sometimes also referred to as the principal or reduced system. In order to arrive at such equation we now assume that the full system, consisting of the principal system and its environment, can be regarded as a closed system and with its density matrix \( \rho(t) \) evolving according to the Liouville–von-Neumann equation (2.167). Accordingly, for an initial state \( \rho(0) \) of the combined system, the state at a later time \( t \) reads

\[ \rho(t) = U(t,0)\rho(0)U^\dagger(t,0), \]

(2.169)

where \( U(t,0) = e^{-iHt} \) is the unitary time evolution operator of the full system. The state of the reduced system \( \rho_S(t) \) is obtained from \( \rho(t) \) by performing a partial trace over the environmental degrees of freedom [1] indicated by a subscript E

\[ \rho_S(t) = \text{Tr}_E \rho(t) = \text{Tr}_E[U(t,0)\rho(0)U^\dagger(t,0)]. \]

(2.170)

Let us now assume that the principal system and its environment are initially uncorrelated, i.e. in a product state, such that we may write the initial state as

\[ \rho(0) = \rho_S(0) \otimes \rho_E(0) \]

(2.171)
with a fixed state of the environment $\rho_E(0)$, i.e. the same for any $\rho_S(0)$. Here $\rho_S(0)$ and $\rho_E(0)$ are the initial states of system and environment, respectively. Since $\rho_E(0)$ is Hermitian and positive semidefinite, we can write it in diagonal form

$$\rho_E(0) = \sum_j \lambda_j |e_j\rangle \langle e_j|,$$

where the $\lambda_j \geq 0$, $\sum_j \lambda_j = 1$ and $\langle e_j| e_k \rangle = \delta_{jk}$. The index $j$ runs over all environmental degrees of freedom which, in principle, can be infinitely many. Inserting $\rho(0)$ from Eq. (2.171) into the right-hand side of Eq. (2.170) and using Eq. (2.172) we arrive at

$$\rho_S(t) = \sum_{j,k} K_{jk}(t) \rho_S(0) K_{jk}^\dagger(t),$$

where we have introduced the so-called Kraus operators

$$K_{jk}(t) = \sqrt{\lambda_j} |e_k\rangle \langle U(t,0)|e_j\rangle.$$

The Kraus operators satisfy the condition

$$\sum_{j,k} K_{jk}^\dagger(t) K_{jk}(t) = 1,$$

as can be verified using their definition in Eq. (2.174).

Considering Eq. (2.173) we can define a map $\mathcal{E}(t,0)$ that maps the initial state of the reduced system $\rho_S(0)$ to another state at time $t$ according to

$$\rho_S(t) = \mathcal{E}(t,0) \rho_S(0) = \sum_{j,k} K_{jk}(t) \rho_S(0) K_{jk}^\dagger(t).$$

The map $\mathcal{E}(t,0)$ is called a dynamical map [69]. More mathematically speaking for fixed $t$ $\mathcal{E}(t,0)$ is a map from the set of operators on the Hilbert space of the reduced system into itself. Let us briefly consider the properties that we expect for such a map.

The dynamical map should map density matrices to density matrices. It is a crucial feature of a density matrix that it has a unit trace and, therefore, we expect the dynamical map to be trace preserving. Furthermore, the density matrix is positive semidefinite and accordingly the dynamical map should preserve the non-negative character of the density matrix. Thus, we demand positivity of the map. $\mathcal{E}(t,0)$ should also be a convex-linear map on the set of density matrices [1], i.e. for a set of probabilities and states $\{p_i, \rho_i\}$, $\sum_i p_i = 1$ we have

$$\mathcal{E} \left( \sum_i p_i \rho_i \right) = \sum_i p_i \mathcal{E}(\rho_i).$$

Finally, it turns out that demanding positivity of the map is not sufficient. Let us consider an $n$-level system with Hamiltonian $H = 0$ far away from the open system such that the two systems do not interact. The time evolution of the full system in their combined Hilbert space is then given by $\mathcal{E}(t,0) \otimes \mathbb{1}$ [70]. Accordingly, the map $\mathcal{E}(t,0) \otimes \mathbb{1}$ has to be positive for all values of $n$. This property is called complete positivity. Summarizing the above considerations, a dynamical map $\mathcal{E}(t,0)$ should have the properties of being

1. trace preserving
2. convex-linear
3. completely positive.

It can be shown that a dynamical map has a Kraus representation as in Eq. (2.173) if and only if it satisfies the three conditions above [1].

If we allow for other values of \( t \), the set \( \{ \mathcal{E}(t_0) | t \geq 0 \} \) constitutes a one-parameter family of dynamical maps. The dynamical maps can be represented by a generator \( \mathcal{L} \) if they satisfy the two following conditions [70]: the map satisfies the semigroup property

\[
\mathcal{E}(t_2,0)\mathcal{E}(t_1,0) = \mathcal{E}(t_2+t_1,0),
\]

and \( \text{Tr}[\mathcal{E}(t,0)\rho A] \) is a continuous function of \( t \) for every \( \rho \) from the set of density matrices and for any bounded linear operator \( A \) acting on the system’s Hilbert space. We then have that [70]

\[
\frac{d}{dt}\rho_S(t) = \mathcal{L}\rho_S(t),
\]

where \( \rho_S(t) = \mathcal{E}(t,0)\rho_S(0) \). Hence, the dynamical map can be represented as

\[
\mathcal{E}(t_2,t_1) = e^{\mathcal{L}(t_2-t_1)},
\]

where we assume that the generator \( \mathcal{L} \) is bounded and we are dealing with a finite dimensional system. Note that the semigroup property, sometimes also called Markov property, can be understood physically as the environment being memoryless. The time evolution does not depend on a particular point in time or on the history of the time evolution. This point should become clearer in the following section.

Starting from Eq. (2.179) and using Eq. (2.173) one can derive the “diagonal” standard form of the generator of completely positive (trace preserving) semigroups [69,70]

\[
\mathcal{L}\rho_S(t) = -\frac{i}{\hbar}[\mathcal{H}_S,\rho_S(t)] + \sum_k \gamma_k \left( A_k\rho_S(t)A_k^\dagger - \frac{1}{2}A_k^\dagger A_k\rho_S(t) - \frac{1}{2}\rho_S(t)A_k^\dagger A_k\right).
\]

The first part on the right-hand side describes the unitary part of the evolution due to a Hamiltonian \( \mathcal{H}_S \). Note that the Hamiltonian \( \mathcal{H}_S \), in general, does not coincide with the Hamiltonian \( H_S \), because \( \mathcal{H}_S \) may include additional contributions due to the interaction with the environment [69]. The operators \( A_k \) appearing in the second part are usually termed Lindblad or jump operators and the \( \gamma_k \) are positive rates. For a finite dimensional system of dimension \( N \), there can be at most \( N^2 \) Lindblad operators. The second part can account for dissipation and dephasing processes and is therefore called the dissipator. We denote it by \( \mathcal{D} \) such that

\[
\mathcal{D}\rho_S(t) = \sum_k \gamma_k \left( A_k\rho_S(t)A_k^\dagger - \frac{1}{2}A_k^\dagger A_k\rho_S(t) - \frac{1}{2}\rho_S(t)A_k^\dagger A_k\right).
\]

The generator \( \mathcal{L} \) is often called Liouvillian. Using the dissipator we may write it as

\[
\mathcal{L}\rho_S(t) = -\frac{i}{\hbar}[\mathcal{H}_S,\rho_S(t)] + \mathcal{D}\rho_S(t).
\]

Let us finally remark that the Lindblad operators are formally derived from the time evolution of the full system consisting of the principal system and its environment [69] which is not known in the vast majority of cases. In a few cases the Lindblad operators can be derived from a microscopic model for the full dynamics of system and environment but in most cases they are chosen phenomenologically [69].
2.4.2. Master equation in the Born-Markov approximation

It is in general not the case that a family of dynamical maps describing the evolution of an open quantum system constitutes a semigroup dynamics. In this section, we consider one case, the weak coupling limit, where the derivation of a semigroup generator as in Eq. (2.181) is possible. It is an instructive example to see which approximations are needed to arrive at the semigroup dynamics. This example is also illustrative to make the connection between the semigroup property and Markovian behavior. We follow the treatment in [69].

Let us start from the full Hamiltonian of system plus environment

\[ H = H_S + H_E + H_I. \]  

(2.184)

Moving to an interaction picture with respect to \( H_S + H_E \) the Liouville–von-Neumann equation (2.167) takes the form

\[ \frac{d}{dt} \rho_I(t) = -\frac{i}{\hbar} [H_I(t), \rho_I(t)], \]  

(2.185)

where \( H_I(t) \) and \( \rho_I(t) \) are the transformed interaction Hamiltonian and density matrix. For clarity of notation we omit the index for the state in the following. We look for an equation of motion for the reduced density matrix

\[ \rho_S(t) = \text{Tr}_E \rho(t). \]  

(2.186)

Formal integration of Eq. (2.185) yields

\[ \rho(t) = \rho(0) - \frac{i}{\hbar} \int_0^t dt_1 [H_I(t_1), \rho(t_1)]. \]  

(2.187)

Inserting the above integral form into the right-hand side of Eq. (2.185) and performing a partial trace over the environmental degrees of freedom we obtain

\[ \frac{d}{dt} \rho_S(t) = -\frac{1}{\hbar^2} \int_0^t dt_1 \text{Tr}_E [H_I(t_1), [H_I(t_1), \rho_S(t_1)]]]. \]  

(2.188)

Here, we assumed that \( \text{Tr}_E[H_I(t), \rho(0)] \) vanishes. Note that the right-hand side of the above equation still contains the state \( \rho(t_1) \) of the full system.

We will now perform a first approximation called the Born approximation. Assuming that the environment is much larger than the system and that its coupling to the system is weak, one expects that the state of the environment is only negligibly affected by the interaction with the system. Accordingly, we approximate \( \rho(t) \) as

\[ \rho(t) \approx \rho_S(t) \otimes \rho_E(0). \]  

(2.189)

The above form for the combined state of system and environment might lead one to the conclusion that at no point in time are there excitations in the bath. This is, however, not the case. It means that we assume that excitations in the environment have already decayed on the time scale of our interest and these effects are not resolved. Together with the Markov approximation we introduce below, we obtain a description of the dynamics on a “coarse-grained” time scale [69].

In the Born approximation Eq. (2.188) becomes

\[ \frac{d}{dt} \rho_S(t) = -\frac{1}{\hbar^2} \int_0^t dt_1 \text{Tr}_E [H_I(t_1), [H_I(t_1), \rho_S(t_1) \otimes \rho_E(0)]]]. \]  

(2.190)

We now perform the Markov approximation. The evolution of \( \rho_S(t) \) still depends on its history through \( \rho_S(t_1) \). The first step of the Markov approximation is to substitute \( \rho_S(t_1) \) by \( \rho_S(t) \). The evolution of the system then only depends on its present state. The resulting master equation

\[ \frac{d}{dt} \rho_S(t) = -\frac{1}{\hbar^2} \int_0^t dt_1 \text{Tr}_E [H_I(t_1), [H_I(t_1), \rho_S(t) \otimes \rho_E(0)]]]] \]  

(2.191)
is called the Redfield equation [69].

In order to achieve a Markovian equation that actually leads to a semigroup dynamics we substitute \( t_1 \) by \( t - t_1 \) in the above equation and extend the upper integration limit to infinity. We are interested in the dynamics on a typical time scale of the system’s evolution which we call \( \tau_S \). The above substitution is justified if the integrand on the right-hand side of Eq. (2.191), which turns out to be proportional to the two-time correlation functions of the environment, disappears sufficiently quickly for \( t_1 \gg \tau_E \). Here \( \tau_E \) is a typical time constant for the decay of correlations in the environment and \( t_1 \) a typical time scale of the evolution we are interested in. The Markov approximation is therefore justified if the typical time scale \( \tau_S \) of the system’s evolution is much larger than the time scale of the decay of the environmental correlations \( \tau_E \) [69]. This is why the Markovian master equation is said to provide a description on a “coarse-grained” time axis. The resulting equation reads

\[
\frac{d}{dt} \rho_S(t) = -\frac{1}{\hbar^2} \int_0^\infty dt_1 \text{Tr}_E[H_t(t), [H_t(t-t_1), \rho_S(t) \otimes \rho_E(0)]].
\]  

(2.192)

Note that the Born approximation already contains a subtle version of this separation of time scales. Over a typical time scale of the system evolution \( \tau_S \) the environmental state is taken to have returned to its initial state, which also presupposes \( \tau_E \ll \tau_S \).

The approximations that we performed so far together are termed the Born-Markov approximation. These approximations, however, do not guarantee that one obtains a completely positive semigroup dynamics [69]. In order to obtain a semigroup dynamics a further approximation called the secular approximation is needed. It is, in essence, a rotating wave approximation. In the remainder of this section we will study the secular approximation.

Let us consider that \( H_S \) has a discrete non-degenerate spectrum such that we may define projectors \( \Pi(\varepsilon) \) onto the subspace of energy \( \varepsilon \). The interaction Hamiltonian can be written in the form

\[
H_t = \sum_j A_j \otimes B_j,
\]  

(2.193)

where \( A_j = A_j^\dagger \) and \( B_j = B_j^\dagger \) are Hermitian operators on the system and environment Hilbert spaces, respectively. Using the projectors \( \Pi(\varepsilon) \) we can define the operators

\[
A_j(\omega) = \sum_{\varepsilon' - \varepsilon = \hbar \omega} \Pi(\varepsilon) A_j(\varepsilon'),
\]  

(2.194)

where the sum extends over all \( \varepsilon \) and \( \varepsilon' \) that yield \( \varepsilon' - \varepsilon = \hbar \omega \) for fixed \( \omega \). Obviously, we have

\[
A_j = \sum_\omega A_j(\omega) = \sum_\omega A_j^\dagger(\omega).
\]  

(2.195)

Using the definition of the \( A_j(\omega) \) it is easy to see that they obey the following commutation relations with the principal system’s Hamiltonian

\[
[H_S, A_j(\omega)] = -\omega A_j(\omega), \quad [H_S, A_j^\dagger(\omega)] = \omega A_j^\dagger(\omega).
\]  

(2.196)

Using the above identity together with Eq. (2.136) we obtain the operators \( A_j(\omega) \) and \( A_j^\dagger(\omega) \) in an interaction picture

\[
e^{\frac{i}{\hbar} H_st} A_j(\omega) e^{-\frac{i}{\hbar} H_st} = A_j(\omega) e^{-i \omega t},
\]  

(2.197)

\[
e^{\frac{i}{\hbar} H_st} A_j^\dagger(\omega) e^{-\frac{i}{\hbar} H_st} = A_j^\dagger(\omega) e^{i \omega t}.
\]  

(2.198)
The Hamiltonian may thus be written as

\[ H_I(t) = \sum_{j, \omega} A_j(\omega)e^{-i\omega t} \otimes B_j(t) = \sum_{j, \omega} A^\dagger_j(\omega)e^{i\omega t} \otimes B^\dagger_j(t), \]  

(2.199)

where we have introduced the interaction picture reservoir operators

\[ B_j(t) = e^{iH_0t}B_j e^{-iH_0t}. \]  

(2.200)

Inserting the interaction picture Hamiltonian Eq. (2.199) into Eq. (2.192) yields [69]

\[ \frac{d}{dt}\rho_S(t) = \frac{1}{\hbar^2} \sum_{j, k, \omega, \omega'} \left[ \Gamma_{jk}(\omega) \right] \left[ A_k(\omega)\rho_S(t)A^\dagger_j(\omega') - A^\dagger_j(\omega')A_k(\omega)\rho_S(t) \right] + \text{H.c.} \]  

(2.201)

Here, we have defined the one-sided Fourier transforms

\[ \Gamma_{jk}(\omega) = \frac{1}{\hbar^2} \sum_{j, k, \omega, \omega'} \left[ \Gamma_{jk}(\omega) \right] A_k(\omega)\rho_S(t)A^\dagger_j(\omega') \]  

(2.202)

of the reservoir correlation functions

\[ \left< B^\dagger_j(t)B_k(t-t_1) \right> = \text{Tr}_E \left( B^\dagger_j(t)B_k(t-t_1)\rho_{E}(0) \right). \]  

(2.203)

In many cases \( \rho_E(0) \) is a thermal state of the reservoir

\[ \rho_E(0) = \rho_B = \frac{e^{-\beta H_E}}{\text{Tr}e^{-\beta H_E}} \]  

(2.205)

at some inverse temperature \( \beta = 1/(k_B T) \), where \( T \) is the temperature and \( k_B \) the Boltzmann constant. In this case \( [\rho_E, H_E] = 0 \) and the correlation functions become independent of \( t \)

\[ \left< B^\dagger_j(t)B_k(t-t_1) \right> = \left< B^\dagger_j(t_1)B_k(0) \right>. \]  

(2.206)

Obviously, in this case the rates \( \Gamma_{jk}(\omega) \) are time-independent. Now, if \( \Gamma_{jk} \ll \omega - \omega' \) for all \( \omega \) and \( \omega' \) we may neglect the contributions with \( \omega \neq \omega' \) in a rotating wave approximation such that we arrive at

\[ \frac{d}{dt}\rho_S(t) = \sum_{j, k, \omega} \Gamma_{jk}(\omega) \left[ A_k(\omega)\rho_S(t)A^\dagger_j(\omega) - A^\dagger_j(\omega)A_k(\omega)\rho_S(t) \right] + \text{H.c.} \]  

(2.207)

Further manipulations of the above equation allow us to cast the master equation into the form [69]

\[ \frac{d}{dt}\rho_S(t) = -\frac{i}{\hbar}[H_{LS}, \rho_S(t)] + \mathcal{D}\rho_S(t). \]  

(2.208)

The Hamiltonian \( H_{LS} \) is called the Lamb shift and takes the form [69]

\[ H_{LS} = \sum_{j, k, \omega} S_{jk}(\omega)A^\dagger_j(\omega)A_k(\omega), \]  

(2.209)

where \( S_{jk} = [\Gamma_{jk}(\omega) + \Gamma_{kj}(\omega)]/(2i) \). Using the properties of the \( A_j(\omega) \) it can readily be seen that \( [H_{LS}, H_S] = 0 \). Thus, \( H_{LS} \) will also appear in the Schrödinger picture. It represents the
additional contributions to the Hamiltonian $\bar{H}_S$ in Eq. (2.181) above. The dissipator on the other hand takes the form [69]

$$\mathcal{D}\rho_S(t) = \sum_{j,k} \gamma_{jk}(\omega) \left[ A_k(\omega)\rho_S(t)A_j^\dagger(\omega) - \frac{1}{2} \{A_j^\dagger(\omega)A_k(\omega),\rho_S(t)\} \right],$$

(2.210)

where $\gamma_{jk}(\omega) = \Gamma_{jk}(\omega) + \Gamma_{kj}(\omega)$ and $\{\cdot,\cdot\}$ denotes the anti-commutator. The above equation can be brought into the standard form of the Markovian master equation given in Eq. (2.181), which represents a semigroup evolution [69].

Let us remark in closing this section that environments are usually modeled by collections of harmonic oscillators. The operators $B_j$ in Eq. (2.193) will therefore be given by linear combinations of mode operators and the environmental correlation functions in Eq. (2.204) will contain mode operators or linear combinations thereof. If these involve a finite number of frequencies, the functions will decay and have a revival after a finite revival time $\tau_{\text{rev}}$. Truly decaying functions can only be obtained for environments with a continuum of frequencies, i.e. for infinite environments [69].

2.4. Master equations, the quantum regression theorem and adiabatic elimination

In the previous sections we have seen the standard form of the Markovian master equation and a derivation of a Markovian master equation in the weak coupling limit. Once we are equipped with the master equation, we can compute the time evolution of the reduced system and also expectation values of operators at different times. But often one is also interested in obtaining correlation functions, which amounts to evaluating the expectation value of products of operators with different time arguments. From such calculations one can obtain, for example, the spectrum of spontaneous emission of a two-level system or the spectrum of a field radiated by a driven cavity [71]. In this section we will introduce a means of computing two-time correlation functions of mode operators and the environmental correlation functions in Eq. (2.204) will contain harmonic oscillators. The operators $B_j$ in Eq. (2.193) will therefore be given by linear combinations of mode operators and the environmental correlation functions in Eq. (2.204) will contain mode operators or linear combinations thereof. If these involve a finite number of frequencies, the functions will decay and have a revival after a finite revival time $\tau_{\text{rev}}$. Truly decaying functions can only be obtained for environments with a continuum of frequencies, i.e. for infinite environments [69].

2.4.3. The quantum regression theorem

In the previous sections we have seen the standard form of the Markovian master equation and a derivation of a Markovian master equation in the weak coupling limit. Once we are equipped with the master equation, we can compute the time evolution of the reduced system and also expectation values of operators at different times. But often one is also interested in obtaining correlation functions, which amounts to evaluating the expectation value of products of operators with different time arguments. From such calculations one can obtain, for example, the spectrum of spontaneous emission of a two-level system or the spectrum of a field radiated by a driven cavity [71]. In this section we will introduce a means of computing two-time correlation functions of operators of an open quantum system whose time evolution is described by a Markovian master equation in Lindblad form. This result is known as the quantum regression theorem and was originally developed in [72, 73]. We will follow the treatment in [71].

Let us consider an open quantum system with Hamiltonian $H = H_S + H_E + H_I$ as in Eq. (2.168) above, whose time evolution is described by a generic Liouvillian $\mathcal{L}$ derived in the weak coupling limit

$$\frac{d}{dt} \rho_S(t) = \mathcal{L}\rho_S(t).$$

(2.211)

We move to the Heisenberg picture to compute the correlation functions. Let us consider two operators $F_1$ and $F_2$ of the principal system. In the Heisenberg picture the operators become time dependent and read

$$F_j(t) = e^{i\bar{H}t} F_j e^{-i\bar{H}t}, \quad j = 1, 2.$$  

(2.212)

The correlation function between the two operators, i.e. the multi-time average of the operators, is given by

$$\langle F_1(t_1)F_2(t_2) \rangle = \text{Tr}[F_1(t_1)F_2(t_2)\rho(0)],$$

(2.213)

where $\rho(0)$ is the initial state of the full system comprising principal system and environment and the trace is also taken over system and environment. Note that we may write the initial state in terms of the state of the system at a later time $t$ as

$$\rho(0) = e^{i\bar{H}t} \rho(t)e^{-i\bar{H}t}.$$  

(2.214)

Using the above property for $t = t_1$, Eq. (2.212) and the cyclic property of the trace we can write the two-time average of the operators as

$$\langle F_1(t_1)F_2(t_2) \rangle = \text{Tr}_S \{ F_2(0) \text{Tr}_E [ e^{-i\bar{H}(t_2-t_1)} \rho(t_1) F_1(0) e^{i\bar{H}(t_2-t_1)} ] \}.$$  

(2.215)
We now assume that \( t_2 \geq t_1 \) such that \( t_2 - t_1 = \tau \geq 0 \) and define the operator
\[
\chi(\tau) = e^{-\frac{i}{\hbar}H\tau} \rho(t_1) F_1(0) e^{\frac{i}{\hbar}H\tau},
\] (2.216)
which is the term in angular brackets in Eq. (2.215). The new operator satisfies the equation of motion
\[
\frac{d}{d\tau} \chi(\tau) = -\frac{i}{\hbar} [H, \chi(\tau)]
\] (2.217)
with the initial condition
\[
\chi(0) = \rho(t_1) F_1(0).
\] (2.218)
In order to eliminate the explicit appearance of the reservoir in Eq. (2.215) we need to perform the trace over the environment and define
\[
\chi_S(\tau) = \text{Tr}_E[\chi(\tau)].
\] (2.219)
For \( \tau = 0 \) we obtain
\[
\chi_S(0) = \text{Tr}_E[\rho(t_1) F_1(0)] = \rho_S(t_1) F_1(0),
\] (2.220)
where \( \rho_S(t_1) \) is the state of the reduced system at time \( t_1 \). Now, we make the assumption of weak coupling between system and environment and make a Born approximation assuming that
\[
\rho(t_1) = \rho_S(t_1) \otimes \rho_E(0).
\]
Inserting this product state into the initial condition Eq. (2.218) and using Eq. (2.220) we obtain
\[
\chi(0) = \chi_S(0) \otimes \rho_E(0).
\] (2.221)
Now, Eqs. (2.217), (2.219) and (2.221) are completely analogous to Eqs (2.185), (2.186) and (2.189) which formed the starting point for the derivation of the master equation in the Born-Markov approximation. Accordingly, following the procedure in the previous chapter one can derive a weak coupling master equation for the operator \( \chi(\tau) \). The Hamiltonian appearing in Eq. (2.217) is the same as the one that dictates the dynamics of the full system. Hence, we will obtain the same Liouvillian for the evolution of \( \chi(\tau) \) as for \( \rho_S(t) \) given in Eq. (2.211) and the evolution of \( \chi_S(\tau) \) obeys the equation of motion [71]
\[
\frac{d}{d\tau} \chi_S(\tau) = \mathcal{L} \chi_S(\tau).
\] (2.222)
The solution of the above equation is
\[
\chi_S(\tau) = e^{\mathcal{L} \tau} \chi_S(0) = e^{\mathcal{L} \tau} [\rho_S(t_1) F_1(0)].
\] (2.223)
Inserting the above result back into Eq. (2.215) yields the final result
\[
\langle F_1(t) F_2(t + \tau) \rangle = \text{Tr}_S \{ F_2(0) e^{\mathcal{L} \tau} [\rho_S(t) F_1(0)] \},
\] (2.224)
where we set \( t_1 = t \). Similarly one obtains [71]
\[
\langle F_1(t + \tau) F_2(t) \rangle = \text{Tr}_S \{ F_1(0) e^{\mathcal{L} \tau} [F_2(0) \rho_S(t)] \}.
\] (2.225)
One is often interested in stationary state correlation functions. Assuming that the asymptotic state is a thermal equilibrium state \( \lim_{\tau \to \infty} \rho_S(t) = \rho_\beta \), we have
\[
\lim_{\tau \to \infty} \langle F_1(t + \tau) F_2(t) \rangle = \langle F_1(\tau) F_2(0) \rangle_\beta = \text{Tr}_S \{ F_1(0) e^{\mathcal{L} \tau} [F_2(0) \rho_\beta] \}.
\] (2.226)
Equations (2.224) and (2.225) are the first formulation of the quantum regression theorem that we present in this section. With these equations, we have found expressions to compute correlation functions for the case when the dynamics of a system is described by a weak coupling.
master equation. One should note though that the above equations are usually as hard to solve as the Liouville–von-Neumann equation. We will now obtain another form to compute the two-time correlation functions if the time evolution is given by a time independent Liouvillian as in Eq. (2.211).

Let us assume that there exists a set of operators \( A_j, j = 1, 2, \ldots \) in the Hilbert space of the open quantum system that is complete in the sense that for an arbitrary operator \( F \) in the open system’s Hilbert space [71]

\[
\text{Tr}_S[A_j \mathcal{L} F] = \sum_k M_{jk} \text{Tr}_S[A_k F],
\]

(2.227)

where the \( M_{jk} \) are constants. It follows from the above equation that we obtain a closed system of equations for the time evolution of the expectation values \( \langle A_j(t) \rangle = \text{Tr}_S[A_j \rho(t)] \)

\[
\frac{d}{dt} \langle A_j(t) \rangle = \text{Tr}_S[A_j \frac{d}{dt} \rho(t)] = \text{Tr}_S[A_j \mathcal{L} \rho(t)] = \sum_k M_{jk} \text{Tr}_S[A_k \rho(t)] = \sum_k M_{jk} \langle A_k(t) \rangle.
\]

(2.228)

Writing the operators \( A_j \) as a vector \( \mathbf{A} = (A_1, A_2, \ldots)^\top \) we can write the above equation as a matrix equation

\[
\frac{d}{dt} \langle \mathbf{A}(t) \rangle = \mathbf{M} \langle \mathbf{A}(t) \rangle.
\]

(2.229)

Let us now consider the correlation function \( \langle F(t)A_j(t + \tau) \rangle, \tau \geq 0 \). Differentiating with respect to \( \tau \) yields

\[
\frac{d}{d\tau} \langle F(t)A_j(t + \tau) \rangle = \text{Tr}_S[A_j(0) \mathcal{L} e^{\mathcal{L} \tau} [\rho_S(t)F(0)]]
\]

\[
= \sum_k M_{jk} \text{Tr}_S[A_k(0)e^{\mathcal{L} \tau} [\rho_S(t)F(0)]]
\]

\[
= \sum_k M_{jk} \langle F(t)A_k(t + \tau) \rangle,
\]

(2.230)

where we have used Eq. (2.224) in the first and last steps and Eq. (2.228) in the second step. We may also write the above equation as

\[
\frac{d}{d\tau} \langle F(t)\mathbf{A}(t + \tau) \rangle = \mathbf{M} \langle F(t)\mathbf{A}(t + \tau) \rangle,
\]

(2.231)

which is the second formulation of the quantum regression theorem. Similarly, one obtains [71]

\[
\frac{d}{d\tau} \langle \mathbf{A}(t + \tau)F(t) \rangle = \mathbf{M} \langle \mathbf{A}(t + \tau)F(t) \rangle,
\]

(2.232)

where \( \tau > 0 \). This form of the regression theorem tells us that the correlation functions obey the same equations of motion as the mean values do. This is a rather surprising result and one would not expect that it is generally true. This expectation is actually correct [74]. Of course, this is because the master equation itself is an approximate treatment and one can only expect it to yield quantitatively valid predictions in its domain of applicability. In Chapter 4 we will discuss in more detail when the regression theorem is applicable.

Finally, one might expect that the second form of the regression theorem in Eqs. (2.231) and (2.232) can only rarely be applied, because it relies on the existence of a set of operators \( A_j \) that satisfies Eq. (2.228). It can be shown, though, that this set of operators always exists if there is a discrete basis \(|n\rangle, n \in \{1, \ldots, N\}\) of the open system’s Hilbert space [71]. Then, the operators \( A_j = A_{nm} = |n\rangle\langle m|, n, m \in \{1, \ldots, N\}\) constitute a set of operators satisfying Eq. (2.228). Note that \( N \) might even be infinite here.
2.4.4. Time-local master equations with memory effects

In the previous sections we have considered the Markovian master equation which is a time local differential equation that allows us to compute the evolution of a quantum system. An essential (but not the only) ingredient for the derivation of the Markovian master equation was that, loosely speaking, the environment relaxes on a time scale that is much faster than the open system’s time evolution and excitations that have entered the environment once do not act back on the system. Although Markovian master equations can be applied to a variety of phenomena, they do not cover a large class of open system dynamics, namely those where there are memory effects and the system’s dynamics depends on its prior evolution.

At first sight one would expect that a differential equation that incorporates memory effects is non-local in time including some memory kernel which includes the past evolution of the system. Indeed, such an equation of motion can be derived from the Liouville–von-Neumann equation (2.185) and is called the Nakajima-Zwanzig equation [69]. The Nakajima-Zwanzig equation is exact and is usually as hard to solve as the Liouville–von-Neumann equation itself.

From a computational point of view it is, however, desirable to obtain a time-local master equation. The following argument [75] illustrates that even in the presence of memory effects it should be possible to obtain a time-local differential equation for the evolution of a quantum system. According to Eq. (2.176) the time-evolved state of the open system reads \( \rho_S(t) = \hat{\gamma}(t,0)\rho_S(0) \). Assuming that the map \( \hat{\gamma}(t,0) \) is differentiable with respect to \( t \) we obtain

\[
\frac{d}{dt} \rho_S(t) = \left[ \frac{d}{dt} \hat{\gamma}(t,0) \right] \rho_S(0) = \hat{\gamma}(t,0)\rho_S(0).
\]

(2.233)

Assuming the inverse \( \hat{\gamma}^{-1}(t,0) \) exists, we may invert the first equality in Eq. (2.176) and write \( \rho_S(0) = \hat{\gamma}^{-1}(t,0)\rho_S(t) \). Inserting this identity into Eq. (2.233) the evolution of the reduced state \( \rho_S(t) \) is given in terms of a time local generator

\[
\frac{d}{dt} \rho_S(t) = K(t)\rho_S(t),
\]

(2.234)

where

\[
K(t) = \hat{\gamma}(t,0)\hat{\gamma}^{-1}(t,0).
\]

(2.235)

Writing \( K(t) \) as in Eq. (2.235) above presupposes the existence of the inverse \( \hat{\gamma}^{-1}(t,0) \), which is not always guaranteed. Thus, \( K(t) \) might not be defined for all times. In many cases, however, it does exist or it exists apart from isolated singularities at certain points in time such that the following results still hold [75].

Let us assume \( K(t) \) exists for all times \( t \). Then, interestingly, master equations of the form Eq. (2.234) can be cast into a form very similar to the standard form of the Markovian master equation reported in Eq. (2.181). Demanding that the generator \( K(t) \) be trace and Hermiticity preserving one can actually show that it can be written in the form [75, 76]

\[
K(t)\rho_S(t) = -\frac{i}{\hbar} [\hat{H}_S(t), \rho_S(t)] + \sum_k \gamma_k(t) \left( A_k(t)\rho_S(t)A_k^+(t) - \frac{1}{2} \{A_k^+(t)A_k(t), \rho_S(t)\} \right),
\]

(2.236)

where now the Lindblad operators and rates are explicitly time dependent. Thus, dynamics including memory effects that are not described by a Markovian master equation can be described by an equation of motion very similar in form.

Two remarks are in order now. First, it is known that the generator in Eq. (2.236) leads to completely positive dynamics for \( \gamma_k(t) > 0 \) for all \( t \) and for all \( k \). It is, however, still an open problem to provide necessary and sufficient conditions when the generator in Eq. (2.236) leads to completely positive dynamics, in general [75, 77]. Second, the generator in Eq. (2.236) can
describe dynamics that include memory effects. This, however, does not mean that it describes non-Markovian dynamics, in general. Actually, it turns out that the concept of non-Markovianity in quantum dynamics is quite subtle and can be defined in different ways as we discuss in Chapter 4.

2.4.5. Adiabatic elimination

In the previous section, we considered how to obtain an effective description of a system interacting with its surrounding environment. The motivation to seek such an effective description was that the environment was very large or even infinite and we needed to reduce the complexity of the problem in order to describe the system’s dynamics. But already for smaller systems it is usually desirable to obtain a description of the problem under scrutiny in a Hilbert space of the smallest possible dimension. An example is the effective two-level description that we applied to the internal degrees of freedom of atoms.

A technique that is commonly used to achieve such a reduction of complexity, also in smaller systems, is adiabatic elimination. Adiabatic elimination is usually applied if one can divide the dynamical degrees of freedom of a system in “fast” and “slow” degrees of freedom and provides a way of eliminating the “fast” degrees of freedom from the dynamics. In atomic physics, adiabatic elimination is often applied to disregard states from a multilevel system that are initially not occupied and only very weakly and off-resonantly coupled to the initially occupied states. After a more general presentation of adiabatic elimination we will discuss an example of this kind. In order to introduce adiabatic elimination we follow the presentation in [78].

Let us consider a system with Hamiltonian $H$ that evolves according to the Schrödinger equation

$$\frac{i\hbar}{\text{d}t} |\psi(t)\rangle = H |\psi(t)\rangle.$$  \hspace{1cm} (2.237)

Assuming the eigenvalues of $H$ can be separated in two sets that are largely separated in magnitude, we define the projector $P$ and the orthogonal projector $Q = 1 - P$ to the two subspaces. We take $P$ as the projector on the subspace of the slowly evolving degrees of freedom, i.e. the small eigenvalues, and $Q$ on that of the fast evolving degrees of freedom, i.e. large eigenvalues. We are interested in finding the dynamics in the subspace associated to $P$. With the shorthand notations $a \equiv P |\psi(t)\rangle$ and $b \equiv Q |\psi(t)\rangle$, the Schrödinger equation may now be written as

$$\frac{i\hbar}{\text{d}t} a = PHa + PHQb,$$  \hspace{1cm} (2.238)

$$\frac{i\hbar}{\text{d}t} b = QHPa + QHQb.$$  \hspace{1cm} (2.239)

Now let $\tau_F$ be a typical time scale in the evolution of the fast subspace, i.e. a typical eigenvalue of $(QHQ)^{-1}$. The time scale we are interested in is a typical time scale of the slowly evolving part of the Hamiltonian $\tau_S \gg \tau_F$. Now, loosely speaking the derivative of $b$ varies very rapidly on the time scale of interest and we can consider its average over many cycles which we set to zero. This is referred to as adiabatic elimination of $b$ from the dynamics. From Eq. (2.239) we then obtain

$$b = -(QHQ)^{-1} QHPa.$$  \hspace{1cm} (2.240)

Inserting this expression back into Eq. (2.238) yields an equation of motion for the degrees of freedom $a$

$$\frac{i\hbar}{\text{d}t} a = (PHP - PHQ(QHQ)^{-1} QHP) a.$$  \hspace{1cm} (2.241)

From the above equation we can identify an effective Hamiltonian for the slowly evolving degrees of freedom

$$H_{\text{eff}} = PHP - PHQ(QHQ)^{-1} QHP.$$  \hspace{1cm} (2.242)
2. Trapped atomic ions in radio-frequency traps

Figure 2.4.: Adiabatic elimination in a three-level system: The left part of the figure shows a three-level system with coherent couplings of levels $|g\rangle$ and $|e\rangle$ to a level $|r\rangle$ described by the Hamiltonian $H = H_{3ls} + H_{\text{int}}$ from Eq. (2.245). For $|\Delta| \gg |\Omega_1|, |\Omega_2|$ the upper level may be adiabatically eliminated leading to an effective two-level description with a coupling between the two levels displayed on the right with effective Hamiltonian in Eq. (2.253). Here we assume $\hbar (|\Omega_1|^2 - |\Omega_2|^2)/(4\Delta) > 0$. See the text for details.

Let us remark that in a similar manner effective dynamics can be found if the dynamics is governed by a Liouvillian if there are two subspaces where one of the two subspaces is rapidly decaying. Then the argument to set the derivative in Eq. (2.239) equal to zero would be that the degrees of freedom in $b$ have relaxed so quickly that they can be assumed to be in their stationary state on the time scale of interest. This is very similar to the coarse-grained description of the Markovian master equation.

In order to illustrate the procedure let us consider a concrete example. We consider a $\Lambda$-type three-level system as depicted on the left-hand side of Fig. 2.4. The three-level system consists of the levels $|g\rangle$, $|e\rangle$ and $|r\rangle$ which could, for example represent, three atomic levels.

The free Hamiltonian of the three levels can be written as

$$H_{3ls} = -\hbar \omega_g |g\rangle \langle g| - \hbar \omega_e |e\rangle \langle e|.$$  

(2.243)

Let us assume that the levels $|g\rangle$ and $|e\rangle$ are coupled to state $|r\rangle$ by a Hamiltonian

$$H_{\text{int}} = \hbar \frac{\Omega_1}{2} |r\rangle \langle g| e^{-i\omega_1 t} + \hbar \frac{\Omega_2}{2} |r\rangle \langle e| e^{-i\omega_2 t} + \text{H.c.},$$  

(2.244)

where the frequencies $\omega_1$ and $\omega_2$ satisfy $\omega_1 = \omega_g - \Delta$ and $\omega_2 = \omega_e - \Delta$. Accordingly, the full Hamiltonian of the system is

$$H = H_{3ls} + H_{\text{int}}.$$  

(2.245)

Moving to an interaction picture with respect to the Hamiltonian $\tilde{H}_{3ls} = -\hbar \omega_g |g\rangle \langle g| - \hbar \omega_e |e\rangle \langle e|$, we obtain the Hamiltonian

$$\tilde{H}_{\text{int}} = -\hbar \Delta (|g\rangle \langle g| + |e\rangle \langle e|) + \left( \frac{\hbar \Omega_1}{2} |r\rangle \langle g| + \frac{\hbar \Omega_2}{2} |r\rangle \langle e| + \text{H.c.} \right).$$  

(2.246)

Adjusting the zero point of the energy we may write the above Hamiltonian as

$$\tilde{H}_{\text{int}} = \hbar \Delta |r\rangle \langle r| + \left( \frac{\hbar \Omega_1}{2} |r\rangle \langle g| + \frac{\hbar \Omega_2}{2} |r\rangle \langle e| + \text{H.c.} \right).$$  

(2.247)

We can now write down the Schrödinger equation for the three level system in an interaction
2.5. Stimulated Raman transitions and spin-dependent optical dipole forces

In this section we study the effects of a so-called stimulated Raman laser configuration on a Λ-type three-level system. There are two parameter regimes which are of particular interest to us. Namely, the regimes of two-photon stimulated Raman transitions and the so-called spin-dependent optical dipole forces. The two-photon stimulated Raman transitions couple two atomic levels using an off-resonant coupling of both levels to a third atomic level. This mechanism allows us to drive transitions with frequencies in the microwave and radio-frequency domain by laser radiation. In particular, this enables one to drive sideband transitions also for
qubits with transition frequencies in these domains. The spin-dependent optical dipole forces facilitate the coupling between internal and external degrees of freedom on magnetic-field sensitive qubit transitions. The coupling of the qubit to the motional degrees of freedom is in the $\sigma^z$ basis and also relies on off-resonant coupling of the qubit states to a third level.

In the above mentioned processes the $\Lambda$ system is described as an effective two-level system. The derivation of the effective Hamiltonians of these processes relies on adiabatic elimination of the upper level of the $\Lambda$ scheme. In contrast to the previous chapter here we also take into account spontaneous emission from the upper level.

The starting point of the treatment is a master equation of the full system that includes the ground and excited state manifolds. The considered situation is depicted in Fig. 2.5. In the above mentioned processes the $\Lambda$ system is described as an effective two-level system.

The effective operators are given by [80]

$$H = H_g + H_{\text{ex}} + \sum_l [V^l_+ (t) + V^l_- (t)] = H_g + H_{\text{ex}} + V_+ (t) + V_- (t).$$

The Lindblad operators $L_k$ represent each a decay process from an excited state to the ground state and already incorporate the decay rates. Under the assumption that the dynamics is governed by the master equation (2.254) and the coupling between the ground and excited states is weak, an adiabatic elimination of the excited states is performed in [80]. Then, one obtains an effective master equation in the ground state manifold

$$\dot{\rho} = -\frac{i}{\hbar} [H_{\text{eff}}, \rho] + \sum_k \left( L_k^\dagger \rho L_k^+ - \frac{1}{2} \{ L_k^\dagger L_k, \rho \} \right).$$

Here, $H$ is the Hamiltonian of the full system consisting of the ground state manifold Hamiltonian $H_g$, the excited state manifold Hamiltonian $H_{\text{ex}}$ and the interaction Hamiltonian $H_{\text{int}} = \sum_l [V^l_+ (t) + V^l_- (t)]$, where each contribution $l$ corresponds to an applied field. The elements $V^l_+ (t)$ excite states from the ground state manifold to the excited state manifold and the elements $V^l_- (t)$ deexcite from the excited state manifold to the ground states. Note that, in particular, we have $V^l_+ (t) = (V^l_- (t))^\dagger$. Accordingly, the total Hamiltonian reads

$$H = H_g + H_{\text{ex}} + \sum_l [V^l_+ (t) + V^l_- (t)] = H_g + H_{\text{ex}} + V_+ (t) + V_- (t).$$

The effective operators are given by [80]

$$H_{\text{eff}} = -\frac{1}{2} \left[ V_- (t) \sum_{l,s} \left( H^{(l,s)}_{\text{NH}} \right)^{-1} V^{(l,s)}_+ (t) + \text{h.c.} \right] + H_g,$$

$$L_k^{\text{eff}} = L_k \sum_{l,s} \left( H^{(l,s)}_{\text{NH}} \right)^{-1} V^{(l,s)}_+ (t).$$

Here, the sum over $s$ runs over all ground states in $H_g$, and $V^{(l,s)}_+ (t)$ is the component of $V^l_+ (t)$ coupling state $s$ of the ground state manifold to the excited states. The $H^{(l,s)}_{\text{NH}}$ are the non-Hermitian Hamiltonians given by [80]

$$H^{(l,s)}_{\text{NH}} = H_{\text{ex}} - \frac{i \hbar}{2} \sum_k \left( L_k^\dagger L_k - \varepsilon_s \mathbb{I}_{\text{ex}} - \hbar \omega_l \mathbb{I}_{\text{ex}} \right).$$
2.5. Stimulated Raman transitions and spin-dependent optical dipole forces

The manifold of ground states with Hamiltonian $H_g$ is coupled to a manifold of decaying excited states with Hamiltonian $H_{ex}$ by the interaction Hamiltonian $H_{int} = V_+ + V_-$. The decay processes are described by Lindblad operators $L_k$ and are indicated by curly lines. Figure derived from [80]. The figure shows an example of the scenario in part a), a three-level Λ-system consisting of the ground states $|g\rangle$ and $|e\rangle$ and the decaying excited state $|r\rangle$. The curly lines in the figure represent spontaneous emission from the excited to the ground states with rates $\Gamma_s, s = e, g$. The ground states are separated in frequency by $\omega_0$ and both feature a dipole-allowed transition to the state $|r\rangle$. We consider that the transitions are driven by two laser beams where laser $l$ has Rabi frequency $\Omega_l$, on transition $|s\rangle \leftrightarrow |r\rangle$. Both lasers are far-detuned from their respective transitions by an amount $\delta_l \approx \Delta_R$. The detuning $\Delta_R$ satisfies $\Delta_R \gg \Omega_l, \Gamma$. Note that a red detuning as sketched here means $\Delta_R < 0$. Depending on the effective laser frequency $\omega_L = \omega_1 - \omega_2$ different effective operations between the ground states can be implemented.

where $s$ labels the ground states in $H_g$ with corresponding energy $\epsilon_s$ and $\omega_l$ is the frequency of field $l$. $I_{ex}$ is the identity operator of the excited state manifold.

We will now focus on the situation depicted in part b) of Fig. 2.5. We consider an ion where the internal levels form a Λ-type three-level system consisting of the ground states $|g\rangle$ and $|e\rangle$, which are separated in energy by $\bar{\hbar}\omega_0$ and have an electric dipole-allowed optical transition to a decaying excited state $|r\rangle$. The Hamiltonian of the system without additionally applied fields reads

$$H_{at} = \sum_{i=e,g,r} \epsilon_i |i\rangle \langle i|,$$

(2.260)

with $\epsilon_i$ the energy of the corresponding state. We assume that the dipole transitions are driven by two laser fields with frequencies $\omega_1$ and $\omega_2$. In general, both lasers couple to both transitions and we denote the Rabi frequency of laser $l$ on transition $|s\rangle \leftrightarrow |r\rangle$, where $s = e, g$, by $\Omega_{l,s}$. The interaction Hamiltonian then reads

$$H_L(t) = \hbar \sum_{l=1,2} \sum_{s=e,g} \frac{\Omega_{l,s}}{2} e^{-i\omega_l t} |s\rangle \langle r| + H.c.,$$

(2.261)

where we have already performed a rotating wave approximation using $\Omega_{l,s} \ll \omega_l$. For ease of notation we have included the phase factors $e^{i(k_l r + \phi_l)}$ into the Rabi frequencies. Here $r$ is the ion’s position operator and $k_l$ and $\phi_l$ denote the laser wave vectors and phases, respectively. Finally, we assume that decay from the excited level to the ground states is described by the dissipator

$$\mathcal{D}_{se} \rho = \sum_{s=e,g} \left( L_s \rho L_s^\dagger - \frac{1}{2} \{ L_s^\dagger L_s, \rho \} \right),$$

(2.262)
where $L_s = \sqrt{\Gamma_s} \langle s \rangle \langle r \rangle$ such that $\Gamma_s$ is the decay rate from state the excited $|r\rangle$ to ground state $|s\rangle$. The subscript “se” stands for “spontaneous emission”. Combining Eqs. (2.260), (2.261) and (2.262) we obtain the master equation for the evolution of the full three level system

$$\dot{\rho} = -\frac{i}{\hbar}[H_{at} + H_L(t), \rho] + D_{se} \rho. \quad (2.263)$$

Let us now introduce the overall decay rate $\Gamma = \Gamma_g + \Gamma_e$ and the detuning $\delta_{l,s}$ of laser $l$ on transition $|s\rangle \rightarrow |r\rangle$

$$\delta_{l,s} = (\varepsilon_r - \varepsilon_s)/\hbar - \omega_l. \quad (2.264)$$

We will now assume that both lasers are far detuned from both transitions $\Delta_R \gg \Omega_{l,s}, \Gamma$ and that the detunings $\delta_{l,s}$ are approximately equal for all transitions

$$\delta_{l,s} \approx \Delta_R. \quad (2.265)$$

Note that this implies $\Delta \gg \omega_0$. Since in this case the lasers are far off resonant for all transitions the ground states are only weakly coupled to the decaying excited state. This is the situation where we can apply the adiabatic elimination procedure from [80] to obtain an effective dynamics in the ground state manifold $|e\rangle, |g\rangle$.

Comparing Eqs. (2.254) and (2.255) with Eqs. (2.260)-(2.263), we obtain the following correspondences:

$$H_g = \sum_{s=e,g} \varepsilon_s |s\rangle \langle s|, \quad H_{ex} = \varepsilon_r |r\rangle \langle r|$$

and the interaction Hamiltonian

$$H_{int} = H_L(t).$$

The Lindblad operators $L_k$ are those from the dissipator $D_{se}$ in Eq. (2.262).

The non-Hermitian Hamiltonians $H_{NH}^{(l,s)}$ in our system read

$$H_{NH}^{(l,s)} = \hbar(\delta_{l,s} - i\frac{\Gamma}{2}) |r\rangle \langle r|. \quad (2.266)$$

We can now evaluate the expressions (2.257) and (2.258) for the effective Hamiltonian and Lindblad operator. The effective Hamiltonian can be written as

$$H_{eff} = H_g' + H_{at} + H_{odf}. \quad (2.267)$$

The first part contains the ground state levels whose energy is shifted due to the off-resonant interaction with the excited level

$$H_g' = \sum_{s=e,g} (\varepsilon_s + \Delta \varepsilon_s) |s\rangle \langle s|, \quad (2.268)$$

where the shifts $\Delta \varepsilon_s$ are given by

$$\Delta \varepsilon_s = -\sum_{l=1,2} \sum_{s=e,g} \frac{\hbar \Omega_{l,s}^2}{4\delta_{l,s}^2 + \Gamma^2} \delta_{l,s}. \quad (2.269)$$

These are the well-known ac-Stark shifts [51] of the ground state levels that arise due to the off-resonant coupling to the excited level through the applied laser beams.

The differential ac-Stark shift between the spin levels leads to a shift of the resonance frequency and could be absorbed into a new resonance frequency. Yet, this is experimentally not desirable because fluctuating laser intensities would then lead to a fluctuating resonance frequency and thus dephasing. Often the differential ac-Stark shift can be canceled in experiments by adjusting polarization, detuning and intensity of the applied laser beams [81] and the problem can be mitigated. Hence, we ignore this contribution in the sequel and take $H_g'$ as

$$H_g' = \frac{\hbar \omega_0}{2} \sigma^z, \quad (2.270)$$
where $\sigma^+ = |e\rangle \langle e| - |g\rangle \langle g|$. The second part, $H_{\text{st}}$, induces couplings between the states and can be written as

$$H_{\text{st}} = \hbar \sum_{l,j} \frac{\Omega_{l,j}^{\text{st}}}{2} \sigma^+ e^{-i(\omega_1 - \omega_0)t} + \text{H.c.},$$

(2.271)

where $\sigma^+ = |e\rangle \langle g| = (\sigma^-)^\dagger$ and

$$\Omega_{l,j}^{\text{st}} = -\frac{\Omega_{l,g}^* \Omega_{l,e}(\delta_{l,g} + \delta_{l,e})}{(2\delta_{l,g} - i\Gamma)(2\delta_{l,e} + i\Gamma)}.$$  

(2.272)

The third part of the effective Hamiltonian induces time-dependent ac-Stark shifts and reads

$$H_{\text{od}} = \hbar \sum_s \Omega_s e^{i(\omega_1 - \omega_2)t}|s\rangle \langle s| + \text{H.c.},$$

(2.273)

where

$$\Omega_s = -\frac{\Omega_{1,s} \Omega_{2,s}(\delta_{2,s} + \delta_{1,s})}{(2\delta_{2,s} - i\Gamma)(2\delta_{1,s} + i\Gamma)}.$$  

(2.274)

While the action of the first part of the effective Hamiltonian is apparent, the second and third parts require a more careful consideration. We will study the effects of these two contributions in the two following subsections. Before that we give the effective Lindblad operators that are found to read

$$L_{g}^{\text{eff}} = \sqrt{\Gamma_g} \left( \frac{\Omega_{1,g} e^{-i\omega_1 t}}{2\delta_{1,g} - i\Gamma} + \frac{\Omega_{2,g} e^{-i\omega_2 t}}{2\delta_{2,g} - i\Gamma} \right) |g\rangle \langle g| + \sqrt{\Gamma_g} \left( \frac{\Omega_{1,e} e^{-i\omega_1 t}}{2\delta_{1,e} - i\Gamma} + \frac{\Omega_{2,e} e^{-i\omega_2 t}}{2\delta_{2,e} - i\Gamma} \right) |e\rangle \langle e|,$$

(2.275)

$$L_{e}^{\text{eff}} = \sqrt{\Gamma_e} \left( \frac{\Omega_{1,e} e^{-i\omega_1 t}}{2\delta_{1,e} - i\Gamma} + \frac{\Omega_{2,e} e^{-i\omega_2 t}}{2\delta_{2,e} - i\Gamma} \right) |e\rangle \langle e| + \sqrt{\Gamma_e} \left( \frac{\Omega_{1,g} e^{-i\omega_1 t}}{2\delta_{1,g} - i\Gamma} + \frac{\Omega_{2,g} e^{-i\omega_2 t}}{2\delta_{2,g} - i\Gamma} \right) |g\rangle \langle g|.$$  

(2.276)

### 2.5.1. Two-photon stimulated Raman transitions

In this paragraph we study the second contribution of the effective Hamiltonian in Eq. (2.267)

$$H_{\text{st}} = \hbar \sum_{l,j} \frac{\Omega_{l,j}^{\text{st}}}{2} \sigma^+ e^{-i(\omega_1 - \omega_0)t} + \text{H.c.}$$

(2.277)

with the Rabi frequencies

$$\Omega_{l,j}^{\text{st}} = -\frac{\Omega_{l,g}^* \Omega_{l,e}(\delta_{l,g} + \delta_{l,e})}{(2\delta_{l,g} - i\Gamma)(2\delta_{l,e} + i\Gamma)}.$$  

(2.278)

in more detail. We will see that this Hamiltonian, if resonant, yields the so-called two-photon stimulated Raman transitions that coherently couple the states $|g\rangle$ and $|e\rangle$. For clarity, we now consider that each laser beam only couples to one of the transitions, say laser 1 to $|g\rangle \leftrightarrow |r\rangle$ and laser 2 to $|e\rangle \leftrightarrow |r\rangle$ such that $\Omega_{1,e} = \Omega_{2,g} = 0$. Experimentally this can be achieved depending on the transition, for example, by choosing suitable polarizations for the laser beams. The Hamiltonian in Eq. (2.277) then reduces to

$$H_{\text{st}} = \hbar \frac{\Omega_{1,2}^{\text{st}}}{2} \sigma^+ e^{-i(\omega_1 - \omega_0)t} + \text{H.c.}$$

(2.279)

We further assume laser frequencies $\omega_1, \omega_2$ such that $\omega_1 - \omega_2 = \omega_0$. Then, the above Hamiltonian produces a resonant coupling between the states $|e\rangle$ and $|g\rangle$. This can be seen by moving
to an interaction picture with respect to \( H'_g \), where \( H_{sf} \) takes the form of the carrier Hamiltonian in Eq. (2.147)

\[
H_{sf} = \hbar \frac{\Omega_{sf}}{2} \sigma^+ + \text{H.c.} \tag{2.280}
\]

Accordingly, for \( \omega_1 - \omega_2 = \omega_0 \) the Hamiltonian \( H_{sf} \) induces coherent transitions between the states \( |g\rangle \) and \( |e\rangle \). These transitions are called two-photon stimulated Raman transitions because we can physically think of the transitions as a two-step process where a photon is absorbed from one laser beam immediately followed by a stimulated emission into the other beam. This technique is commonly used to drive Rabi oscillations between hyperfine states, but it can also be used to drive sideband transitions for hyperfine qubits [39, 42]. To see this we now consider that the ion is trapped. The Hamiltonian describing the motional degrees of freedom then reads

\[
H_m = \sum_{\alpha=x,y,z} \hbar \omega_\alpha a_\alpha^+ a_\alpha. \tag{2.281}
\]

Writing out the phase factors \( e^{i(k_\alpha t + \phi_\alpha)} \) we included into the Rabi frequencies and keeping the assumption \( \Omega_{1,\epsilon} = \Omega_{2,\epsilon} = 0 \), the Hamiltonian \( H_{sf} \) becomes

\[
H_{sf} = \hbar \frac{\Omega_{sf}}{2} e^{i(k_1 - k_2) t} e^{i(\phi_1 - \phi_2)} e^{-i(\omega_1 - \omega_2) t} + \text{H.c.} \tag{2.282}
\]

Introducing the effective wave vector \( k_{\text{eff}} = k_1 - k_2 \), effective phase \( \phi_{\text{eff}} = \phi_1 - \phi_2 \) and effective frequency \( \omega_{\text{eff}} = \omega_1 - \omega_2 \), the Hamiltonian in Eq. (2.282) takes the form

\[
H_{sf} = \hbar \frac{\Omega_{sf}}{2} e^{i(k_{\text{eff}} t + \phi_{\text{eff}})} e^{-i\omega_{\text{eff}} t} + \text{H.c.} \tag{2.283}
\]

Comparing with Eq. (2.132) this is exactly the form of the Hamiltonian of a trapped ion interacting with a monochromatic field of wave vector \( k_{\text{eff}} \), phase \( \phi_{\text{eff}} \) and frequency \( \omega_{\text{eff}} \). We could now apply the treatment after Eq. (2.132) to \( H_{sf} \) in Eq. (2.283) and obtain the red and blue sideband Hamiltonians in Eqs. (2.148) and (2.149) after moving to an interaction picture with respect to \( H_0 = H'_g + H_m \).

Recall, however, that we need a non-negligible Lamb-Dicke factor to obtain spin-motion coupling. For this, in turn, we need an optical wave vector \( k_{\text{eff}} \). Usually, the Raman detuning \( \Delta_R \) is still small compared to the transition frequencies \( (\epsilon_e - \epsilon_s)/\hbar \) and the modulus of the wave vectors \( k_1 \) and \( k_2 \) is almost equal \( |k_1| \approx |k_2| = k \). Thus, if we direct the two beams \( k_1 \) and \( k_2 \) such that they are perpendicular, we obtain an effective wave vector with modulus \( |k_{\text{eff}}| \approx \sqrt{2} k \). Due to the optical wavelength of the laser radiation this leads to a non-negligible Lamb-Dicke factor. If, on the other hand, the two wave vectors are parallel, we have \( k_{\text{eff}} \approx 0 \) and there will be no coupling to the motion.

Let us finally note that if we have that one laser only couples to one transition, e.g. \( \Omega_{1,\epsilon} = \Omega_{2,\epsilon} = 0 \) as above, the third part \( H_{\text{odf}} \) of the effective Hamiltonian vanishes. If all Rabi frequencies \( \Omega_{i,\epsilon} \) are non-zero and we are in the regime of the stimulated two-photon Raman transitions, i.e. \( \omega_1 - \omega_2 \approx \omega_0 \), the contributions in \( H_{\text{odf}} \) are typically highly off-resonant and can be neglected in a rotating wave approximation.

### 2.5.2. Optical dipole forces

In this section we investigate the effects of the third part of the effective Hamiltonian

\[
H_{\text{odf}} = \hbar \sum_s \frac{\Omega_{sf}}{2} e^{i(\omega_0 - \omega_2) t} |s\rangle \langle s| + \text{H.c.}, \tag{2.284}
\]
2.5. Stimulated Raman transitions and spin-dependent optical dipole forces

where

\[ \Omega_{\text{odf}} = \frac{\Omega_{1,s} \Omega_{2,s} (\delta_{2,s} + \delta_{1,s})}{(2 \delta_{1,s} - i \Gamma) (2 \delta_{1,s} + i \Gamma)}. \]  \hspace{1cm} (2.285)

The contributions of \( H_{\text{odf}} \) produce a time-dependent ac-Stark shift of the ground states that can be used to create the optical dipole forces (“odf”). We can rewrite \( H_{\text{odf}} \) using the Pauli operator \( \sigma^z = |e\rangle\langle e| - |g\rangle\langle g| \) to obtain

\[ H_{\text{odf}} = \hbar \frac{\Omega_{\text{odf}}}{2} e^{-i(\alpha t - \omega t)} \sigma^z + \hbar \frac{\Omega_{\text{rw}}}{2} e^{-i(\omega t - \omega t)} I + \text{H.c.}. \]  \hspace{1cm} (2.286)

Here have introduced the Rabi frequencies

\[ \Omega_{\text{odf}} = \frac{1}{2} (\Omega_e^* - \Omega_g^*) \quad \text{and} \quad \Omega_{\text{rw}} = \frac{1}{2} (\Omega_e^* + \Omega_g^*). \]  \hspace{1cm} (2.287)

In order to assess the action of \( H_{\text{odf}} \), we again consider that the ion is trapped and we have an additional motional Hamiltonian \( H_m \), given in Eq. (2.281). The spin-dependent optical dipole forces are obtained when the beatnote between the two lasers matches one of the motional frequencies \( \omega_m \), \( \omega_1 - \omega_2 \approx \omega_m \). Normally \( \omega_m \ll \omega_a \) so that the stimulated Raman processes in \( H_{\text{sr}} \) are highly off-resonant and can be neglected in a rotating wave approximation. Writing out the phase factors that were absorbed into the Rabi frequencies \( H_{\text{odf}} \) becomes

\[ H_{\text{odf}} = \hbar \frac{\Omega_{\text{odf}}}{2} e^{i(k_{\text{eff}} r + \phi_{\text{eff}})} e^{-i \alpha t} \sigma^z + \hbar \frac{\Omega_{\text{rw}}}{2} e^{i(k_{\text{eff}} r + \phi_{\text{eff}})} e^{-i \omega t} + \text{H.c.}, \]  \hspace{1cm} (2.288)

where we have introduced the effective laser frequency \( \omega_{\text{eff}} = \omega_1 - \omega_2 \), phase \( \phi_{\text{eff}} = \phi_1 - \phi_2 \) and wave vector \( k_{\text{eff}} = k_1 - k_2 \) as above.

Choosing the coordinate system such that \( \varepsilon^0 = 0 \), we can write

\[ (k_{\text{eff}} r)_z = k_{\text{eff},z} \sqrt{\frac{\hbar}{2m \omega_z}} (a + a^\dagger), \]  \hspace{1cm} (2.289)

where \( a = a_e \) and \( a^\dagger = a_e^\dagger \). Let us now consider that \( k_{\text{eff}} \) is directed along the \( z \)-direction and that we are in the Lamb-Dicke limit. Keeping with our previous analysis we approximate the exponentials containing \( k_{\text{eff}} r \) by a first order expansion in the Lamb-Dicke factor \( \eta = |k_{\text{eff}}| \sqrt{\hbar/(2m \omega_z)} \). This yields

\[ H_{\text{odf}} \approx \hbar \frac{\Omega_{\text{odf}}}{2} e^{i \phi_{\text{eff}}} \left[ 1 + i \eta (a + a^\dagger) \right] e^{-i \omega t} \sigma^z + \hbar \frac{\Omega_{\text{rw}}}{2} e^{i \phi_{\text{eff}}} \left[ 1 + i \eta (a + a^\dagger) \right] e^{-i \omega t} + \text{H.c.}. \]  \hspace{1cm} (2.290)

If we move to an interaction picture with respect to \( H_0 = H'_g + H_m \) with \( H'_g \) and \( H_m \) from Eqs. (2.270) and (2.281) and choose \( \omega_{\text{eff}} = \omega_z - \delta \) where \( \delta > 0 \) and \( \delta \ll \omega_z \), the resonant part of \( H_{\text{odf}} \) reads

\[ \hat{H}_{\text{odf}} = \hbar i \eta \frac{\Omega_{\text{odf}}}{2} e^{i \phi_{\text{eff}}} a \sigma^z e^{i \delta t} + \hbar i \eta \frac{\Omega_{\text{rw}}}{2} e^{i \phi_{\text{eff}}} a^\dagger e^{i \delta t} + \text{H.c.}. \]  \hspace{1cm} (2.291)

In the above equation we have neglected the remaining terms in a rotating wave approximation relying on \( \Omega_{\text{odf}}, \Omega_{\text{rw}} \ll \omega_z \). Now we are left with a contribution coupling spin and motion and a second contribution that only couples to the motion. This second term causes an unwanted displacement of the harmonic oscillator describing the motion. Going back to Eq. (2.287) we see that the Rabi frequencies \( \Omega_{\text{odf}} \) and \( \Omega_{\text{rw}} \) are different combinations of the Rabi frequencies \( \Omega_s \) of (2.285). Choosing the right polarizations, intensities and detunings of the applied laser
beams one can make $\Omega_{\text{cw}}$ vanish while obtaining an appreciable $\Omega_{\text{off}}$ [81]. Assuming that the parameters are chosen appropriately we are left with

$$\tilde{H}_{\text{off}} = \hbar \eta \frac{\Omega_{\text{off}}}{2} e^{i\phi_{\text{off}}} a^\dagger \sigma^z e^{i\delta t} + \text{H.c.} \quad (2.292)$$

We can now understand the term “spin-dependent optical dipole forces”. To this end, we move to yet another interaction picture with respect to the Hamiltonian $\tilde{H}_m = -\hbar \delta a^\dagger a$. The above Hamiltonian then becomes

$$\tilde{H}_{\text{off}} = \hbar \delta a^\dagger a + \hbar \left( i \eta \frac{\Omega_{\text{off}}}{2} e^{i\phi_{\text{off}}} a^\dagger + \text{H.c.} \right) \sigma^z. \quad (2.293)$$

Let us consider that the system spin plus mode is initially in the state $|e\rangle |\Omega\rangle_2$. Trapped atomic ions in radio-frequency traps

$$\text{beams one can make } \Omega \text{ vanish while obtaining an appreciable } \Omega_{\text{off}} \text{ [81]. Assuming that the parameters are chosen appropriately we are left with}$$

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$$\text{Let us consider that the system spin plus mode is initially in the state } |e\rangle |\Omega\rangle_2, \text{ where } |0\rangle \text{ denotes the vacuum state of the mode. Since the action of } \sigma^z \text{ on the state } |e\rangle \text{ simply yields } |e\rangle \text{ and subspaces associated to } |g\rangle \text{ and } |e\rangle \text{ do not interact, we can substitute } \sigma^z \text{ by } 1 \text{ in the propagator. Doing the same in the Hamiltonian we arrive at}$$

$$\tilde{H}_{\text{off}} = \hbar \delta a^\dagger a + \hbar (\hat{\alpha} a^\dagger + \hat{\alpha}^* a), \quad (2.294)$$

$$\text{where we have defined } \hat{\alpha} = i \eta \frac{\Omega_{\text{off}}}{2} e^{i\phi_{\text{off}}} / 2. \text{ Let us assume for the moment that } \hat{\alpha} \text{ is real. We may then write}$$

$$\tilde{H}_{\text{off}} = \hbar \delta a^\dagger a + F_0 \hat{\xi}, \quad (2.295)$$

$$\text{where } F_0 = \hbar \hat{\alpha} \sqrt{2m\omega_c / \hbar} = i \hbar \Omega_{\text{off}} \langle k_{\text{eff}} \rangle e^{i\phi_{\text{off}}} \text{. Then, } \tilde{H}_{\text{off}} \text{ in Eq. (2.295) is just the Hamiltonian of a classical force acting on the harmonic oscillator representing the normal mode [82]. If the state of the qubit were } |g\rangle \text{ and the state of the total system } |g\rangle |0\rangle, \text{ the action of } \sigma^z \text{ would yield } -1 \text{ and the force would act in the opposite direction. Accordingly, the force is called state-dependent.}$$

$$\text{It is interesting to note that the time evolution of the state dependent force in Eq. (2.292) can be solved exactly. The time evolution operator of } \tilde{H}_{\text{off}} \text{ can be computed in a second-order Magnus expansion [83, 84] and takes the form}$$

$$U(t, 0) = e^{\frac{\hat{\alpha}}{\hbar} (e^{-it\hbar} - 1) \sigma^z a - \hbar c} e^{i \frac{\hbar^2}{2} \left( t - \frac{\sin(\delta t)}{\delta} \right)}, \quad (2.296)$$

$$\text{The second term in this case only produces an irrelevant global phase and we neglect it for the moment. Let us now consider again the situation that the system is initially in the state } |e\rangle |0\rangle \text{ as above. We can then again substitute } \sigma^z \text{ in the propagator by } 1 \text{ and defining}$$

$$\alpha(t) = -\frac{\hat{\alpha}}{\delta} (e^{i\delta t} - 1), \quad (2.297)$$

$$\text{we can write the propagator as}$$

$$U(t, 0) = D[\alpha(t)] = e^{\alpha(t) a^\dagger - \alpha^*(t) a}. \quad (2.298)$$

$$\text{Thus, we realize that the time evolution operator in this case is a displacement operator. } D[\alpha(t)] \text{ displaces the motional state in phase space and creates a coherent state of the motion [54]. The state of the system at time } t \text{ is then given by [82]}$$

$$|\psi(t)\rangle = |e\rangle |\alpha(t)\rangle, \quad (2.299)$$

$$\text{where}$$

$$|\alpha(t)\rangle = e^{-\frac{|\alpha(t)|^2}{2}} \sum_{n=0}^{\infty} \frac{\alpha(t)^n}{\sqrt{n!}} |n\rangle \quad (2.300)$$
Inspecting the time dependence of $\alpha(t)$ in Eq. (2.297) we see that the motional state is displaced along circular trajectories in phase space. In particular after a time $t = 2\pi/\delta$ the system returns to its initial state.

If several trapped ions are subject to a spin-dependent force, interactions between the internal levels of the ions can be created. Then, the second term in the time evolution operator of Eq. (2.296) becomes important because spin-spin interaction terms appear here, see e.g. [85, 86]. We do not discuss the creation of spin-spin interactions in more detail here because in this thesis we do not analyze models that involve spin-spin interactions.

Let us close this section by summarizing that we obtain three effects on the spin states of the $\Lambda$ system irradiated by a stimulated Raman laser configuration. The first is an ac-Stark shift of the spin levels due to the laser fields, which we normally can ignore because the differential ac-Stark shift between spin levels can often be canceled in experiments [81]. Depending on the choice of the laser frequencies there are two regimes. If one chooses the frequency difference between lasers close to the transition frequency between the spin states $\omega_1 - \omega_2 \approx \omega_0$, the second part of $H_{\text{eff}}$, namely $H_{\text{sr}}$ in Eq. (2.277), is resonant and one can drive coherent two-photon stimulated Raman transitions between the spin states. In this case, the third contribution $H_{\text{odf}}$ is usually highly off-resonant and can be neglected in a rotating wave approximation. If, however, one chooses the frequency difference between lasers close to a motional mode frequency, e.g. $\omega_1 - \omega_2 \approx \omega_z$, we are in the regime of the optical dipole forces where the Hamiltonian $H_{\text{odf}}$ in Eq. (2.284) is resonant. The stimulated Raman transitions are typically highly off-resonant in this case and $H_{\text{sr}}$ can be neglected in a rotating wave approximation.

Dissipative effects are described by the Lindblad operators in Eqs. (2.275) and (2.276). By keeping only the dominant contributions, i.e. those parts of the action of the Lindblad operators that are time independent, and using $\delta l_s \approx \Delta R$ we obtain effective operators

\begin{align}
L_{ee} &= \frac{1}{2} \sqrt{\Gamma_e} \sum_l \frac{|\Omega_{l,e}|^2}{4\Delta_R^2} \sigma^z, \\
L_{gg} &= \frac{1}{2} \sqrt{\Gamma_g} \sum_l \frac{|\Omega_{l,g}|^2}{4\Delta_R^2} \sigma^z, \\
L_{eg} &= \sqrt{\Gamma_e} \sum_l \frac{|\Omega_{l,e}|^2}{4\Delta_R^2} \sigma^+, \\
L_{ge} &= \sqrt{\Gamma_g} \sum_l \frac{|\Omega_{l,g}|^2}{4\Delta_R^2} \sigma^-.
\end{align}

The first two terms have the form of pure dephasing noise. They describe so-called Rayleigh scattering [87] where the spin state is not altered upon a scattering event. These processes can randomize the phase and introduce dephasing. The other operators describe so-called Raman scattering [87] where the spin state is changed upon a scattering event which also leads to decoherence.

Let us finally estimate the decoherence rates. If we assume the modulus of the Rabi frequencies is approximately equal $|\Omega_{l,s}| \approx \Omega_0$, we can estimate the effective scattering rate $\Gamma_{\text{eff}} \approx \Gamma \Omega_L / \Delta_R$, where $\Omega_L = \Omega_0^2 / (2\Delta_R)$ is the approximate effective laser Rabi frequency. Hence, decoherence can be largely suppressed if we choose $\Delta_R$ large enough. Note that the above rates only constitute a worst case estimate. For a realistic level scheme involving more than one excited state Rayleigh scattering can be largely suppressed by choosing the right parameters for the applied lasers [87].

2.6. Laser cooling of trapped ions

Cooling nowadays constitutes the first step in basically all experiments with trapped ions. It is essential for the operation of atomic clocks [60] and quantum information experiments [61] as well as for quantum simulation experiments [12, 55]. In most cases, it is desirable to initialize
the ions in the motional ground state. Although perfect preparation of the ground state is not possible, very low occupation numbers can be achieved. As we will see in Chapter 4, cooling can also be used for environment engineering.

There are several techniques of laser cooling. Usually, cooling is a two step process, where the first stage of cooling is so-called Doppler cooling which is then followed by a second stage of a more sophisticated cooling technique, such as resolved-sideband cooling or EIT cooling [42]. After a brief account of Doppler cooling we will consider the derivation of the equations of motion for EIT cooling.

2.6.1. Doppler cooling

For Doppler cooling the ions are illuminated with light that is red detuned, i.e. tuned to a frequency slightly smaller than the actual transition frequency, with respect to a dipole-allowed transition. Usually, dipole transitions have a linewidth that is larger than the motional frequencies and the motional sideband transitions cannot be resolved. Each absorption event transfers a momentum \( h\kappa \) in the direction of light propagation to the ion. Here \( \kappa \) is the modulus of the wave vector of the applied light. The following spontaneous emission also transfers a momentum \( h\kappa \) to the atom but in a random direction such that the net effect of recoil on the motion is zero in a first approximation.

Let us now denote the excited state of the dipole transition by \( |e\rangle \), the transition frequency \( \omega_0 \) and the frequency of the applied light by \( \omega \). If the spontaneous emission rate \( \Gamma \) of the excited state of the dipole transition is much larger than the motional frequency \( \Gamma \gg \omega_m \), one can model the radiation pressure of the applied field as a velocity-dependent force on the ion [88]. The rate of the momentum transfer is given by the product of the probability for the ion to be in the excited state, given by the matrix element \( \rho_{ee} \), times the rate of spontaneous emission from the excited state \( \Gamma \). The force can then be written as [42]

\[
F = h\kappa \Gamma \rho_{ee} = h\kappa \Gamma \frac{s_0/2}{1 + s_0 + (2\Delta_{\text{eff}}/\Gamma)^2},
\]

where we have introduced the saturation parameter \( s_0 = 2|\Omega|^2/\Gamma^2 \) that includes the Rabi frequency \( \Omega \) of the applied radiation. The detuning \( \Delta_{\text{eff}} \) is composed of the bare detuning \( \Delta = \omega - \omega_0 \) of the applied radiation and the Doppler shift \( -\kappa \cdot \mathbf{v} \), where \( \mathbf{v} \) is the ion’s velocity and \( \kappa \) the wave vector of the applied radiation, [42]

\[
\Delta_{\text{eff}} = \Delta - \kappa \cdot \mathbf{v}.
\]

Towards the end of the cooling process one may expand the force linearly in the ion’s velocity. For simplicity, we only consider one-dimensional motion from now on such that we substitute \( \kappa \rightarrow k \) and \( v \rightarrow v \). The force in Eq. (2.303) then becomes [42]

\[
F \approx F_0 (1 + \kappa v),
\]

where

\[
F_0 = \frac{h\kappa \Gamma s_0/2}{1 + s_0 + (2\Delta/\Gamma)^2} \quad \text{and} \quad \kappa = \frac{8k\Delta/\Gamma^2}{1 + s_0 + (2\Delta/\Gamma)^2}. \tag{2.306}
\]

The force \( F_0 \) is the average radiation pressure that displaces the ion from the potential zero of the trap while \( \kappa \) can be seen as a “friction coefficient”. From Eqs. (2.305) and (2.306) we see that we obtain cooling of the ionic motion for red detuning, i.e. \( \Delta = \omega - \omega_0 < 0 \), while blue detuning yields heating.

There is an appealing pictorial description for this damping force. In case of a red detuning the applied radiation appears more resonant to the ion when it travels opposite to the direction
of propagation of the light. The ion is then more likely to absorb a photon, which will slow down the ion. On the other hand, the light appears even more detuned when the ion travels in the direction of propagation of the light and the ion is much less likely to absorb a photon that increases its momentum. In the same way one can understand the heating effect in case of a blue detuning.

The average energy loss in the picture that we studied so far is given by [42]

\[ \dot{E}_c = \langle Fv \rangle = F_0 \langle \langle v \rangle + \kappa \langle v^2 \rangle \rangle = F_0 \kappa \langle v^2 \rangle, \]  
(2.307)

where we used the force from Eq. (2.305) and that \( \langle v \rangle = 0 \) for a trapped ion. Note that if \( \langle v \rangle \) were not zero, the ion would eventually escape from the trap. Following Eq. (2.307) the motion would be cooled to its ground state. This is not what happens in practice and is only due to the simplified picture that is considered here. Taking into account spontaneous emission, one finds that Doppler cooling will yield final temperatures \( T_D \) of the order [42]

\[ T_D \approx \frac{\hbar \Gamma}{2k_B}, \]  
(2.308)

where \( k_B \) is the Boltzmann constant.

Let us consider \( ^{40}\text{Ca}^+ \) as an example. The \( 4^2S_{1/2} \) to \( 4^2P_{1/2} \) transition near 397 nm with a natural linewidth \( \Gamma/2\pi \approx 20 \text{ MHz} \) can be used for Doppler cooling. Note that the transition is not closed and in order to avoid optical pumping, another laser has to be applied. The theoretical Doppler temperature for a single \( ^{40}\text{Ca}^+ \) ion is \( T_D = 0.5 \text{ mK} \) [89]. If the ion is confined in a trap with trap frequencies \( (\omega_x, \omega_y, \omega_z) = 2\pi \times (1.69, 1.62, 3.32) \text{ MHz} \), the theoretical final occupation numbers after Doppler cooling are [89]

\[ n_x,D \approx n_y,D \approx 6 \quad \text{and} \quad n_z,D \approx 3. \]  
(2.309)

In practice, they will be higher due to experimental imperfections.

After Doppler cooling, the ions are often in the Lamb-Dicke regime, Eq. (2.144). It is instructive to treat the final stages of Doppler cooling in the Lamb-Dicke regime, because it provides some physical insights that are useful also for the understanding of EIT cooling in the following. Our treatment is based on those of [42] and [88].

Let us consider a cooled ion and assume we know its scattering rate at rest

\[ W(\Delta) = \Gamma \rho_{ee}(\Delta), \]  
(2.310)

where \( \Delta \) is the detuning of the applied radiation from resonance and \( \rho_{ee}(\Delta) \) is the probability of finding the ion in the excited state for a detuning \( \Delta \). The interaction of the ion with the applied radiation is described by the Hamiltonian in Eq. (2.141). This Hamiltonian couples an initial state \( |g, n\rangle \) to states of the form \( |e, n'\rangle \). The ion can also emit on sidebands and thus after spontaneous emission the system finds itself in a state \( |g, n''\rangle \). A complete scattering, i.e. absorption-emission, cycle hence consists of the transitions \( |g, n\rangle \rightarrow |e, n'\rangle \rightarrow |g, n''\rangle \). One could now calculate the transition rates for all combinations \( (n, n', n'') \) and write down a set of equations for the probabilities \( p_n \) to find the motion in state \( |n\rangle \). This set could then be solved to find the populations at the end of the cooling process.

Assuming that the previous cooling process has left the mode in a thermal state in the Lamb-Dicke regime the absorption and emission processes will be dominated by the carrier and first red- and blue-sideband transitions. All other processes are of higher order in \( \eta \) and can therefore be neglected [42]. This is consistent with the Hamiltonian in Eq. (2.146) which we obtained for
the interaction of an ion with externally applied electromagnetic radiation in the Lamb-Dicke regime. This Hamiltonian also only features the first red- and blue-sideband transitions.

According to Eq. (2.142) the probability of making a carrier transition from state $|g,n\rangle$ will be proportional to $\Omega^2$, where $\Omega$ is the Rabi frequency of the applied radiation, while the probability of making a transition on the first red- or blue-sidebands is proportional to $\eta^2 n|\Omega|^2$ and $\eta^2 (n + 1)|\Omega|^2$, respectively. Recall that $\eta \ll 1$ in the Lamb-Dicke regime. Also, spontaneous emission from a state $|e,n\rangle$ on the red- and blue-sidebands is suppressed by factors $\eta^2 n$ and $\eta^2 (n + 1)$ with respect to the carrier. Here, $\eta$ is the Lamb-Dicke factor of spontaneous emission and is in general not equal to $\eta$ because it involves the angle of spontaneous emission with respect to the direction of motion. This can be seen from Eq. (2.339) below. For simplicity, let us consider $\eta = \eta$.

The process that is most likely to happen in this situation is absorption on the carrier followed by emission on the carrier, which does not change the motional state. The only scattering cycles are those starting in $|e,n\rangle$ and $|g,n\rangle$ ending $|g,n\pm 1\rangle$ via intermediate states $|e,n\pm (0,1)\rangle$. These processes are illustrated in Fig. 2.6 a). In total there are four processes, two of which lead to heating while the other two lead to cooling. The heating processes are: absorption on the carrier followed by emission on the first blue sideband with rate [42]

$$R_{n\rightarrow n+1}^{(1)} = W(\Delta)\eta^2(n + 1)$$  \hspace{1cm} (2.311)

and absorption on the blue sideband followed by emission on the carrier with rate [42]

$$R_{n\rightarrow n+1}^{(2)} = W(\Delta - \omega_m)\eta^2(n + 1).$$  \hspace{1cm} (2.312)

Combining the above rates we obtain the overall rate

$$R_{n\rightarrow n+1} = W(\Delta)\eta^2(n + 1) + W(\Delta - \omega_m)\eta^2(n + 1).$$  \hspace{1cm} (2.313)

Note that the second process involves absorption at $\Delta - \omega_m$ because a portion $\hbar\omega_m$ of the energy of the radiation goes into the motion. Considering the analogous cooling processes involving the red sideband, we obtain the rate [42]

$$R_{n\rightarrow n-1} = W(\Delta)\eta^2 n + W(\Delta + \omega_m)\eta^2 n.$$  \hspace{1cm} (2.314)

With these rates we can write down an equation of motion for the populations $p_n$ [42]

$$\frac{dp_n}{dt} = R_{n-1\rightarrow n}p_{n-1} + R_{n+1\rightarrow n}p_{n+1} - (R_{n\rightarrow n+1} + R_{n\rightarrow n-1})p_n.$$  \hspace{1cm} (2.315)

Introducing the $n$ independent coefficients

$$A_{\pm} = \eta^2 [W(\Delta) + W(\Delta \pm \omega_m)],$$  \hspace{1cm} (2.316)

we may write the above equation as

$$\frac{dp_n}{dt} = A_- [p_{n+1}(n + 1) - p_n n] + A_+ [p_{n-1} n - p_n(n + 1)].$$  \hspace{1cm} (2.317)

The processes described by the above equation are depicted in Fig. 2.6 b).

Multiplying Eq. (2.317) by $n$ and summing over all $n$ we obtain an equation of motion for the mean occupation number $\bar{n}(t) = \frac{1}{\Omega^2} \sum_n np_n(t) = \langle n(t) \rangle$ from the above equation [88]

$$\frac{d}{dt}\bar{n}(t) = -(A_- - A_+)\bar{n}(t) + A_+.$$  \hspace{1cm} (2.318)
2.6. Laser cooling of trapped ions

In Eq. (2.318) we can identify

\[ W_c = A_- - A_+ \] (2.319)

as the cooling rate. For \( W_c > 0 \), that is for \( A_- > A_+ \), the processes annihilating quanta of motion dominate and we obtain cooling. In the opposite case, one obtains heating.

Let us now assume that we are in the cooling regime \( A_- > A_+ \). The final occupation number is reached when \( \frac{d}{dt} \bar{n}(t) = 0 \). From Eq. (2.318) we then obtain the asymptotic occupation number

\[ \bar{n}_\infty = \frac{A_+}{A_- - A_+}. \] (2.320)

Inserting the rates \( A_\pm \) from Eq. (2.316) we can write the asymptotic occupation number as

\[ \bar{n}_\infty = \frac{W(\Delta) + W(\Delta - \omega_m)}{W(\Delta + \omega_m) - W(\Delta - \omega_m)}. \] (2.321)

There are several pieces of information that we can retrieve from the above equation. First, we see that

\[ A_- - A_+ = W(\Delta + \omega_m) - W(\Delta - \omega_m). \] (2.322)

This means that the most efficient cooling happens if the scattering rate on the red sideband is much larger than that on the blue sideband. Furthermore, the smallest final occupation numbers are achieved when \( W(\Delta + \omega_m) \gg W(\Delta - \omega_m) \), \( W(\Delta) \), i.e. when the scattering rate on the red-sideband transition is also much larger than that on the carrier. Finally, inserting a Lorentzian absorption profile with \( \Gamma \gg \omega_m \) into Eq. (2.321) we recover the Doppler limit of Eq. (2.308).

We can write this limit in terms of the mean occupation number yielding [42]

\[ k_B T_D = \bar{n}_\infty h \omega_m \approx \hbar \Gamma / 2. \] (2.323)

2.6.2. EIT cooling of a single trapped ion

After Doppler cooling, trapped ions can be cooled to temperatures considerably lower than the Doppler temperature using more sophisticated cooling techniques [90]. There are two techniques which are commonly used called resolved-sideband cooling and EIT cooling, where EIT
2. Trapped atomic ions in radio-frequency traps

stands for electromagnetically induced transparency. Here we focus on EIT cooling and give a brief outline of the derivation of the equations of motion of EIT cooling. This technique has been realized in different laboratories. It has the advantage of yielding high cooling rates and allows the cooling of several modes at the same time [91,92].

We start by considering cooling of a single ion and in one spatial direction. Our treatment follows that of [93]. The extension to cooling of the normal modes of ion crystals is straightforward and will be outlined in the next section.

EIT cooling requires a three-level Λ system as depicted in Fig. 2.7 a). We label the three energy levels \( |g_1 \rangle, |g_2 \rangle \) and \( |e \rangle \). Choosing the energy scale in a way that the excited state has zero energy we can write the Hamiltonian of the three-level system as

\[
H_{\text{at}} = -\hbar \omega_l |g_1 \rangle \langle g_1| - \hbar \omega_s |g_2 \rangle \langle g_2|,
\]

where \(-\hbar \omega_s\) is the energy of state \(|g_s \rangle \), \(s = 1, 2\). We consider that the system is illuminated by two lasers with frequencies \(\omega_{L,1}\) and \(\omega_{L,2}\) and that each laser couples only one of the ground states to the excited level. We further assume that both beams are detuned by an amount \(\Delta_{\text{EIT}} = \omega_s - \omega_{L,s}, s = 1, 2\). Note that by this definition \(\Delta_{\text{EIT}} < 0\) corresponds to a blue detuning. The interaction of the atom with the lasers is described by the Hamiltonian

\[
V = \sum_{s=1,2} \frac{\hbar \Omega_s}{2} |e \rangle \langle g_s| e^{i \left( k_s z \cos \theta_s - \omega_{L,s} t \right)} + \text{H.c.},
\]

where \(k_s = |k_s|\) is the modulus of the wave vector of laser \(s\) and \(\theta_s\) is the angle between the direction of propagation of the lasers and the \(z\)-axis. Note that we have set the laser phases to zero. Moving to an interaction picture with respect to \(H_0 = -\hbar \sum_{s=1,2} \omega_{L,s} |g_s \rangle \langle g_s|\) and performing a rotating wave approximation, the Hamiltonian of the system can be written as

\[
H_{\text{EIT}} = \tilde{H}_{\text{at}} + V_{\text{int}},
\]

\[
\tilde{H}_{\text{at}} = -\hbar \Delta_{\text{EIT}} \sum_s |g_s \rangle \langle g_s|,
\]

\[
V_{\text{int}} = \sum_{s=1,2} \frac{\hbar \Omega_s}{2} |e \rangle \langle g_s| e^{i \left( k_s z \cos \theta_s \right)} + \text{H.c.}
\]

We will now consider the evolution of the internal states neglecting spin-motion coupling. To this end we define

\[
H_{\text{EIT},0} = \tilde{H}_{\text{at}} + V_0,
\]

where \(V_0\) is the zeroth order approximation of \(V_{\text{int}}\) in the Lamb-Dicke parameter:

\[
V_0 = \sum_{s=1,2} \frac{\hbar \Omega_s}{2} |e \rangle \langle g_s| + \text{H.c.}
\]

Neglecting spin-motion coupling, spontaneous emission from the excited state is accounted for by the dissipator

\[
\mathcal{D}_0 \rho = \sum_{s=1,2} \Gamma_s |g_s \rangle \langle g_s| \rho |g_s \rangle \langle g_s| - \frac{\Gamma}{2} \{ |e \rangle \langle e|, \rho \},
\]

where again the total spontaneous emission rate \(\Gamma = \Gamma_{g_1} + \Gamma_{g_2}\) is the sum of the individual emission rates.

Let us now assume the \(\Omega_l\) are real. The eigenstates of \(H_{\text{EIT},0}\) can be calculated analytically and are given by [93]

\[
|\psi_0 \rangle = \frac{1}{\Omega} \left( \Omega_2 |g_1 \rangle - \Omega_1 |g_2 \rangle \right),
\]

54
2.6. Laser cooling of trapped ions

Figure 2.7.: a) Three-level $\Lambda$ system with the ground states $|g_1\rangle$ and $|g_2\rangle$ which feature a transition to the decaying excited state $|e\rangle$. The transitions are driven by two lasers where each laser only couples to one of the transitions. $\Omega_s$ denotes the Rabi frequency on transition $|g_s\rangle \rightarrow |e\rangle$. The laser frequencies are chosen such that both beams are detuned by an amount $\Delta_{\text{EIT}}$ from their respective transitions. Spontaneous emission from the excited state to the two ground states is represented by the curly lines and $\Gamma_s$ is the decay rate to state $|g_s\rangle$. Figure derived from [93]. b) Absorption spectrum of a three-level system as in part a). We assume the system has evolved to the dark state $\rho_d$, Eq. (2.337), and is probed by a weak beam with Rabi frequency $\Omega_p \ll \Omega_1, \Omega_2$ on the transition $|g_1\rangle \rightarrow |e\rangle$. $\Delta_p$ denotes the detuning of the probe beam. Parameters are $\Delta_{\text{EIT}}/2\pi = -100\,\text{MHz}$, $\Omega_1/2\pi = 35\,\text{MHz}$, $\Omega_2/2\pi = 11\,\text{MHz}$, $\Gamma/2\pi = 41.4\,\text{MHz}$ and $\Gamma_1 = 2\Gamma_2 = 2\Gamma/3$. The parameters are chosen on the basis of energy levels in $^{24}\text{Mg}^+$ which can be used to approximately form a three-level system as in a) [91].

where $\Omega = \sqrt{\Omega_1^2 + \Omega_2^2}$ and

$$|\psi_+\rangle = \cos \varphi |e\rangle - \sin \varphi |\psi_c\rangle,$$

$$|\psi_-\rangle = \sin \varphi |e\rangle - \cos \varphi |\psi_c\rangle,$$

(2.333)

(2.334)

where $|\psi_c\rangle = 1/\Omega (\Omega_1 |g_1\rangle + \Omega_2 |g_2\rangle)$. The angle $\varphi$ obeys

$$\tan \varphi = \frac{\sqrt{\Delta_{\text{EIT}}^2 + \Omega^2} - \Delta_{\text{EIT}}}{\Omega}.$$

(2.335)

The states $|\psi_+\rangle$ and $|\psi_-\rangle$ involve the excited state $|e\rangle$ and therefore decay. The third eigenstate $|\psi_d\rangle$ is a dark state of the dynamics generated by the Liouvillian

$$\mathcal{L}_0 \rho = -\frac{i}{\hbar} [H_{\text{EIT}}, \rho] + \mathcal{D}_0 \rho,$$

(2.336)

which can easily be checked inserting $\rho_d = |\psi_d\rangle \langle \psi_d|$ into the above equation. Hence, under $\mathcal{L}_0$ the internal atomic levels will evolve into the dark state

$$\rho_d = |\psi_d\rangle \langle \psi_d|.$$

(2.337)

The internal levels evolve to the dark state on a characteristic time scale $\Gamma_d$, which is given by the smallest decay rate of the other eigenstates of the Hamiltonian.

It is now interesting to inspect the absorption spectrum of the three-level system at rest when it has evolved into the dark state. We consider the absorption of a weak probe beam with Rabi
frequency $\Omega_p \ll \Omega_1, \Omega_2$ on the $|g_1 \rangle \rightarrow |e \rangle$ transition. In Fig. 2.7 b) we plot the absorption as a function of the probe beam detuning $\Delta_p = \omega_l - \omega_p$, where $\omega_p$ is the probe beam frequency. In App. B we show how this absorption profile can be calculated. The absorption vanishes for a detuning $\Delta_p = \Delta_{EIT}$ of the probe beam and there is a resonance on either side of the absorption null. The resonance on the left is broad while the one on the right is quite narrow. The resonances can be identified with transitions to the other eigenstates of $H_{EIT,0}$ [93]. The absorption null tells us that the ion does not absorb photons from the dressing beams at a detuning $\Delta$ can be identified with transitions to the other eigenstates of $H_{EIT,0}$ [93]. The absorption null tells us that the ion does not absorb photons from the dressing beams at a detuning.

Hence, one can achieve much stronger absorption on the red-sideband transition than on the blue-sideband transition while there are no carrier excitations. Accordingly, high cooling rates on the red- and blue-sideband transitions determines the final occupation number of the motional direction. From Eqs. (2.321) and (2.322) we know that the difference in the scattering rates on the red- and blue-sideband transitions determines the final occupation number of the mode and the cooling rate. By an appropriate choice of the laser parameters the red-sideband absorption is located in the narrow feature to the right of the absorption null, and the absorption of the blue sideband is located in the tail of the broad resonance to the left of the absorption null. As we pointed out earlier, absorption from the dressing beams on the carrier is suppressed. Hence, one can achieve much stronger absorption on the red-sideband transition than on the blue-sideband transition while there are no carrier excitations. Accordingly, high cooling rates and small final occupation numbers are possible.

After this intuitive approach to the cooling mechanism we proceed to the derivation of the equations of motion for EIT cooling. To this end, we now take into account the motion of the ion along the $z$-axis of motion. In the Lamb-Dicke regime the ion will then be probed by sidebands at frequencies $\omega$ and $\omega_m$ in the rest frame of the ion. In the Lamb-Dicke regime the ion will then be probed by sidebands at frequencies $\Delta_{EIT} \pm \omega_m$ by the applied lasers, if their $k$-vectors have a component along the motional direction. From Eqs. (2.321) and (2.322) we know that the difference in the scattering rates on the red- and blue-sideband transitions determines the final occupation number of the mode and the cooling rate. By an appropriate choice of the laser parameters the red-sideband absorption is located in the narrow feature to the right of the absorption null, and the absorption of the blue sideband is located in the tail of the broad resonance to the left of the absorption null. As we pointed out earlier, absorption from the dressing beams on the carrier is suppressed. Hence, one can achieve much stronger absorption on the red-sideband transition than on the blue-sideband transition while there are no carrier excitations. Accordingly, high cooling rates and small final occupation numbers are possible.

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The Liouvillian $\mathcal{L}$ describing the evolution of the full system including the internal and external degrees of freedom is now given by

$$\rho = \mathcal{L}\rho = -\frac{i}{\hbar}[H_{\text{EIT}}, \rho] + \mathcal{D}\rho,$$  \hspace{5cm} (2.342)

with $H_{\text{EIT}}$ and $\mathcal{D}$ from Eqs. (2.338) and (2.339), respectively.

We assume that the motion of the ion has been brought to the Lamb-Dicke regime by some other cooling mechanism such that we can assume $\eta_r = k_s \sqrt{\hbar/(2m\omega_r)} \ll 1$ in the following. As in the previous sections, we expand the factors $e^{ik_z \cos \theta_r} = e^{i \eta_r \cos \theta_r (\alpha + \alpha^* )}$ appearing in the Liouvillian of Eq. (2.342) in powers of $\eta_r$, where we used $\zeta = \zeta^0 + q_z$ and assumed $\zeta^0 = 0$. To second order we obtain

$$\rho = (\mathcal{L}_0 + \mathcal{L}_1 + \mathcal{L}_2)\rho,$$  \hspace{5cm} (2.343)

where $\mathcal{L}_i$ is the $i$-th order expansion of $\mathcal{L}$ in the $\eta_r$. $\mathcal{L}_0$ is defined in Eq. (2.336). Now we have to add the motional Hamiltonian $H_m = \hbar \omega_r a^+ a$ such that we obtain

$$\mathcal{L}_0\rho = -\frac{i}{\hbar}[H_{\text{EIT},0} + H_m, \rho] + \mathcal{D}_0\rho.$$  \hspace{5cm} (2.344)

The first and second order contributions read

$$\mathcal{L}_1\rho = -\frac{i}{\hbar}[V_1 q_z, \rho],$$  \hspace{5cm} (2.345)

$$\mathcal{L}_2\rho = -\frac{i}{\hbar}[V_2 q_z^2, \rho] + \mathcal{D}_2\rho.$$  \hspace{5cm} (2.346)

Here we have used definitions

$$V_1 = \frac{i\hbar}{2} \sum_s k_s \cos \theta_s \Omega_s (|e\rangle \langle g_s| - |g_s\rangle \langle e|),$$

$$V_2 = -\frac{\hbar}{4} \sum_s k_s^2 \cos^2 \theta_s \Omega_s (|e\rangle \langle g_s| + |g_s\rangle \langle e|)$$

and

$$\mathcal{D}_2\rho = \sum_s \alpha_s^2 k_s^2 \Gamma_s |g_s\rangle \langle e| \left( q_s \rho q_s - \frac{1}{2} q_s^2 \rho - \frac{1}{2} \rho q_s^2 \right) |e\rangle \langle g_s|,$$

where $\alpha_s^2 = \frac{1}{2} \int_0^1 d(\cos \theta) \mathcal{N}(\cos \theta) \cos^2 \theta$. For an electric dipole transition $\mathcal{N}(\cos \theta) = \frac{3}{4} (1 + \cos^2 \theta)$ [94] such that the first moment vanishes. Note the we have not written out the Lamb-Dicke factors for clarity but remember that products $(k_s q_z)^\alpha$ are of order $\eta_r^\alpha$.

As we pointed out earlier, in this setting $\rho_d$ is not a dark state of the Liouvillian anymore. The assumption that we use to proceed is that the timescale of relaxation of the internal levels, which we call $\tau_{\text{int}}$, is much smaller than a typical time scale $\tau_{\eta}$ of the processes coupling internal and external degrees of freedom in $\mathcal{L}_1$ and $\mathcal{L}_2$. These processes are slower due to the smallness of the $\eta_r$. The internal degrees of freedom can then be taken to have relaxed to their stationary state for a typical time scale of the coupling processes. In this regime, one can adiabatically eliminate the internal degrees of freedom from the dynamics [88].

More formally, to zeroth order in the $\eta_r$ the internal and external degrees of freedom are decoupled. Consider the eigenstates of the Liouvillian $\mathcal{L}_0$. These are of the form

$$\mathcal{L}_0 (\rho_d \otimes |n\rangle \langle k|) = \lambda_{n,k} (\rho_d \otimes |n\rangle \langle k|),$$

where the eigenvalues are given by $\lambda_{n,k} = -i(n-k)\omega_k$ and the subspaces for each of the $\lambda_{n,k}$ are infinitely degenerate. The different subspaces are coupled by the processes in $\mathcal{L}_1$ and $\mathcal{L}_2$ that
couple the internal and motional degrees of freedom. If the energy separation of the subspaces is much larger than the coupling, i.e. \( \omega_\ell \gg \eta_\ell \Omega_\ell \), an equation of motion for the dynamics in \( \lambda_{n,k} = 0 \) subspace can be derived in second order perturbation theory in the \( \eta_\ell \) [88]. Let us define the projector on the \( \lambda_{n,k} = 0 \) subspace

\[
P_0^\dagger \rho (\rho_i \otimes P_k) \rho = \rho_i \otimes P_k \text{Tr}_i (\rho) = \rho_i \otimes \sum_{n=0}^\infty |n\rangle \langle n| \text{Tr}_i (\rho) |n\rangle = \rho_i \otimes \mu,
\]

where \( \text{Tr}_i \) denotes the trace over the internal degrees of freedom and the projector to the orthogonal subspace is

\[
Q_0 = I - P_0.
\]

The equation of motion in the \( \lambda_{n,k} = 0 \) subspace can now be written as [88, 93, 94]

\[
\frac{d}{dt} P_0 \rho (t) = \left[ P_0 L_2 P_0 + P_0 L_1 Q_0 (\lambda_{n,k} = 0) Q_0 \lambda_{n,k} = 0 \right] \rho (t)
\]

\[
= \left[ P_0 L_2 P_0 + \int_0^\infty \tau \rho_0 \lambda_{n,k} = 1 e^{\lambda_{n,k} = 1} \lambda_{n,k} = 1 \rho_0 \lambda_{n,k} = 1 \right] \rho (t).
\]

The second equation can be achieved by assigning a small negative real part \( -\varepsilon \), \( \varepsilon > 0 \) to the subspaces with a purely imaginary eigenvalue and taking the limit \( \varepsilon \to 0 \) in the end. Here, it was already used that \( P_0 \lambda_{n,k} = 0 = 0 \), which can readily be checked.

After considerable algebra which is summarized in App. A, one obtains an equation of motion for the motional state \( \mu (t) = P_k \text{Tr}_i \rho (t) \). After tracing out the internal degrees of freedom in Eq. (2.353) this equation reads [93]

\[
\lambda (t) = \Lambda_- [a(t) a^\dagger - a^\dagger a(t)] + \Lambda_+ [a^\dagger (t) a - aa^\dagger (t)] + \text{H.c.}
\]

(2.354)

The coefficients \( \Lambda_\pm \) are determined by the fluctuation spectra of \( V_1 \)

\[
S(\pm \omega_\ell) = \frac{1}{\hbar^2} \int_0^\infty d\tau \langle V_1 (\tau) V_1 (0) \rangle_{ss} e^{\pm i \omega_\ell \tau}
\]

(2.355)

and read

\[
\Lambda_\pm = z_0^2 S(\pm \omega_\ell) = \frac{z_0^2}{\hbar^2} \int_0^\infty d\tau \langle V_1 (\tau) V_1 (0) \rangle_{ss} e^{\pm i \omega_\ell \tau},
\]

(2.356)

where we have introduced \( z_0 = \sqrt{\hbar/(2m\omega_\ell)} \). Here the subscript “ss” stands for stationary state and it means that the expectation values are evaluated for the internal degrees of freedom in their stationary state, which is the dark state \( \rho_d \) in Eq. (2.337).

From Eq. (2.354), we can obtain a master equation for the populations \( \mu_{n,n} = \langle n | \mu (t) | n \rangle \). Defining the rates

\[
A_\pm = 2 \text{Re} (\Lambda_\pm)
\]

(2.357)

this master equation can be written as

\[
\frac{d}{dt} \mu_{n,n} = (n + 1) A_- \mu_{n+1,n+1} - [(n + 1) A_+ + n A_-] \mu_{n,n} + n A_+ A_{n-1,n-1}.
\]

(2.358)

This is actually the same equation as Eq. (2.317) above, which we obtained from the simplified rate treatment. Completely analogous to Eq. (2.318) we obtain an equation of motion for the mean occupation number \( \bar{n} (t) = \langle n (t) \rangle \) of the cooled mode

\[
\frac{d}{dt} \langle n (t) \rangle = \frac{d}{dt} \sum_{n=0}^\infty n \mu_{n,n} = - (A_- - A_+) \langle n (t) \rangle + A_+,
\]

(2.359)
where
\[ W = A_- - A_+ \quad \text{(2.360)} \]
is the cooling rate. Again we obtain cooling for \( W > 0 \) and the final occupation number that is reached for cooling is given by
\[ \langle n \rangle_\infty = \frac{A_+}{A_- - A_+}. \quad \text{(2.361)} \]
The fluctuation spectra \( S(\pm \omega) \) from Eq. (2.355) can actually be evaluated analytically for the three-level system we considered at the beginning of this section. This allows us to obtain an analytical expression for the final occupation number from Eq. (2.361) and obtains \[93\]
\[ A_\mp = \frac{1}{4} \left( \frac{\Omega_1 \Omega_2}{\Omega} \right)^2 \frac{\eta_{\text{eff}}^2 \Gamma \omega_z^2}{\Omega^2/4 - \omega_z (\omega_z - \Delta_{\text{EIT}})} + \Gamma^2 \omega_z^2/4, \quad \text{(2.362)} \]
where we have introduced the shorthand notation \( \eta_{\text{eff}} = \eta_1 \cos \theta_1 - \eta_2 \cos \theta_2 \). The steps of the evaluation of the rates are sketched in App. A. One can then also evaluate the theoretical asymptotic mean occupation number from Eq. (2.361) and obtains \[93\]
\[ \langle n \rangle_\infty = \frac{4 \Omega^2/4 - \omega_z (\omega_z - \Delta_{\text{EIT}})^2 + \Gamma^2 \omega_z^2}{4 \omega_z |\Delta_{\text{EIT}}| (\Omega^2 - 4 \omega_z^2)}. \quad \text{(2.363)} \]
Finally, using the rates from Eq. (2.362) we can cast the master equation (2.354) into the form
\[ \dot{\mu}(t) = -i[\Delta \omega a^\dagger a, \mu(t)] + A_- [a \mu(t) a^\dagger - \frac{1}{2} \{a^\dagger a, \mu(t)\}] + A_+ [a^\dagger \mu(t) a - \frac{1}{2} \{aa^\dagger, \mu(t)\}]. \quad \text{(2.364)} \]
We see that the cooling not only introduces dissipation on the mode but it also causes a shift of \( \Delta \omega = \text{Im}(\Lambda_-) + \text{Im}(\Lambda_+) \) of the mode’s frequency. This shift can also be evaluated analytically and yields
\[ \Delta \omega_z = \eta_{\text{eff}}^2 \left( \frac{\Omega_1 \Omega_2}{\Omega} \right)^2 \frac{8 \omega_z^2 \Delta_{\text{EIT}} \left[ \left( \Omega_z^2 - 4 \omega_z^2 \right)^2 - 4 \Gamma^2 \omega_z^2 - 16 \omega_z^2 \Delta_{\text{EIT}} \right]}{[4 \Gamma^2 \omega_z^2 + (\Omega_z^2 + 4 \omega_z (\Delta_{\text{EIT}} - \omega_z))^2][4 \Gamma^2 \omega_z^2 + (\Omega_z^2 - 4 \omega_z (\Delta_{\text{EIT}} + \omega_z))^2]. \quad \text{(2.365)} \]
Given that we are in the cooling regime \( A_- > A_+ \) we can write the master equation (2.364) in terms of the cooling rate \( W \equiv 2 \kappa \) and the asymptotic occupation number \( \langle n \rangle_\infty \equiv \bar{n} \) from Eqs. (2.360) and (2.361), respectively. The resulting equation reads
\[ \dot{\bar{n}}(t) = -i[\Delta \omega a^\dagger a, \mu(t)] + 2 \kappa (\bar{n} + 1) [a \mu(t) a^\dagger - \frac{1}{2} \{a^\dagger a, \mu(t)\}] + 2 \kappa \bar{n} [a^\dagger \mu(t) a - \frac{1}{2} \{aa^\dagger, \mu(t)\}]. \quad \text{(2.366)} \]
The dissipator is that of the damped quantum harmonic oscillator [69]. This form of the cooling master equation is quite instructive since we can extract the final occupation number \( \bar{n} \) and the rate \( 2 \kappa \) at which energy is dissipated directly from the equation. The effect of the dissipator is that it takes the populations of the state \( \mu(t) \) to a thermal state with mean occupation \( \bar{n} \) at a rate \( 2 \kappa \). The thermal state of an oscillator with mean occupation number \( \bar{n} \) can be written as \[53\]
\[ \rho_\beta = \sum_{n=0}^{\infty} \frac{\bar{n}^n}{(1 + \bar{n})^{1+n}} |n\rangle\langle n|, \quad \text{(2.367)} \]
where the mean occupation number is given by
\[ \bar{n} = \frac{1}{e^{\beta \omega} - 1}. \quad \text{(2.368)} \]
Here $\beta$ is the inverse temperature $\beta = (k_B T)^{-1}$ with $k_B$ the Boltzmann constant.

It is interesting to note that Doppler cooling in a traveling or standing wave also obeys the master equation (2.364) but with different coefficients [94].

In order to illustrate the validity of the above derivation, we compare the effective dissipative dynamics of the mode given by Eq. (2.364) with the dynamics of the full system including the three-level system given in Eq. (2.343) in a numerical simulation. We consider one mode of motion of a single trapped $^{24}\text{Mg}^+$ ion with a mode frequency $\omega_z / 2\pi = 5\text{MHz}$. We consider $^{24}\text{Mg}^+$ because it has a suitable internal structure and EIT cooling has already been successfully implemented with it [91]. The Λ-system can be realized by the $|m_J = \mp 1/2 \rangle = |g_1, g_2 \rangle$ states of the $^2S_{1/2}$ ground state manifold of $^{24}\text{Mg}^+$ and one of the $^2P_{1/2}$ states, e.g. $|m_J = +1/2 \rangle$ [91]. Here, we neglect the $|m_J = -1/2 \rangle$ $P$ level for simplicity. The branching ratio for the two decay channels is $\Gamma_{g_1} = 2\Gamma_{g_2}$ and the total decay rate is $\Gamma/2\pi = 41.4\text{MHz}$. In part a) of Fig. 2.8 we set $\Omega_1/2\pi = 35\text{MHz}$, $\Omega_2/2\pi = 11\text{MHz}$ and $\Delta_{\text{EIT}}/2\pi = -100\text{MHz}$. The bare Lamb-Dicke parameter in this case is $\eta \approx 0.15$ for both lasers and we assume the beams to enclose angles $\theta_1 = \frac{2}{5}\pi$ and $\theta_2 = \frac{3}{5}\pi$ with the axis of motion. According to the rates in Eq. (2.362) we expect a cooling rate $2\kappa/2\pi = 4.6\text{kHz}$. We consider initial conditions $\rho(0) = \rho_{\text{d}} \otimes \rho_{\beta}$ with the internal levels in the dark state and the motional degree of freedom in thermal state with mean phonon number $\bar{n}(0) = 1$. We truncate the motional Hilbert space at $n_{\text{max}} = 15$ excitations. The full line illustrates the full dynamics and the circles the effective dynamics, displaying very good agreement between the two descriptions.

![Figure 2.8.](image-url)

Figure 2.8.: The figure shows the mean occupation number $\langle \bar{n}(t) \rangle$ of a single trapped $^{24}\text{Mg}^+$ ion. The solid line shows the dynamics of the full system including the internal levels given by Eq. (2.343), circles show the effective dynamics according to Eq. (2.364). The remaining parameters are given in the text.

For the parameters of the simulation in Fig. 2.8 the final occupation number of the mode according to Eq. (2.363) is $\bar{n}_\infty \approx 0.07$. This is considerably lower than the final occupation numbers of $^{40}\text{Ca}^+$ after Doppler cooling reported in Eq. (2.309). Indeed, in [89] the $y$- and $z$-mode of the trapped $^{40}\text{Ca}^+$ ion were cooled from their Doppler temperatures, theoretically $\bar{n}_{y,D} \approx 6$ and $\bar{n}_{z,D} \approx 3$, to $\bar{n}_{y,\text{EIT}} \approx 0.18$ and $\bar{n}_{z,\text{EIT}} \approx 0.1$. Depending on the ion masses and frequencies these occupation numbers can be even lower.

In addition to the small occupation numbers that can be achieved, EIT cooling can yield higher cooling rates than other cooling schemes such as resolved sideband cooling [91, 92]. In [91] and [92] cooling rates of several kHz were reported in the cooling mixed- and single-species crystals. Another benefit of EIT cooling is that several modes can be cooled at the same time if
their frequencies are close enough to accommodate their red sidebands in the narrow feature of the absorption spectrum in Fig. 2.7 b).

### 2.6.3. Sympathetic cooling of mixed species crystals of trapped ions

In this section we want to generalize the description of EIT cooling of the previous section from a single ion to Coulomb crystals incorporating several ions of possibly different species. Specifically, we derive the cooling master equation for a crystal of ions and generalize it to the case of mixed-species crystals. This second form allows us to describe sympathetic cooling of a mixed-species crystals, where the cooling light only interacts with one species of the crystal and this species cools the whole crystal.

Let us consider a string of \( N \) trapped ions of equal mass and internal structure. We assume the internal levels of each ion are described by the Hamiltonian in Eq. (2.324) and that they are interacting with the laser configuration depicted in Fig. 2.7 a). Again we restrict the considerations to the \( z \)-direction of motion. Analogous to Eq. (2.342) the starting point is the Liouvillian

\[
\dot{\rho} = \mathcal{L}^{(N)} \rho = -\frac{i}{\hbar} [H_{\text{at}}^{(N)} + H_{\text{m}}^{(N)} + V^{(N)}, \rho] + \mathbb{D}^{(N)} \rho, \tag{2.369}
\]

where the superscript \( (N) \) indicates generalization of the Hamiltonians and the dissipator to \( N \) ions. Note that we have already incorporated the motional Hamiltonian into the Liouvillian. Assuming that we have already moved to an interaction picture with respect to \( H_0 = -\hbar \sum_{j=1}^{N} \sum_{n=1,2} \omega_{n,j} |g_s\rangle \langle g_s| \), where \( |g_s\rangle \langle g_s| \) acts on the internal levels of ion \( j \), the internal levels are described by

\[
H_{\text{at}}^{(N)} = -\hbar \Delta_{\text{EIT}} \sum_{j=1}^{N} |g_s\rangle \langle g_s|, \tag{2.370}
\]

generalizing Eq. (2.327) to \( N \) ions. According to Eq. (2.57) the motional Hamiltonian reads

\[
H_{\text{m}}^{(N)} = \sum_{n=1}^{N} \hbar \omega_n a_n^\dagger a_n, \tag{2.371}
\]

where \( a_n \) and \( a_n^\dagger \) are the ladder operators of mode \( n \) with frequency \( \omega_n \). The laser-ion interaction is described by

\[
V^{(N)} = \sum_{j=1}^{N} \sum_{k=1,2} \left( \frac{\Omega_j}{2} |e\rangle \langle g_s| e^{ikz_j \cos \theta} + \text{H.c.} \right), \tag{2.372}
\]

which is the Hamiltonian of Eq. (2.328) for \( N \) ions. The dissipator now takes the form

\[
\mathbb{D}^{(N)} \rho = -\frac{\Gamma}{2} \sum_{j=1}^{N} \{ |e\rangle \langle e|, \rho \}
+ \frac{\Gamma}{2} \sum_{j=1}^{N} \sum_{k=1,2} \int_{-1}^{1} d(\cos \theta) \mathcal{N} (\cos \theta) |g_s\rangle \langle g_s| e^{ikz_j \cos \theta} \rho e^{-ikz_j \cos \theta} |e\rangle \langle e| \langle g_s| \rangle \tag{2.373}
\]

Recalling Eqs. (2.52) and (2.55) we write the \( z \)-positions as

\[
z_j = z_j^0 + q_{jz}, \quad \text{where} \quad q_{jz} = \sum_{n=1}^{N} B_{jn} Q_{nz} = \sum_{n=1}^{N} B_{jn} \sqrt{\frac{\hbar}{2 m \omega_n}} (a_n + a_n^\dagger) \tag{2.374}
\]

and follow the same procedure as for the single ion case. Assuming that all modes are in the Lamb-Dicke regime, we expand the laser-ion interaction and the momentum transfer term in the dissipator to second order in the Lamb-Dicke parameters

\[
\eta_{kn} = k_{n} \sqrt{\frac{\hbar}{2 m \omega_n}} \ll 1. \tag{2.375}
\]
where the motion state onto the populations of the modes are equally illuminated. The projector of the external degrees of freedom \( P \) projects the internal levels of the ions to the dark state \( \rho = (\mathcal{L}_0^{(N)} + \mathcal{L}_1^{(N)} + \mathcal{L}_2^{(N)}) \rho \).

(2.376)

We have

\[
\mathcal{L}_0^{(N)} \rho = -\frac{i}{\hbar} [H_\text{int}^{(N)} + V_0^{(N)}, \rho] + \mathcal{D}_0^{(N)} \rho,
\]

(2.377)

where \( V_0^{(N)} \) and \( \mathcal{D}_0^{(N)} \) are obtained by setting the exponentials in Eqs. (2.372) and Eq. (2.373) equal to 1 and in the case of \( \mathcal{D}_0^{(N)} \) we additionally use the normalization of \( \mathcal{N}(\cos \theta) \), Eq. (2.340). The first and second order contributions take the form

\[
\mathcal{L}_1^{(N)} \rho = -\frac{i}{\hbar} \sum_{n=1}^{N} [V_{1,n} Q_{n,z}, \rho],
\]

(2.378)

\[
\mathcal{L}_2^{(N)} \rho = -\frac{i}{\hbar} \sum_{n,m=1}^{N} [V_{2,nm} Q_{n,z} Q_{m,z}, \rho] + \mathcal{D}_0^{(N)} \rho.
\]

(2.379)

The interaction terms \( V_{1,n} \) and \( V_{2,nm} \) read

\[
V_{1,n} = \frac{i \hbar}{2} \sum_{j=1}^{N} \sum_{s=1,2} B_{j n} k_s \cos \theta_s \Omega_s \left( |g_s \rangle \langle e| - |e \rangle \langle g_s| \right),
\]

(2.380)

\[
V_{2,nm} = -\frac{\hbar}{4} \sum_{j=1}^{N} \sum_{s=1,2} B_{j n} B_{j m} k_s^2 \cos \theta_s \Omega_s \left( |g_s \rangle \langle e| + |e \rangle \langle g_s| \right)
\]

(2.381)

and the second order dissipator reads

\[
\mathcal{D}_2^{(N)} \rho = \sum_{j=1}^{N} \sum_{s=1,2} \alpha_j^2 k_s^2 \Gamma_s |g_s \rangle \langle g_s| \left( q_{jz} \rho q_{jz} - \frac{1}{2} q_{jz}^2 \rho - \frac{1}{2} \rho q_{jz}^2 \right) |e \rangle \langle e|. \]

(2.382)

Note that the first order terms vanish and that we have not expanded the \( q_{jz} \) in terms of the normal modes in order to keep the expression more transparent.

The scenario is basically the same as in the single ion case: To zeroth order in the Lamb-Dicke factor there is no coupling between internal levels and motion and the cooling is brought about by the terms \( \mathcal{L}_1 \) and \( \mathcal{L}_2 \). In order to obtain an equation of motion for the cooling the second order contributions can be neglected assuming that the cooling dynamics is much slower than the relaxation of the internal states. In order to apply Eq. (2.353) we define a projection operator \( P^{(N)} \) to the zero eigenvalue subspaces of \( \mathcal{L}_0^{(N)} \)

\[
P_0^{(N)} \rho = \left( P_1^{(N)} \otimes P_E^{(N)} \right) \rho.
\]

(2.383)

The operator \( P_1^{(N)} \) projects the internal levels of the dark state to the dark state

\[
P_1^{(N)} \rho = \rho_d^{(N)} \otimes \text{Tr}_I \rho = \otimes_{j=1}^{N} |\psi_d \rangle \langle \psi_d| \otimes \mu^{(N)}.
\]

(2.384)

where \( \text{Tr}_I \) again denotes the trace over the internal states of the ions and we defined \( \mu^{(N)} = \text{Tr}_I \rho \).

The above equation implies that all ions reach the same stationary state, which assumes that all ions are equally illuminated. The projector of the external degrees of freedom \( P_E^{(N)} \) projects the motional state onto the populations of the modes

\[
P_E^{(N)} (\rho_d^{(N)} \otimes \mu^{(N)}) = \rho_d^{(N)} \otimes_{m=1}^{N} \left[ \sum_{n=0}^{\infty} \langle n | \text{Tr}_{\text{modes}} \mu^{(N)} | n \rangle m | n \rangle_m \langle n \rangle \right] = \rho_d^{(N)} \otimes \mu.
\]

(2.385)
It is easy to check that the states \( P_0^{(N)} \rho \) live in the subspace of the zero eigenvalue of \( \mathcal{L}_0^{(N)} \).

We are now in a position to use Eq. (2.353) to obtain the dynamics in the “relevant” subspace \( P_0^{(N)} \) substituting \( P_0 \rightarrow P_0^{(N)} \), \( \mathcal{L}_0 \rightarrow \mathcal{L}_0^{(N)} \) and so on. Following the same steps as in the single ion case as outlined in App. A, a lengthy calculation yields the effective master equation

\[
\dot{\rho}(t) = \sum_{\mu} \left( \Lambda_{-\mu} [a_n \mu(t) a_n^\dagger - a_n^\dagger a_n \mu(t)] + \Lambda_{+\mu} [a_n^\dagger \mu(t) a_n - a_n a_n^\dagger \mu(t)] \right) + \text{H.c.,}
\]  

(2.386)

where the coefficients \( \Lambda_{\pm \mu} \) read

\[
\Lambda_{\pm \mu} = \frac{2 z_{0,\mu}}{\hbar} \int_0^\infty d\tau \langle V_{1,\mu}(\tau) V_{1,\mu}(0) \rangle_{ss} e^{\pm i \omega_\mu \tau}.
\]

(2.387)

In the above expression we have introduced \( z_{0,\mu} = \sqrt{\hbar/(2m \omega_\mu)} \). Inserting the definition for \( V_{1,\mu} \) from Eq. (2.380) and realizing that the correlation function between internal levels of different ions is zero, we actually find that the rates \( \Lambda_{\pm \mu} \) can be written in terms of the single particle fluctuation spectra \( S(\pm \omega) \) from Eq. (2.355)

\[
\Lambda_{\pm \mu} = \frac{2 z_{0,\mu}^2}{\hbar^2} \sum_j B_{jn}^2 \int_0^\infty d\tau \langle V_1(\tau) V_1(0) \rangle_{ss} e^{\pm i \omega_\mu \tau} = z_{0,\mu}^2 S(\pm \omega_\mu) \sum_j B_{jn}^2,
\]

(2.388)

where we have used \( V_1 \) from Eq. (2.347).

If all ions participate in the cooling process, we can sum over all \( j \) in the above rates and can substitute \( \sum_j B_{jn}^2 = 1 \). It is interesting to analyze the cooling rate if not all ions interact with the cooling laser. Then, the sum on the right-hand side of Eq. (2.388) only extends over those ions \( j \) which are actually subject to the cooling laser. In this case it is still possible to cool a whole crystal if for each mode, at least one of the ions participating in cooling has a non-vanishing amplitude. Depending on the amplitudes this might lead to different time scales for the cooling of the modes.

Let us now comment on the validity of the above treatment. By defining the projector \( P_0^{(N)} \) in Eq. (2.383), which allowed us to derive Eq. (2.386), we are assuming that the normal mode picture is valid during the cooling process and that the cooling does not introduce correlations between the modes. We considered cooling in the Lamb-Dicke regime and in this regime it is usually assumed that the modes are cooled individually [90] such that our treatment is justified. The underlying assumption is that in the Lamb-Dicke regime the sideband spectrum reduces to the carrier and first red- and blue-sideband transitions of the individual modes. Then, in a scattering event the occupation number of only one mode is changed by one quantum. Processes coupling the modes or changing the occupation number of more than one mode are of higher order in the Lamb-Dicke parameter and are neglected. However, cooling broadens the resonances of the modes in frequency space and if these resonance overlap, we cannot consider the modes to be independent anymore. Hence, our treatment is restricted to the regime

\[
2 \kappa_n \ll |\omega_n - \omega_m| \quad \forall n, m,
\]

(2.389)

where \( 2 \kappa_n = 2 \text{Re}(\Lambda_{-n}) - 2 \text{Re}(\Lambda_{+n}) \) is the cooling rate of mode \( n \). Satisfying Eq. (2.389) we make sure the modes can be regarded as isolated from each other. An interesting question is whether we are strictly bound to the regime in Eq. (2.389). This could be checked by numerically integrating the full master equation Eq. (2.369). But while this is an interesting topic in its own right, it is beyond the scope of our treatment here.

Let us finally come to the case of a mixed-species crystal. We consider the case of a crystal incorporating two species, where one species interacts with the cooling light while the cooling
light is highly off-resonant with all transitions in the second species. The cooling process then involves only the ions that interact with the applied light. If the normal mode picture is valid, the cooling extracts motional energy from the whole crystal due to the nonlocal nature of the normal modes. In order to achieve efficient cooling for all modes, one has to take care that the cooled ions have an appreciable amplitude for all modes. On the other hand, the cooling rate cannot be stronger than the Coulomb interaction between ions, which is weak in comparison to the trap potential in the radial directions. Cooling a subset of the ions too strongly would decouple the cooled ions from the rest of the crystal, and a description of the ions in terms of local harmonic oscillators is then more appropriate [17, 33].

Sympathetic cooling has been applied in several laboratories, see e.g. [91,95,96], also including EIT cooling. The cooling and heating rates for sympathetic EIT cooling in a mixed species crystal are obtained from Eq. (2.388). In this case the normal mode amplitudes $B_{jn}$ of a single species crystal have to be substituted by that of a mixed species crystal $\tilde{B}_{jn}$ from Eq. (2.69) and one has to take into account that according to Eq. (2.79) the “ground state wave packet spread” $\tilde{z}_{0,n}$ now depends on $j$

$$\tilde{z}_{0,n} \rightarrow \tilde{z}_{0,jn} = \sqrt{\frac{\hbar}{2m_j\omega_n}}. \quad (2.390)$$

The rates $\Lambda_{\mp,n}$ from Eq. (2.388) for sympathetic cooling in a mixed-species crystal then become

$$\Lambda_{\mp,n} = \sum_j \tilde{B}_{jn}^2 \tilde{z}_{0,jn}^2 S(\pm \omega_n), \quad (2.391)$$

where the sum $\sum_j$ extends only over those ions interacting with the cooling lasers.

In order to obtain the highest cooling rates for sympathetic cooling, it is desirable to work with ions of similar masses. Then, the coefficients $\tilde{B}_{jn}$ do not differ too much from those of a single species crystal. If the difference in masses becomes very large, it can happen that the coefficients $\tilde{B}_{jn}$ become very small for some mode for one of the species such that motion of the ions effectively decouples. Then, no effective cooling of the crystal is possible [97]. Furthermore, for sympathetic cooling one has to make sure that the cooling ions possess an appreciable amplitude $B_{jn}$ for all modes to be cooled since otherwise we are facing the same effect mentioned before, namely, that cooling becomes inefficient or for a vanishing coupling cannot be realized at all for some modes.

In this section, we have discussed Doppler and EIT laser cooling. We discussed the cooling mechanisms as well as the achievable cooling rates and final temperatures. In particular, it should have become clear that the difference in scattering rates on the red- and blue-sidebands is decisive for efficient cooling. After reviewing the derivation of the master equation for EIT cooling of a single ion, we generalized the master equation to the cooling of crystals. Finally, we generalized the master equation further to mixed-species crystals such that it can also be used for the description of sympathetic cooling.

64
3. Two-dimensional spectroscopy for the study of Coulomb crystals

Two-dimensional (2D) spectroscopy is a spectroscopic technique originally developed in the context of nuclear magnetic resonance (NMR) experiments [30, 98, 99], where it was also first applied successfully. It is an extremely versatile tool for the investigation of complex systems and therefore has been adapted to various other fields of physics. It has been applied to investigate anharmonic molecular vibrational spectra in the infrared [100], electronic dynamics in molecular aggregates [31], a special case of which are the excitonic dynamics in photosynthetic pigment protein complexes [101, 102], structure and dynamics of polymers [103] as well as photochemical reactions [104].

The popularity of 2D spectroscopy is due to the wealth of information that its methods can reveal. For example, 2D spectroscopy can witness interactions in complex molecules and thus allows one to obtain a topology of interactions in the molecule. It facilitates the separation of interactions due to different physical origin to orthogonal frequency axes, which can be very helpful in unraveling spectra containing overlapping resonances. And also dynamical processes, as for example spin diffusion in solids, can be investigated using 2D spectroscopy.

On the other hand, experiments with trapped ions have experienced tremendous progress in the field of quantum computation and quantum simulation. While quantum computation experiments usually involve only a small number of ions, recent quantum simulations of spin systems involved up to 53 ions in linear Paul traps [23] and more than one hundred ions in Penning traps [62]. More than that, trapped ions have also become a testbed for the statistical mechanics of systems in and out of equilibrium [13, 17, 20–22, 28, 29, 105], where experiments often involve of the order of ten or even tens of ions. This increased complexity demands new experimental tools for the investigation of trapped ions.

As should have become clear from the introductory paragraph, 2D spectroscopy is a very powerful tool and in this chapter we present a protocol for the application of the technique to trapped ions. The protocol is designed to investigate nonlinear dynamics of ion Coulomb crystals and as such represents a new route to the investigation of the dynamics of crystals of trapped ions. We should remark that the application of 2D spectroscopy has been proposed and studied also in related work [106–108]. The cited references, however, consider different scenarios for the application of 2D spectroscopy than we do here.

In the previous chapter we have seen that the Coulomb interaction, which is intrinsically nonlinear, is usually linearized in the description of the dynamics of ions in a trap. In practice, this description agrees excellently with the observed dynamics and therefore the question arises when nonlinear dynamics actually occur in trapped-ion experiments. Nonlinearities can be added to a trap, for example, by applying a standing wave which induces a position dependent ac-Stark shift [33]. Such added nonlinearities have been proposed for the simulation of the Bose-Hubbard model [33] and the Frenkel-Kontorova model [14–16]. Nonlinear effects also play a crucial role in the linear-to-zigzag structural transition, where the ions in a crystal leave the linear structure and form a two-dimensional zigzag structure [109–111]. In the vicinity of the transition the usual harmonic treatment of the Coulomb potential breaks down and higher order terms start to play a role [32, 112]. Another example of nonlinear dynamics are cross-Kerr
type interactions due to third- and fourth order effects of the Coulomb potential that can lead to effective dephasing [113, 114]. Apparently, there are several instances of nonlinear dynamics in ion traps. In this chapter we will show how 2D spectroscopy can be used to study nonlinear interactions in ion traps and focus on two examples: the nonlinear interactions appearing in the vicinity of the linear-to-zigzag structural transition and resonant energy exchange between modes due to third-order effects of the Coulomb potential.

This chapter is organized as follows. We start with a brief introduction to 2D spectroscopy in the context of NMR followed by a discussion of the linear-to-zigzag transition of trapped ion crystals. Thereafter, we will turn to the calculation of nonlinear corrections to the Coulomb potential in the harmonic approximation. We then present our 2D spectroscopy protocol that is able to detect nonlinear interactions and illustrate the protocol on the basis of two examples. In the first example, we show how the onset of the linear-to-zigzag structural transition can be witnessed by 2D spectroscopy and in the second example, we show how resonant energy transfer between modes due to the third order-effects in the Coulomb potential can be studied with our protocol. We close the chapter with a summary of our results and future perspectives for 2D spectroscopy with trapped ions. The results of this chapter were published in [L1]. Accordingly, the presentation follows that in [L1].

3.1. Basic principles of Two-Dimensional Spectroscopy

In the introduction we have seen that 2D spectroscopy comes in different flavors and can reveal different information. In this section we want to introduce the basic structure of 2D spectroscopy experiments and discuss two realizations of 2D spectroscopy in the context of NMR experiments. In these two examples we will see how couplings between different spins can be witnessed by 2D spectroscopy and how different contributions of a Hamiltonian can be separated to different frequency axes, which can be used to unravel information that is contained in 1D spectra but cannot be retrieved from them. At the end of the section we will discuss the impact of different sources of noise on 2D spectra.

Before we dive into the analysis of 2D spectroscopy protocols, we should clarify what we mean by a two-dimensional spectrum. The somehow obvious answer is that we refer to a spectrum that has two frequency axes instead of one. The question now is how we obtain these two frequency axes. For example, one could think of an experiment where we excite a system with two fields of frequency $\omega_1$ and $\omega_2$ and record some signal describing the system’s response. Scanning the frequencies $\omega_1$ and $\omega_2$ and plotting the obtained signal we would obtain a two-dimensional spectrum. The 2D spectroscopy experiments that we want to discuss here are time-domain 2D spectroscopy experiments. This means we measure some signal $s(t_1, t_2)$ with a suitable segmentation of the time axis and then perform a Fourier transformation in both time variables. In this way we obtain a signal $S(\omega_1, \omega_2)$ as a function of two frequency variables.

This type of two-dimensional spectroscopy experiments can be understood as a special case of multidimensional spectroscopy [30, 103]. The general sequence of a multidimensional spectroscopy experiment is depicted in Fig. 3.1 a). After the system is prepared in an initial state $\rho_0$, it is irradiated by $n$ pulses of electromagnetic radiation separated by intervals of free evolution of variable length $t_i, i = 1, \ldots, n$. Let us denote the action of pulse $i$ by a superoperator $\hat{R}_i$ such that

$$\hat{R}_i \rho = R_i \rho R_i^\dagger,$$

where the operator $R_i$ describes the action of the pulse on a state $|\psi\rangle$. Similarly we associate a
3.1. Basic principles of Two-Dimensional Spectroscopy

Liouvillian $\mathcal{L}_i$

$$\mathcal{L}_i \rho = -\frac{i}{\hbar} [H_i, \rho] + \mathcal{D}_i \rho$$

(3.2)

with the free evolution period $i$ with Hamiltonian $H_i$ and a dissipator $\mathcal{D}_i$ that accounts for dissipative processes during $t_i$. The state $\rho(t_1, \ldots, t_n)$ after the sequence of $n$ pulses and free evolutions reads

$$\rho(t_1, \ldots, t_n) = e^{\mathcal{L}_{n} t_n} \cdots e^{\mathcal{L}_{1} t_1} \rho.$$  \hspace{1cm} (3.3)

At the end of the sequence some observable $M$ is measured yielding a signal

$$s(t_1, \ldots, t_n) = \text{Tr}[M \rho(t_1, \ldots, t_n)]$$ \hspace{1cm} (3.4)

that depends on the time variables $t_i$. A spectrum $S(\omega_i)$ containing spectral information on the Liouvillian $\mathcal{L}_i$ is obtained by a Fourier transformation of the signal with respect to time $t_i$. A two-dimensional spectrum $S(\omega_i, \omega_j)$ is obtained by Fourier transforming the signal $s(t_1, \ldots, t_n)$ in the time variables $t_i$ and $t_j$

$$S(\omega_i, \omega_j) = \int_{-\infty}^{\infty} dt_i e^{-i\omega_i t_i} \int_{-\infty}^{\infty} dt_j e^{-i\omega_j t_j} s(t_1, \ldots, t_n).$$ \hspace{1cm} (3.5)

In the same way higher order spectra can be obtained. We will use the short-hand notation $S(\omega_1, \omega_2) = \mathcal{F}[s(t_1, t_2)]$ for the two-dimensional Fourier transform in the following.

Here, we want to focus on two-dimensional spectra. The typical sequence of an NMR time domain 2D spectroscopy experiment is illustrated in Fig. 3.1 b) [30]. Here, after preparation of the initial state there is a first evolution period $t_1$ under a Liouvillian $\mathcal{L}_1$ which is called the evolution time. The time interval $t_m$ between the free evolution times typically consists of one or more pulses and possibly free evolutions described by a Liouvillian $\mathcal{L}_m$ and is called mixing time. The last interval of length $t_2$ is called detection time. This is because in NMR experiments the detected signal is the free induction decay of the magnetization that has built up in the course of the experiment recorded during $t_2$. Although naming conventions might be different, this is the general sequence for a 2D spectroscopy experiment. The type of information that one obtains in the spectrum, neglecting noise for the moment, depends on the Hamiltonians $H_i$ in the different evolution periods, which we will see in more detail in the next section.
3.1.1. Correlation spectroscopy

Correlation spectroscopy (COSY) is a method to identify couplings between nuclear spins commonly used in NMR [30]. Here, we will examine the simplest example of COSY which considers two weakly coupled nuclear spins. We will see that the coupling between the two spins leads to off-diagonal peaks in the 2D spectrum, which illustrates how 2D spectroscopy can be used to witness couplings in a system of interacting spins. The case we consider is that of homonuclear correlation spectroscopy, i.e. we consider two nuclear spins of the same species.

In a magnetic field $B_0$ the levels of a spin are separated in energy by the Larmor frequency $\omega_0 = \gamma_s B_0$, where $\gamma_s$ is the gyromagnetic ratio of the spin under consideration. Let us consider two spin-$1/2$ nuclei in a magnetic field that is directed along the $z$-direction. Keeping our previous notation, the bare spin Hamiltonian can be written as

$$H_0 = \hbar \omega_0 \sum_{k=1,2} \sigma_z^k, \quad (3.6)$$

where $\omega_0 = \gamma_s B_0$ and $\sigma_z^k$ is the Pauli operator of spin $k$. In a molecule or solid the spins interact with their surroundings which leads to a local shift of their resonance frequency, so-called chemical shifts [30, 103]. Their free Hamiltonian therefore becomes

$$H_0 = \sum_{k=1,2} \hbar \omega_{0,k} \sigma_z^k, \quad (3.7)$$

where $\omega_{0,k}$ is the shifted resonance frequency of spin $k$.

Let us now assume that the spins are initially in a thermal equilibrium state

$$\rho_\beta = \frac{e^{-\beta H_0}}{Z} = \frac{e^{-\beta H_0}}{\text{Tr}[e^{-\beta H_0}]}, \quad (3.8)$$

where $Z = \text{Tr}[e^{-\beta H_0}]$ is the partition function. We consider the high temperature limit $\beta \hbar \omega_{0,k} = \hbar \omega_{0,k}/(k_B T) \ll 1$ and expand the state as

$$\rho_\beta \approx \frac{1}{Z} \left( 1 - \sum_k \frac{\hbar \beta \omega_{0,k}}{2} \sigma_z^k \right). \quad (3.9)$$

We will now investigate how this initial state evolves under the typical COSY sequence. Since the unit operator is invariant under the dynamics, only the second term plays a role for the dynamics. In order to keep the notation as clear as possible, we adopt the somewhat unfamiliar convention sometimes used in NMR that the initial state is just written as [30, 103]

$$\rho_\beta = \sum_k \sigma_z^k. \quad (3.10)$$

Note that writing the state in the above form is consistent with the high temperature limit if we assume that the chemical shifts are much smaller than the Larmor frequency $\omega_0$. The COSY protocol is illustrated in Fig. 3.2 a). The preparation period consists of a $\pi/2$ rotation about the $x$-axis applied to the thermal equilibrium state which is followed by the first evolution period $t_1$. The mixing period consists of a rotation by a variable angle $\varphi$ around the $x$-axis. It is followed by the second evolution period $t_2$. The measured quantity in NMR experiments is the free induction decay. Using quadrature detection the signal is given by [30]

$$s(t_1, t_2) = \langle \rho(t_1, t_2) S^+ \rangle = \langle \rho(t_1, t_2) \sum_k \sigma_z^k \rangle. \quad (3.11)$$
3.1. Basic principles of Two-Dimensional Spectroscopy

Figure 3.2.: Correlation spectroscopy. Part a) shows the simplest COSY pulse sequence. Part b) shows the positive frequency part of a 2D spectrum of two uncoupled nuclear spins whose frequencies are separated by the difference of their chemical shifts $\omega_1$ and $\omega_2$. This difference is given by the distance of the peaks along each of the frequency axes. A non-vanishing coupling $J$, see Eq. (3.13), between the nuclear spins leads to the COSY spectrum in part c). The peaks in the multiplets are separated by $2J$ along each of the frequency axes. The cross-peaks far from the main diagonal indicate coupling between the two spins. If the resolution is not sufficient to resolve $J$, and thus the multiplets, we obtain a spectrum of the form presented in part d). The cross peaks still witness the coupling between the spins. Figure derived from [30].

Note that although $\sigma_k^+$ is not Hermitian, we may write it as a linear combination of two Hermitian operators $\sigma_k^+ = \sigma_k^x + i\sigma_k^y$ which are valid observables. In particular, the measured observable is a combined measurement of the transverse magnetization in $x$ and $y$.

The Hamiltonian in the evolution periods is obtained by adding the coupling to $H_0$ in Eq. (3.7). For weakly coupled spins, i.e. when the coupling $J \ll |\omega_{0,1} - \omega_{0,2}|$, the full Hamiltonian may be written as [30]

$$ H = \sum_{k=1,2} \hbar \frac{\omega_{0,k}}{2} \sigma_k^x + \hbar J \frac{1}{2} \sigma_1^z \sigma_2^z, \quad (3.12) $$

where interactions between the spins in the transverse directions have been neglected. Note again that we restrict our analysis to the case of two nuclear spins. The pulses are more conveniently described in an interaction picture that rotates with the frequency of the applied radiation $\omega_{rf}$ which often lies in the radio frequency domain. We move to an interaction picture with respect to the Hamiltonian $H_0 = \sum_{k=1,2} \hbar \omega_{0,k} \sigma_k^z$ such that the Hamiltonian becomes

$$ H = \sum_k \hbar \frac{\omega_k}{2} \sigma_k^x + \hbar J \frac{1}{2} \sigma_1^z \sigma_2^z, \quad (3.13) $$

where $\omega_k = \omega_{0,k} - \omega_{rf}$. Note that the weak coupling condition $J \ll |\omega_{0,1} - \omega_{0,2}|$ is equivalent to $J \ll |\omega_1 - \omega_2|$. Let us now proceed to analyze the dynamics. After the preparation pulse the state of the spins reads

$$ \rho(t_p) = -\sum_{k=1,2} \sigma_k^y. \quad (3.14) $$
This initial state evolves for a period $t_1$ under the Hamiltonian in Eq. (3.12) and then reads

$$
ρ(t_1) = \sum_k [-σ_k^x \cos(\tilde{ω}_kt_1) + σ_k^y \sin(\tilde{ω}_kt_1)] \cos(Jt_1)
+ \sum_{k,l} [σ_k^l σ_l^x \sin(\tilde{ω}_kt_1) + σ_k^l σ_l^y \cos(\tilde{ω}_kt_1)] \sin(Jt_1).
$$

(3.15)

The mixing period only consists of a single rotation around the $x$-axis by an angle $φ$. Here, we consider $φ = \frac{π}{2}$ such that the state before the second evolution period reads

$$
ρ(t_1) = \sum_k [-σ_k^x \cos(\tilde{ω}_kt_1) + σ_k^y \sin(\tilde{ω}_kt_1)] \cos(Jt_1)
- \sum_{k,l} [σ_k^l σ_l^x \sin(\tilde{ω}_kt_1) + σ_k^l σ_l^y \cos(\tilde{ω}_kt_1)] \sin(Jt_1).
$$

(3.16)

Considering the signal in Eq. (3.11), we see that the first term in the first sum and the second term in the second sum do not contribute to the measured signal. We can therefore neglect them. It is now interesting to analyze the remaining terms. The second term in the first sum only involves single particle coherences. These will evolve in time with the same frequencies $±\tilde{ω}_k ± J$ during the second time evolution as they did during the first, where the previous expression refers to all four combinations. Restricting our considerations to positive frequencies we obtain four contributions located at $(\tilde{ω}_1, \tilde{ω}_2) = (\tilde{ω}_k ± J, \tilde{ω}_k ± J)$ for each value of $k$. Two peaks appear on the diagonal and two off the diagonal. Since we consider the weak coupling regime $J \ll |\tilde{ω}_1 - \tilde{ω}_2|$, the off-diagonal contributions are close to the main diagonal.

The more interesting contribution for us is the first term in the second sum. Comparing Eqs. (3.15) and (3.16) we see that coherence has been exchanged from spin $k$ to spin $l$ by the second $\frac{π}{2}$-pulse. Thus, the contributions that evolved with $±\tilde{ω}_k ± J$ during the first period evolve with $±\tilde{ω}_k ± J$, where $k \neq l$, during the second. These terms will produce contributions at points $(\tilde{ω}_1, \tilde{ω}_2) = (\tilde{ω}_k ± J, \tilde{ω}_l ± J)$, $k \neq l$, in the spectrum. These peaks are located off the diagonal but share each of their coordinates with one of diagonal peaks. Due to the weak coupling $J \ll |\tilde{ω}_1 - \tilde{ω}_2|$ these so-called cross peaks appear well off the main diagonal. The appearance of these terms is a direct consequence of the coupling term $J$ in the Hamiltonian in Eq. (3.12).

In order to make the above considerations more quantitative, we compute the state of the system after the second evolution period. Considering only terms that contribute to the signal we arrive at the state

$$
ρ(t_1, t_2) = \sum_k \sin(\tilde{ω}_kt_1) \cos(Jt_1) \left[ σ_k^x \cos(\tilde{ω}_kt_2) + σ_k^y \sin(\tilde{ω}_kt_2) \right] \cos(Jt_2)
+ \sum_{k,l} \sin(\tilde{ω}_kt_1) \sin(Jt_1) \left[ σ_l^x \cos(\tilde{ω}_kt_2) + σ_l^y \sin(\tilde{ω}_kt_2) \right] \sin(Jt_2).
$$

(3.17)

According to Eq. (3.11) the measured signal is then given by

$$
\mathcal{S}(t_1, t_2) = \sum_k \sin(\tilde{ω}_kt_1) \cos(Jt_1) e^{i\tilde{ω}_kt_2} \cos(Jt_2) + \sum_{k,l} \sin(\tilde{ω}_kt_1) \sin(Jt_1) e^{i\tilde{ω}_kt_2} \sin(Jt_2).
$$

(3.18)

A two-dimensional Fourier transform of the above signal confirms our previous considerations. From the first term we obtain contributions at $(\tilde{ω}_1, \tilde{ω}_2) = (±\tilde{ω}_k ± J, ±\tilde{ω}_k ± J)$ which are located at or close to the main diagonal $(J \ll |\tilde{ω}_1 - \tilde{ω}_2|)$ in the positive frequency quadrant. The second term yields peaks at $(\tilde{ω}_1, \tilde{ω}_2) = (±\tilde{ω}_k ± J, ±\tilde{ω}_l ± J)$ with $k \neq l$.

We illustrate the above considerations in Fig. 3.2 b) - d) where we schematically show the absolute value of the positive frequency part of a 2D spectrum given by a signal of the form
in Eq. (3.18). As we discussed, the peak structure consists of multiplets at and close to the diagonal and the off-diagonal multiplets indicate coupling between the spins.

Note that the obtained spectra are in general complex quantities and contain phase information. This can lead to different peak structures. We will not discuss these features here because we focus on absolute value spectra for the proposed version of 2D spectroscopy with trapped ions. Furthermore, noise and decoherence will affect the peak structure which we will discuss later. Detailed treatments of these topics may be found in [30, 103].

3.1.2. Separation of interactions

The second example of a 2D spectroscopy protocol we want to consider is the separation of interactions. We will show how 2D spectroscopy allows us to separate interactions of different physical origin to orthogonal frequency axes. In this way one can, for example, separate overlapping resonances that cannot be resolved in 1D spectra. Let us again consider the case of weak homonuclear coupling, i.e. two weakly coupled nuclear spins of the same species. The starting point is again the Hamiltonian of Eq. (3.13)

\[
H = \sum_{k=1,2} \left( \frac{\hbar}{2} \sigma^+_k + \frac{\hbar}{2} J \sigma^+_k \sigma^-_k \right).
\]  

(3.19)

One pulse sequence that leads to a separation of interactions is shown in Fig. 3.3 a). As in the case of correlation spectroscopy the preparation period consists of a single \( \pi \)-pulse along the \( x \)-direction. Accordingly, we obtain the same initial state as before, given in Eq. (3.14). Then after half of the first evolution period, i.e. \( t_1/2 \), a \( \pi \)-pulse along the \( y \)-direction is added. There is no mixing pulse or pulse sequence in this case. By adding the \( \pi \)-pulse along \( y \) the effective Hamiltonians in the evolution periods \( t_1 \) and \( t_2 \) read

\[
H_1 = \frac{\hbar}{2} \sigma^-_1 \sigma^+_2,
\]

(3.20)

\[
H_2 = \sum_{k=1,2} \left( \frac{\hbar}{2} \sigma^+_k + \frac{\hbar}{2} J \sigma^+_k \sigma^-_k \right).
\]

(3.21)

Obviously, during the first evolution period only the scalar coupling \( J \) plays a role while both the chemical shifts and scalar couplings appear in the second time evolution. For an initial state \( \rho(t_p) \) as in Eq. (3.14) the pulse sequence in Fig. 3.3 a) leads to a final state

\[
\rho(t_1, t_2) = \sum_k \left[ -\sigma^+_k \cos(Jt_1) \cos(\bar{\omega}_k t_2) \cos(Jt_2) + \sigma^+_k \cos(Jt_1) \sin(\bar{\omega}_k t_2) \cos(Jt_2) \\
-\sigma^+_k \sin(Jt_1) \sin(\bar{\omega}_k t_2) \sin(Jt_2) + \sigma^+_k \sin(Jt_1) \cos(\bar{\omega}_k t_2) \sin(Jt_2) \right],
\]

(3.22)

where we again only report terms that lead to an observable signal according to Eq. (3.11). The signal due to the above state reads

\[
s(t_1, t_2) = -\frac{i}{2} \sum_k \left[ e^{i\bar{\omega}_k t_2} e^{i(\bar{\omega}_k + J)t_1} + e^{-i\bar{\omega}_k t_2} e^{i(\bar{\omega}_k - J)t_1} \right].
\]

(3.23)

Hence we obtain peaks at \( \pm J, \bar{\omega}_k \pm J \).

The spectrum resulting from Eq. (3.23) is obtained using Eq. (3.5). It is schematically shown in part b) of Fig. 3.3. Part c) of the figure displays the projection of the spectrum to the \( \omega_2 \)-axis. While for this simple case the line structure is obvious, this will, in general, not be the case for systems including more spins. In such systems there can be overlapping multiplets where the physical origin of each line is not immediately clear. It should be clear that a 2D
3. Two-dimensional spectroscopy for the study of Coulomb crystals

\[ H = \sum_k \frac{\hbar \tilde{\omega}_k}{2} \sigma_k^x + \sum_{j<k} \frac{\hbar J_{jk}}{2} \sigma_j^z \sigma_k^z. \]  

(3.24)

The evolution operator for the time \( t_1 \) taking into account the \( \pi \)-pulse in \( y \) reads

\[ U(t_1) = e^{-\frac{i}{\hbar} \frac{1}{2} H_\pi} R_\pi e^{\frac{i}{\hbar} \frac{1}{2} H_\pi}, \]  

(3.25)

where \( R_\pi = \Pi_j (\sigma_j^y) \). Using that the two parts of the Hamiltonian commute with each other and inserting \( \Pi = (R_\pi)\dagger R_\pi \) twice we obtain

\[ U(t_1) = e^{-\frac{i}{\hbar} \frac{1}{2} H_\pi} R_\pi e^{-\frac{i}{\hbar} \frac{1}{2} \sum_i \frac{\hbar}{2} \sigma_i^z} (R_\pi)\dagger R_\pi e^{-\frac{i}{\hbar} \frac{1}{2} \sum_i \frac{\hbar}{2} \sigma_i^z} (R_\pi)\dagger. \]  

(3.26)

In order to see how we can proceed, let us define \( H_{12} = \frac{J_{12}}{2} \sigma_1^z \sigma_2^z \) and \( H_{ij} \) accordingly. Then let us consider the term \( R_\pi^* e^{-\frac{i}{\hbar} \sum_i \frac{\hbar}{2} \sigma_i^z} (R_\pi)\dagger \) for the case of three spins. We have

\[ R_\pi^* e^{-\frac{i}{\hbar} \sum_i \frac{\hbar}{2} \sigma_i^z} (R_\pi)\dagger = \sigma_1^x \sigma_2^x \sigma_3^x e^{-iH_{12}} \frac{1}{2} e^{-iH_{13}} \frac{1}{2} e^{-iH_{23}} \frac{1}{2} \sigma_1^y \sigma_2^y \sigma_3^y \]

\[ = \sigma_1^x \sigma_2^x \sigma_3^x e^{-iH_{12}} \frac{1}{2} \sigma_3^x e^{-iH_{13}} \frac{1}{2} \sigma_1^y e^{-iH_{23}} \frac{1}{2} \sigma_2^y \sigma_3^y. \]  

(3.27)
Now, using $[\sigma_j^x, \sigma_k^z] = 0$ for $j \neq k$ and $(\sigma_j^y)^2 = \mathbb{I}$, we can insert unity between the $e^{-iH_{\text{tot}}t}$, for example $(\sigma_1^y)^2(\sigma_2^y)^2$ after $e^{-iH_1t/2}$, to obtain

$$R_\pi^y e^{-i\Sigma_{\delta \ell} J_{\delta \ell} \hat{\sigma}_\delta^z \hat{\sigma}_\ell^z} (R_\pi^y)^\dagger = (\sigma_1^y \sigma_2^y e^{-iH_{\text{tot}}t/2} \sigma_1^y \sigma_2^y)(\sigma_1^y \sigma_3^y e^{-iH_{\text{tot}}t/2} \sigma_1^y \sigma_3^y)(\sigma_2^y \sigma_3^y e^{-iH_{\text{tot}}t/2} \sigma_2^y \sigma_3^y).$$

(3.28)

After this little detour it should be clear that we can write the propagator $U(t_1)$ as

$$U(t_1) = e^{-i\frac{H_{\text{tot}}}{2}t_1} R_\pi^y e^{-i\Sigma_{\delta \ell} J_{\delta \ell} \hat{\sigma}_\delta^z \hat{\sigma}_\ell^z} (R_\pi^y)^\dagger = e^{-i\frac{H_{\text{tot}}}{2}t_1} \left[ \sigma_j^z e^{-i\Sigma_{\delta \ell} J_{\delta \ell} \hat{\sigma}_\delta^z \hat{\sigma}_\ell^z} \right] \Pi_{j \neq k} \left[ \sigma_j^z \sigma_k^z e^{-iJ_{\delta \ell} \hat{\sigma}_\delta^z \hat{\sigma}_\ell^z} \right] R_\pi^y.$$

(3.29)

Expanding the exponentials as a power series and using $\sigma_j^z \sigma_k^z = -\sigma_k^z$ yields

$$U(t_1) = e^{-i\Sigma_{\delta \ell} J_{\delta \ell} \hat{\sigma}_\delta^z \hat{\sigma}_\ell^z} e^{-i\Sigma_{\delta \ell} J_{\delta \ell} \hat{\sigma}_\delta^z \hat{\sigma}_\ell^z} e^{-i\Sigma_{\delta \ell} J_{\delta \ell} \hat{\sigma}_\delta^z \hat{\sigma}_\ell^z} e^{-i\Sigma_{\delta \ell} J_{\delta \ell} \hat{\sigma}_\delta^z \hat{\sigma}_\ell^z} e^{-i\Sigma_{\delta \ell} J_{\delta \ell} \hat{\sigma}_\delta^z \hat{\sigma}_\ell^z} R_\pi^y.$$

(3.30)

If we now realize that $R_\pi^y \rho(t_p)(R_\pi^y)^\dagger$ leaves the initial state $\rho(t_p)$ from Eq. (3.14) invariant, we find that we can associate the effective Hamiltonian in Eq. (3.20) to the evolution period $t_1$.

### 3.1.3. Homogeneous and inhomogeneous broadening

In the previous two sections we have seen on the basis of two examples from NMR how 2D spectroscopy can witness couplings between nuclear spins and how interactions can be separated to the orthogonal axes of the spectrum. In both examples we have only considered the unitary evolution of the system neglecting dissipation and decoherence. In this section we will introduce the concept of **homogeneous** and **inhomogeneous** broadening which distinguish the action of different sorts of noise on the 2D spectra.

In Section 2.4 we discussed the interaction of a quantum system with an environment. In particular, we considered that the bath dynamics occurred on time scales much shorter than the time scale of the system evolution. The interaction of the system with such a fast environment leads to dephasing and relaxation. Typically, these effects lead to an exponential decay of the coherences on a timescale $T_2^*$ and we associate the decay rate $\Gamma = 1/T_2^*$ to it. Including dephasing the time evolution of a coherence and thus signal during a period $t_i$ becomes

$$s(t_i) \sim e^{-(\Gamma-i\omega_0)t_i}.$$  \hspace{1cm} (3.31)

The damping leads to a broadening of the peak in frequency space

$$S(\omega) \sim \frac{1}{\Gamma - i(\omega - \omega_0)} = \frac{\Gamma}{\Gamma^2 + (\omega - \omega_0)^2} + i\frac{\omega - \omega_0}{\Gamma^2 + (\omega - \omega_0)^2}. \hspace{1cm} (3.32)$$

It is important to note that the interaction with a “fast” environment leads to broadening of the line along one of the frequency axes. We can understand this in the following way. The interaction with the fast environment causes fluctuations of the system’s evolution frequency on a time scale faster than the typical time scale of the system’s evolution. Thus, after many repetitions of the experiment or an average over a sample with many spins, there is a distribution of frequencies usually centered at the unperturbed frequency contributing to the spectrum for the considered evolution period. Hence, fluctuations during a certain evolution period lead to a distribution of frequencies on the associated frequency axis. This distribution is, however, uncorrelated to the distribution on the other frequency axis due to the fast time scale of the fluctuations. This broadening of the lines due to dephasing and relaxation is termed **homogeneous broadening** [31].

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73
3. Two-dimensional spectroscopy for the study of Coulomb crystals

Figure 3.4.: Homogeneous and inhomogeneous broadening. Part a) shows a 2D spectrum of two uncoupled spins with different Larmor frequencies. Part b) shows the effect of a “fast” environment leading to dephasing and dissipation during each experimental run. The peaks are broadened along the frequency axes. This phenomenon is called homogeneous broadening. Part c) depicts the effect of inhomogeneous broadening. Here, frequencies are constant during each experimental run but fluctuate from run to run due to effective static disorder. In the considered case, this leads to broadening of the peaks along the main diagonal of the spectrum.

On the other hand, it can also happen that the evolution frequency of a spin, or more generally the system of interest, varies between experimental runs but can be considered constant during one experimental run. An equivalent situation would be that the local environments of the same sort of spin in a solid are static but vary slightly. In this case the evolution of each of the spins will differ but be constant during the evolution periods. In both cases we have a distribution of effective static frequencies. The signal of a period $t_i$ is then given by

$$s(t_i) \sim \sum_k e^{-i(\omega_k + \Delta\omega_k)t_i}$$

and the frequency space signal feature peaks at locations $(\omega_1, \omega_2) = (\omega_l + \Delta\omega_l, (\omega_k + \Delta\omega_k))$. The effects of this type of noise are most transparent for diagonal peaks. Here, the peaks will be spread along the diagonal. For off-diagonal peaks more complicated structures can arise. If the fluctuations are independent for the different spins, the effective static distribution of frequencies will lead to a spreading of the off-diagonal peaks on a “square lattice”. This type of broadening of the lines is called inhomogeneous broadening [31].

Thus, summarizing, the effects of the two considered types of noise are quite different. Fast fluctuations lead to broadening of the peaks along the frequency axes. Effective static disorder, on the other hand, leads to broadening of diagonal peaks along the diagonal. The effects of these two types of noise are illustrated for diagonal peaks in Fig. 3.4. Obviously, 2D spectroscopy is able to distinguish these two types of noise as can be appreciated in the figure.

3.2. The linear-to-zigzag structural transition

When talking about the motion of ions in a trap, so far we considered harmonic trap potentials and trapping conditions such that the ions form a linear string. Then a second order approximation of the Coulomb potential, Eq. (2.28), led us to the description of the motion in terms of uncoupled sets of normal modes. According to Eqs. (2.48) and (2.49) the normal mode frequen-
3.2. The linear-to-zigzag structural transition

Figure 3.5.: Part a) shows the radial $x$ normal mode frequencies of a crystal of three $^{40}$Ca$^{+}$ ions as a function of the anisotropy $\xi_x$. The frequency of the zigzag mode goes to zero approaching the linear-to-zigzag transition while the other frequencies remain finite. Part b) shows the effective potential for the zigzag mode for the radial potential $\omega_x$ larger then the critical value $\omega_{\text{crit}}$, see Eq. (3.35). There is only one stable minimum leading to the depicted linear configuration of the ions. Part c) shows the effective potential for the zigzag mode for radial potential $\omega_x$ below the critical anisotropy $\xi_x$, see Eq. (3.35). Now there are two minima which correspond to the two stable zigzag configurations depicted below.

Normal mode frequencies in the radial directions are given by

$$\omega_{n,\alpha} = \sqrt{\frac{1}{\xi_\alpha} + \frac{1}{2} \frac{\lambda_{n,z}}{\omega_\alpha^2}}, \quad \alpha = x, y,$$

(3.34)

where the $\lambda_{n,z}$ are the eigenvalues of the Hessian of the potential in the axial direction, Eq. (2.29), and $\xi_\alpha = \omega_\alpha^2/\omega_\alpha^2$ is defined in Eq. (2.30).

Let us now consider that the potential is kept stiff in one of the radial directions, say $y$, such that $\omega_y \gg \omega_x, \omega_z$, but the potential in $x$, which initially also satisfied $\omega_x \gg \omega_z$, is relaxed. As a consequence the normal mode frequencies in the $x$-direction decrease. Note that decreasing the anisotropy between the potentials in $x$ and $z$ leads to larger values of $\xi_x$ for our definition of the $\xi_\alpha$. In Fig. 3.5 we show the normal mode frequencies $\omega_{n,x}$ of a crystal of three $^{40}$Ca$^{+}$ as a function of $\xi_x$. We observe the expected behavior in the figure.

The zigzag mode frequency is the smallest mode frequency and, as we would expect from Eq. (3.34), it eventually tends to zero when we decrease the anisotropy between the potentials in the $x$- and $z$-directions. Decreasing this anisotropy even further would lead to a purely imaginary frequency of the zigzag mode which indicates that the structure of the crystal becomes unstable because there is no restoring force for this mode anymore. This is actually what is observed in the experiment [109–111].

Decreasing the anisotropy between $\omega_x$ and $\omega_z$ below a critical value, i.e. $\omega_x < \omega_{\text{crit}}$, the ions leave the linear string configuration and form a planar zigzag structure. This behavior is sketched in parts b) and c) of Fig. 3.5. The figure also shows that there are actually two degenerate configurations of the zigzag structure. Note that decreasing the anisotropy between the potentials below a critical value corresponds to $\xi_x$ being larger than a critical value $\xi_{\text{crit}}$. 


We further remark that a crystal can also be driven to the zigzag phase by increasing the axial frequency $\omega_z$.

Clearly, the description of the motion in terms of uncoupled normal modes in all three spatial directions does not apply in this planar configuration. For the configuration we have considered, i.e. for a much stiffer confinement of the ions in one direction, the motion in the planar phase can be described by two sets of normal modes. One in the plane where the ions form the zigzag structure and one perpendicular to that plane [32].

The linear-to-zigzag transition has attracted a lot of attention and was investigated from different perspectives. In [32] it was shown that the linear-to-zigzag transition is a second order phase transition in the thermodynamic limit where the zigzag mode is the soft mode and the distance of the ions from the $z$-axis is the order parameter. It was also conjectured that the transition is a quantum phase transition [112, 115].

The critical value of the anisotropy $\xi_{\text{crit}}$ depends on the number of ions in the trap and different power law scalings connecting the number of ions and the critical anisotropy were proposed, see [111] and references therein. In the cited publication the values of $\xi_{\text{crit}}$ were determined experimentally and showed very good agreement with the values one would expect from Eq. (3.34).

Let us try and understand the transition in a bit more detail. In the regime where each direction of motion can be described by a set of normal modes the overall potential of the ions, consisting of trap and Coulomb potentials, is treated in harmonic approximation. In this regime also the potential for each of the normal mode coordinates is harmonic, see Eq. (2.42). Furthermore, in the string configuration the potential energy due to the Coulomb interaction is much smaller than the trap potential in the radial directions.

If we now decrease the anisotropy between one of the radial directions and the axial direction, the Coulomb interaction becomes stronger in comparison with the trapping potential in the radial direction. Eventually, the harmonic approximation of the Coulomb potential is not valid anymore and higher order terms becomes important. Close to the critical anisotropy where the transition takes place the potential of the zigzag mode can be approximated in the form [32]

$$V(q_{zz}) = v^{(2)} q_{zz}^2 + v^{(4)} q_{zz}^4,$$

(3.35)

where $q_{zz}$ is the generalized coordinate of the zigzag mode. At the transition point the coefficient $v^{(2)}$ changes sign from positive to negative [32]. Accordingly, the potential for the soft mode instead of a single minimum exhibits two degenerate minima which correspond to the two degenerate configurations of the zigzag structure. The situation is depicted in parts b) and c) of Fig. 3.5.

Since there are two degenerate configurations of the zigzag phase, driving the system over the critical point can lead to the formation of defects which in this case are boundaries between the two possible structures. These were indeed observed [28, 29, 105]. The rate of production of defects in this transition was modeled using the Kibble-Zurek mechanism [27] and the predictions were confirmed experimentally [28, 29].

Let us finally note that when we consider the linear-to-zigzag transition in this chapter, we always assume that we approach the transition but the crystal remains in the linear structure.

### 3.3. Nonlinear interactions in crystals of trapped ions

Up to this point we have described the motion of ions in a trap by the Hamiltonian

$$H_0 = \sum_{n, \alpha} \hbar \omega_{n, \alpha} a_{n, \alpha}^\dagger a_{n, \alpha}. $$

(3.36)
This Hamiltonian was obtained from the harmonic approximation $V^{(2)}$, Eq. (2.27), of the full potential $V$, Eq. (2.18), experienced by the ions in the trap. It is a good description of the motion of the ions in the string configuration unless nonlinearities are added on purpose, for example by means of additional optical fields [33]. Now, in the previous section we have seen that close to the linear-to-zigzag transition the harmonic approximation of the Coulomb potential does not suffice to describe the dynamics. Here, we report the first two nonlinear corrections to the harmonic approximation of the Hamiltonian for the case that no nonlinearities are added to the trap potential. In this case these corrections are the third- and fourth-order expansions of the Coulomb potential in the small displacements of the ions from their equilibrium positions.

3.3.1. Third-order expansion of the Coulomb potential

In this section we start by reporting the third-order expansion of the Coulomb potential in the small displacements of the ions from their equilibrium positions, which is the first correction to the harmonic approximation. For the evaluation of these contributions we will assume that the ions are in the linear string configuration. Then, their equilibrium positions are $x_i^0 = y_i^0 = 0$ while the $c_{ij}^0$ are obtained from Eq. (2.23) together with Eq. (2.22). The third order expansion of the Coulomb interaction of a crystal of $N$ ions of equal mass $m$ is given by

$$V^{(3)} = \frac{1}{3!} \sum_{i,j,k=1}^{N} \sum_{\mu,\nu,\gamma} \frac{\partial^3 V_C}{\partial r_{i\mu} \partial r_{j\nu} \partial r_{k\gamma}} q_{i\mu} q_{j\nu} q_{k\gamma}. \tag{3.37}$$

where the $q_{i\mu}, q_{j\nu}, q_{k\gamma}$ are the small amplitude displacement from equilibrium in Eq. (2.58).

In order to ease notation, we introduce the notation $q_{i\mu} = \delta_{i \mu}, \mu \in \{x, y, z\}$. In this way the different spatial coordinates are easier to identify. Evaluating the above expression and using the dimensionless equilibrium positions from Eq. (2.23), we obtain after some algebra [116]

$$V^{(3)} = \frac{m \omega^2}{2 \ell_z} \sum_{i,j,k} C_{ijk}^{(3)} (3 \delta_{ij} \delta x_j + 3 \delta_{ij} \delta y_j - 2 \delta_{ij} \delta z_j) \delta z_k \tag{3.38}$$

with the tensor $C_{ijk}^{(3)}$

$$C_{ijk}^{(3)} = \begin{cases} \sum_{\mu \neq k} \frac{\text{sgn}(q_{ij}^0 - q_{ik}^0)}{|q_{ij}^0 - q_{ik}^0|^2} & \text{if } i = j = k \\ \sum_{\mu \neq j} \frac{\text{sgn}(q_{ij}^0 - q_{ik}^0)}{|q_{ij}^0 - q_{ik}^0|^2} & \text{if } i = j \neq k \\ \frac{\text{sgn}(q_{ij}^0 - q_{ik}^0)}{|q_{ij}^0 - q_{ik}^0|^2} & \text{if } i \neq j = k \\ \frac{\text{sgn}(q_{ij}^0 - q_{ik}^0)}{|q_{ij}^0 - q_{ik}^0|^2} & \text{if } i \neq j \neq k \end{cases} \tag{3.39}$$

Note that $C_{ijk}^{(3)}$ is symmetric under exchange of two indices and for $i \neq j$ we have $C_{ij}^{(3)} = -C_{jii}^{(3)}$. With the help of these two properties we can show that

$$\sum_k C_{ijk}^{(3)} = 0. \tag{3.40}$$

The above identity follows directly from the properties of $C_{ijk}^{(3)}$ [116]: For $i \neq j$ we have $\sum_k C_{ijk}^{(3)} = C_{ij}^{(3)} + C_{ij}^{(3)} = C_{ij}^{(3)} - C_{ij}^{(3)} = 0$, while for $i = j$ we have $\sum_k C_{iik}^{(3)} = C_{iij}^{(3)} + \sum_{k \neq i} C_{iik}^{(3)}$ which is seen to be zero using the definition of $C_{iij}^{(3)}$. We will use the property in Eq. (3.40) in a moment.
3. Two-dimensional spectroscopy for the study of Coulomb crystals

For a perturbation expansion the third order effects need to be small compared with the harmonic terms. In this case the normal mode picture is still valid and we can expand the coordinates in terms of the quantized normal modes as in Eq. (2.58). For clarity of notation we use $a_{n,x} = a_n$, $a_{n,y} = b_n$ and $a_{n,z} = c_n$. The Hamiltonian associated with the third-order expansion of the Coulomb potential then takes the form [116]

\[ H^{(3)}_{\text{full}} = \frac{z_0}{4\hbar c} \hbar \omega_x \sum_{n,m,p} D^{(3)}_{nmp} \left[ \frac{3(a_n + a_n^\dagger)(a_m + a_m^\dagger)}{\sqrt{\lambda_{n,x}\lambda_{m,x}}} + \frac{3(b_n + b_n^\dagger)(b_m + b_m^\dagger)}{\sqrt{\lambda_{n,y}\lambda_{m,y}}} ight. \]

\[ \left. - \frac{2(c_n + c_n^\dagger)(c_m + c_m^\dagger)}{\sqrt{\lambda_{n,z}\lambda_{m,z}}} \right] \quad \text{(3.41)} \]

where we have introduced the tensor $D^{(3)}_{nmp}$

\[ D^{(3)}_{nmp} = \sum_{i,j,k} C^{(3)}_{ijk} B_{in} B_{jm} B_{kp}. \quad \text{(3.42)} \]

Here, $B$ is the matrix diagonalizing the second order expansion of the full potential given in Eq. (2.44) and $z_0 = \sqrt{\hbar/(2m\omega_x)}$ is the spread of the longitudinal center-of-mass ground state wave packet of a single ion with frequency $\omega_x$. Due to the symmetry of $C^{(3)}_{ijk}$ under exchange of indices also $D^{(3)}_{nmp}$ is symmetric under exchange of indices. Moreover, all elements of $D^{(3)}_{nmp}$ involving the center-of-mass modes, i.e. all $D^{(3)}_{nmp}$ with at least one of the indices equal to one, vanish. This can be seen using the property of $C^{(3)}_{ijk}$ in Eq. (3.40). From Eq. (2.46) we know that $B_{1i} = 1/\sqrt{N}$ for all $i$ such that we obtain

\[ D^{(3)}_{n1j} = \sum_{i,j,k} C^{(3)}_{ijk} B_{in} B_{jm} B_{k1} = \frac{1}{\sqrt{N}} \sum_{i,j} B_{in} B_{jm} \sum_k C^{(3)}_{ijk} = 0. \quad \text{(3.43)} \]

It is interesting to note that we should have expected this property of $D^{(3)}_{nmp}$ on physical grounds. For $D^{(3)}_{n1j} \neq 0$ quanta of excitation could be transferred from the other modes to the center-of-mass mode changing the momentum of the crystal as a whole. However, the Coulomb repulsion of the crystal of the ions is an internal force of the crystal and as such should conserve the momentum of the crystal. Accordingly, $D^{(3)}_{n1j} = 0$ is an expression of momentum conservation.

### 3.3.2. Resonant interactions in the third-order Hamiltonian

In order to analyze the effects of the third-order correction Hamiltonian $H^{(3)}_{\text{full}}$ one moves to an interaction picture with respect to the harmonic Hamiltonian in Eq. (3.36). Note that the energy scale of the third order processes is smaller than the harmonic contributions by a factor $z_0/l_z$. For a trapping frequency in the MHz range and typical ion masses $z_0/l_z \approx 10^{-3}$ and thus third-order effects will only be observable if they are resonant. An analysis of possible resonances has been carried out in [116]. Reporting only terms that can yield a resonance the third-order Hamiltonian can be approximated as

\[ H^{(3)}_{\text{full}} \approx -\frac{3z_0}{4\hbar c} \hbar \omega_x \sum_{n,m,p} D^{(3)}_{nmp} \left[ 2 \frac{a_n^\dagger a_m c_p}{\sqrt{\lambda_{n,x}\lambda_{m,x}\lambda_{p,z}}} e^{i\Delta_{nmp}\omega_t} + 2 \frac{b_n^\dagger b_m c_p}{\sqrt{\lambda_{n,y}\lambda_{m,y}\lambda_{p,z}}} e^{i\Delta_{nmp}\omega_t} ight. \]

\[ \left. + \frac{a_n^\dagger a_m^\dagger c_p}{\sqrt{\lambda_{n,x}\lambda_{m,x}\lambda_{p,z}}} e^{i\Delta_{nmp}\omega_t} + \frac{b_n^\dagger b_m^\dagger c_p}{\sqrt{\lambda_{n,y}\lambda_{m,y}\lambda_{p,z}}} e^{i\Delta_{nmp}\omega_t} \right] + \text{H.c.}, \quad \text{(3.44)} \]
where \( \Delta_{\text{rup}, \alpha} = \sqrt{\lambda_{n, \alpha} + \lambda_{m, \alpha} - \lambda_{p, z}} \). As is apparent from the above Hamiltonian, there are only two types of resonances that can occur: In the first type of resonance, due to the first two terms in the brackets, one radial phonon is created while one axial and one radial phonon are annihilated. Of course, the reverse process is also possible due to the Hermitian conjugate. In the second type of resonance two radial phonons are created while one axial phonon is annihilated, where again the reverse process is included by the Hermitian conjugate.

For one of the processes to be resonant the associated \( \Delta_{\text{rup}, \alpha} \) needs to vanish. From Eqs. (2.43) and (2.49) we know that the axial and radial frequencies are not independent but are connected by the anisotropy \( \xi_a \). A list of possible resonances reporting the involved modes and associated values of \( \xi_a \) for strings of up to \( N = 10 \) ions is given in [116]. Resonances of the first type always have to involve two different radial modes since all \( \lambda_{n, z} > 0 \). It is interesting to note that these resonances do not occur in crystals involving six or less ions. Furthermore, there is a minimal value for \( \xi_{a}^{\text{res}} \) such that no resonances occur for \( \xi_a < \xi_{a}^{\text{res}} \). The difference between the trapping potentials and thus in mode frequencies between the axial and radial directions is then so large that it forbids any resonances.

Let us now consider an example of a resonant interaction in the third-order Hamiltonian \( H^{(3)}_{\text{full}} \), Eq. (3.41). We will investigate the resulting effective Hamiltonian using 2D spectroscopy below. We consider a crystal with \( N = 3 \) ions and assume that the degeneracy between the radial modes is lifted keeping the \( y \) modes sufficiently stiff, \( \xi_y < \xi_{y}^{\text{res}} \). Accordingly, there are no resonances involving the \( y \)-modes and we focus on the radial \( x \)-direction.

For a three ion crystal there can only be what we called a resonance of the second type due to the last two terms in the brackets in Eq. (3.44). In this type of resonance one axial phonon is created while two radial phonons are annihilated and due to the Hermitian conjugate also the reverse process is included by the Hermitian conjugate. For \( \xi_x = 20/63 \) there is a resonant coupling between the \( x \)-zigzag mode and the axial stretch mode. The resonant contribution of the third-order Hamiltonian in Eq. (3.41) is then given by

\[
H^{(3)}_{\text{res}} = \hbar \Omega_T \left[ a_3^\dagger c_2 + (a_3^\dagger)^2 c_2 \right],
\]  

where we have introduced the resonant coupling strength

\[
\Omega_T = 3 \zeta_0 \omega_x D^{(3)}_{332} / (4 \sqrt{\lambda_{3, x} \lambda_{2, z}}).
\]  

To make the association clearer, we write \( a_3 \equiv a_{zz}, c_2 \equiv c_{\text{str}} \) and \( \omega_{3, x} \equiv \omega_x, \omega_{2, z} \equiv \omega_{\text{str}} \) in the sequel. With this notation the resonant part of the Hamiltonian can be written as

\[
H^{(3)}_{\text{res}} = \hbar \Omega_T \left[ a_{zz}^\dagger c_{\text{str}} + (a_{zz}^\dagger)^2 c_{\text{str}} \right].
\]  

Note that this resonance implies \( \omega_{\text{str}} = 2 \omega_x \). For this Hamiltonian to be the effective Hamiltonian all other terms in the third-order Hamiltonian, Eq. (3.41), must be negligible as well as the effects due to the fourth-order Hamiltonian in Eq. (3.54) below. For the considered parameters this should be the case and we will, therefore, work with the effective Hamiltonian \( H^{(3)}_{\text{res}} \) for the remainder of this section.

It should be noted, though, that a third-order Hamiltonian can only be an approximation for a small excitation numbers, i.e. for sufficiently small oscillation amplitudes around equilibrium, since its energy spectrum is not bounded from below. For larger numbers of excitation the fourth-order terms have to be taken into account. These render the energy spectrum lower-bounded and thus restore the stability of the system.
Table 3.1.: Eigenvalues and corresponding eigenstates of $H_{\text{res}}^{(3)}$, Eq. (3.47), in the small occupation number manifolds of the zigzag and stretch modes.

| Manifold $|n_{zz},n_{\text{str}}\rangle$ | States | Eigenvalues |
|---------------------------------------|--------|-------------|
| $|0,1\rangle,|2,0\rangle$ | $\frac{1}{\sqrt{2}}(|0,1\rangle \pm |2,0\rangle)$ | $\pm \sqrt{2}\Omega_T$ |
| $|1,1\rangle,|3,0\rangle$ | $\frac{1}{\sqrt{2}}(|1,1\rangle \pm |3,0\rangle)$ | $\pm \sqrt{6}\Omega_T$ |
| $|0,2\rangle,|2,1\rangle,|4,0\rangle$ | $\frac{1}{\sqrt{2}}\left(\pm \frac{1}{2}|0,2\rangle \pm |2,1\rangle \pm \frac{\sqrt{3}}{2}|4,0\rangle\right)$ | $\pm 4\Omega_T$ |
| $|0,2\rangle,|2,1\rangle,|4,0\rangle$ | $\frac{\sqrt{3}}{2}|0,2\rangle - \frac{1}{2}|4,0\rangle$ | 0 |
| $|1,2\rangle,|3,1\rangle,|5,0\rangle$ | $\frac{1}{\sqrt{2}}\left(\pm \frac{\sqrt{3}}{2}|1,2\rangle \pm |3,1\rangle \pm \frac{\sqrt{5}}{2}|5,0\rangle\right)$ | $\pm \sqrt{2}\Omega_T$ |
| $|1,2\rangle,|3,1\rangle,|5,0\rangle$ | $\frac{\sqrt{5}}{2}|1,2\rangle - \frac{\sqrt{3}}{2}|5,0\rangle$ | 0 |

Let us now proceed assuming the system is correctly described by $H_{\text{res}}^{(3)}$ from Eq. (3.47). The eigenvalues and eigenvectors of this Hamiltonian in the manifolds involving only a few excitations in the two modes can be found analytically. To this end note that

$$ [H_{\text{res}}^{(3)}, \hat{n}_{zz} + 2\hat{n}_{\text{str}}] = 0, \quad (3.48) $$

where $\hat{n}_{zz}$ and $\hat{n}_{\text{str}}$ are the number operators of the zigzag and stretch mode, respectively. Recalling that $\omega_{\text{str}} = 2\omega_{zz}$, the above equation tells us that $H_{\text{res}}^{(3)}$ only induces transitions between states which are degenerate with respect to the harmonic Hamiltonian $H_0$ in Eq. (3.36). The eigenstates and eigenvalues of $H_{\text{res}}^{(3)}$ are then found inserting linear combinations of degenerate eigenstates of $H_0$ into $H_{\text{res}}^{(3)}|\psi_n\rangle = E_n|\psi_n\rangle$. The eigenvalues and eigenvectors of $H_{\text{res}}^{(3)}$ in the manifolds with only a few excitations in the two modes are reported in Tab. 3.1. Note that the states $|n_{zz} = 0, n_{\text{str}} = 0\rangle$ and $|n_{zz} = 1, n_{\text{str}} = 0\rangle$ are eigenstates with eigenvalue zero.

### 3.3.3. Fourth-order terms in the Coulomb potential

In this section we will present the fourth-order corrections to the harmonic approximation of the Coulomb potential. The fourth-order expansion of the Coulomb potential is given by the expression

$$ V^{(4)} = \frac{1}{4!} \sum_{\mu,\beta,\gamma,\delta} \frac{\partial^4 V_C}{\partial r_\mu \partial r_\beta \partial r_\gamma \partial r_\delta} \mathcal{V}_{\mu,\beta,\gamma,\delta}^{(4)} q_\mu q_\beta q_\gamma q_\delta \cdot \quad (3.49) $$

Using the equilibrium positions $x_i^0 = y_i^0 = 0$ and writing the $z_i^0 = u_i^0 l_z$ as in Eq. (2.22), where the $u_i^0$ are determined by Eqs. (2.23), after considerable algebra we obtain

$$ V^{(4)} = \frac{3\epsilon^2}{4! 4\pi \epsilon_0 l_z^3} \sum_{i,j,k,l} C_{ijkl}^{(4)} \left(3\delta x_i \delta x_j \delta x_k \delta x_l + 3\delta y_i \delta y_j \delta y_k \delta y_l + 8\delta z_i \delta z_j \delta z_k \delta z_l + 6\delta x_i \delta z_j \delta y_k \delta y_l + 24\delta x_i \delta x_j \delta z_k \delta z_l - 24\delta y_i \delta y_j \delta z_k \delta z_l \right), \quad (3.50) $$
where we used $q_{i\mu} = \delta \mu_i$ for $\mu \in \{x,y,z\}$ as above and introduced the tensor

\[
C_{ijkl}^{(4)} = \delta_{ij} \delta_{jk} \delta_{kl} \sum_{p \neq l} \frac{1}{|u_p^0 - u_p|^5} + \delta_{ij} \delta_{jk} (1 - \delta_{ik}) \frac{-1}{|u_p^0 - u_l|^5} + \delta_{ij} (1 - \delta_{jk}) \delta_{kl} \frac{-1}{|u_p^0 - u_l|^5} + (1 - \delta_{ij}) \delta_{jk} \delta_{kl} \frac{-1}{|u_p^0 - u_l|^5} + (1 - \delta_{ij}) \delta_{jk} \delta_{kl} \frac{-1}{|u_p^0 - u_l|^5} + (1 - \delta_{ij}) \delta_{jk} \delta_{kl} \frac{1}{|u_p^0 - u_l|^5}.
\]

From the above equation we see that also $C_{ijkl}^{(4)}$ is symmetric with respect to exchange of two indices. With this information we may write $C_{ijkl}^{(4)}$ more compactly as

\[
C_{ijkl}^{(4)} = \begin{cases} 
\frac{1}{|u_p^0 - u_p|^5} & \text{if } i = j = k = l \\
\frac{-1}{|u_p^0 - u_l|^5} & \text{if } i = j = k \neq l \\
\frac{1}{|u_p^0 - u_l|^5} & \text{if } i = j \neq k = l \\
0 & \text{else.}
\end{cases} \tag{3.52}
\]

Analogous to the property of $C_{ijkl}^{(3)}$ in Eq. (3.40), $C_{ijkl}^{(4)}$ satisfies

\[
\sum_i C_{ijkl}^{(4)} = 0. \tag{3.53}
\]

This can be shown in a similar manner as above: For $i \neq j \neq k$ all elements $C_{ijkl}^{(4)}$ appearing in the sum are zero as follows from the definition of $C_{ijkl}^{(4)}$ in Eq. (3.52); if $i = j \neq k$, the sum becomes $\sum_i C_{ijkl}^{(4)} = C_{ikij}^{(4)} + C_{ikjk}^{(4)} = 0$ because $C_{ikij}^{(4)} = -C_{ikjk}^{(4)}$, finally, if $i = j = k$, we have $\sum_i C_{ijkl}^{(4)} = C_{ijkl}^{(4)} + \sum_{i \neq j} C_{ijkl}^{(4)}$, which is again found to be zero by inserting the expressions for $C_{ijkl}^{(4)}$ and $C_{ijkl}^{(4)}$ from the definition Eq. (3.52).

Under the assumption that the fourth-order contributions are weak in comparison with the harmonic contributions, we expand the ions’ coordinates in terms of the normal modes as in Eq. (2.58). Then we can write the fourth-order correction to the harmonic Hamiltonian in Eq. (3.36) as

\[
H_{\text{full}}^{(4)} = \left( \frac{\hbar \omega_z}{4 \lambda_c^2} \right)^2 \sum_{n,m,p,q} D_{mpq}^{(4)} \begin{aligned} &\left[ \frac{3}{(\lambda_c^2 \lambda_m \lambda_n \lambda_p \lambda_q)^2} \right] (a_n + a_n^\dagger)(a_m + a_m^\dagger)(a_p + a_p^\dagger)(a_q + a_q^\dagger) \\
+ &\frac{3}{\lambda_c^2 \lambda_m \lambda_n \lambda_p \lambda_q} (b_n + b_n^\dagger)(b_m + b_m^\dagger)(b_p + b_p^\dagger)(b_q + b_q^\dagger) \\
+ &\frac{8}{\lambda_c^2 \lambda_m \lambda_n \lambda_p \lambda_q} (c_n + c_n^\dagger)(c_m + c_m^\dagger)(c_p + c_p^\dagger)(c_q + c_q^\dagger) \\
+ &\frac{6}{(\lambda_c^2 \lambda_m \lambda_n \lambda_p \lambda_q)^2} (a_n + a_n^\dagger)(a_m + a_m^\dagger)(b_p + b_p^\dagger)(b_q + b_q^\dagger) \\
- &\frac{24}{\sqrt{\lambda_c^2 \lambda_m \lambda_n \lambda_p \lambda_q}} (a_n + a_n^\dagger)(a_m + a_m^\dagger)(c_p + c_p^\dagger)(c_q + c_q^\dagger) \\
- &\frac{24}{\sqrt{\lambda_c^2 \lambda_m \lambda_n \lambda_p \lambda_q}} (b_n + b_n^\dagger)(b_m + b_m^\dagger)(c_p + c_p^\dagger)(c_q + c_q^\dagger), \end{aligned} \tag{3.54}
\]
where we have introduced

\[ D_{nmpq}^{(4)} = \sum_{i,j,k,l} C_{ijkl}^{(4)} M_n M_{jm} M_{kp} M_l. \]  

(3.55)

Note that \( D_{nmpq}^{(4)} \) is symmetric under exchange of two indices because \( C_{ijkl}^{(4)} \) is. From the analysis of the third order expansion we expect that there are no couplings to the center-of-mass modes, i.e. \( D_{nmp1}^{(4)} = 0 \). This is actually the case and easily verified using \( M_{l1} = 1/\sqrt{N}, l = 1, \ldots, N \) and Eq. (3.53):

\[ D_{nmp1}^{(4)} = \sum_{i,j,k,l} C_{ijkl}^{(4)} M_n M_{jm} M_{kp} \frac{1}{\sqrt{N}} = \frac{1}{\sqrt{N}} \sum_{i,j,k} M_n M_{jm} M_{kp} \sum_{l} C_{ijkl}^{(4)} = 0. \]  

(3.56)

Thus, \( D_{nmpq}^{(4)} = 0 \) if at least one of the indices is one. Again, this is a manifestation of momentum conservation.

### 3.3.4. Resonant interactions in the fourth-order Hamiltonian

We now proceed to identify the regimes in which terms in \( H_{\text{full}}^{(4)} \), Eq. (3.54), have an appreciable effect on the motional dynamics. The fourth-order contributions come with a prefactor \( [z_0/(4l_z)]^2 \). As we pointed out previously under typical operating conditions \( z_0/l_z \approx 10^{-3} \) and the fourth-order corrections are roughly a factor \( 10^{-6} \) smaller than the elements of the harmonic Hamiltonian in Eq. (3.36).

It is interesting to note, though, that even these weak fourth-order corrections can have an appreciable effect under normal trapping conditions. Terms of the form \( a_n^\dagger a_m^\dagger c_m c_n \) and alike that appear in \( H_{\text{full}}^{(4)} \), Eq. (3.54), will always be resonant. Let us consider for simplicity that we have a system of only two modes. If we want to probe one mode and the other one is in a thermal state, these Kerr-type interactions will act as effective dephasing for the first mode. Indeed, in Ref. [113] it was found that these Kerr-type interactions were the leading contribution to dephasing of a motional mode under normal trapping conditions. The reference studied the coherence of the axial stretch mode of a two-ion crystal and Kerr-type interactions of the above form, coupling the stretch mode to the radial rocking modes, were found to be the limiting factor of the stretch mode’s coherence time. This is remarkable because the couplings only led to shifts of the stretch mode frequency of \( \sim 10\text{Hz per quantum} \) in the rocking modes. If not taken care of, such shifts can even limit the achievable fidelities in quantum information processing experiments [113].

On the other hand, the nonlinear effects can also be quite strong. As we discussed above this is, for example, the case when approaching the linear-to-zigzag transition. In the vicinity of the transition the effective potential, at least for the zigzag mode, becomes quartic, see Eq. (3.35). We can actually see this behavior from Eq. (3.54).

Let us consider that the trapping potential in \( x \)-direction is relaxed while the potential in \( y \) is kept stiff. As we have seen in Fig. 3.5 for the case of three ions, approaching the critical anisotropy the normal mode frequencies in \( x \) decrease. Consequently, the terms in \( H_{\text{full}}^{(4)} \), involving the \( \lambda_{n,x} \) will increase as one moves towards the transition point and can assume appreciable values. This decrease is most pronounced for the zigzag mode frequency, which tends to zero approaching the transition point. Hence, also the eigenvalue \( \lambda_{N,x} \) will tend to zero approaching the transition point and especially the terms involving \( \lambda_{N,x} \) will increase. It follows from these considerations that approaching the linear-to-zigzag transition the dominant contributions
3.3. Nonlinear interactions in crystals of trapped ions

in $H_{\text{full}}^{(4)}$ of Eq. (3.54) are those including the modes of the $x$-direction

$$H_{\text{full}}^{(4)} \approx H^{(4)} = 3 \left( \frac{z_0}{4 \xi_c} \right)^2 \hbar \omega \sum_{n,m,p,q} D_{nmpq}^{(4)} \frac{1}{\sqrt{\lambda_{n,x} \lambda_{m,x}}} \left[ (a_n^+ + a_n^*) (a_m^+ + a_m^*) \frac{1}{\sqrt{\lambda_{p,x} \lambda_{q,x}}} \frac{1}{\sqrt{\lambda_{p,y} \lambda_{q,y}}} \frac{1}{\sqrt{\lambda_{p,z} \lambda_{q,z}}} \right].$$

(3.57)

However, also the dominant part of the full fourth-order Hamiltonian features many contributions and therefore demands further consideration to sort out the most important contributions for the dynamics. In order to structure the analysis, we divide $H^{(4)}$ into three parts

$$H^{(4)} = H_{xx} + H_{xy} + H_{xz},$$

(3.58)

where the first term solely involves the interactions between the $x$-modes and the terms $H_{xy}$ and $H_{xz}$ the interactions of the $x$-modes with the $y$- and $z$-modes, respectively. The simplest case where a transition to the zigzag phase can occur is for $N = 3$ ions and therefore we will restrict our attention to crystals consisting of three ions. Treating more ions follows the same steps but increases the complexity of the treatment.

We start by considering $H_{xx}$ which, collecting all terms involving only interactions between the $x$-modes from Eq. (3.57), reads

$$H_{xx} = 3 \left( \frac{z_0}{4 \xi_c} \right)^2 \hbar \omega \sum_{n,m,p,q} D_{nmpq}^{(4)} \frac{1}{\sqrt{\lambda_{n,x} \lambda_{m,x} \lambda_{p,x} \lambda_{q,x}}} (a_n^+ + a_n^*) (a_m^+ + a_m^*) (a_p^+ + a_p^*) (a_q^+ + a_q^*).$$

(3.59)

The resonant terms in $H_{xx}$ are again found by moving to an interaction picture with respect to $H_0$, Eq. (3.36). Obviously, there are many resonant cross-Kerr type couplings between two modes. These arise when two indices are equal, e.g. for $n = m$ and $p = q$. We then find resonances of the form $a_n^* a_n a_p^* a_p$ and similarly for the cases $n = p$, $m = q$ as well as $n = q$, $m = p$. These terms will not acquire a time dependence in the rotating frame. As we discussed earlier, these terms lead to effective dephasing of the motional modes for thermal occupations of the modes. Terms which are a permutation of $a_n^* a_n a_p^* a_p$ will also yield contributions $\sim a_n^* a_n$ and $\sim a_p^* a_p$ when rearranging them into the form $a_n^* a_n a_p^* a_p$. Accordingly, these resonant terms also cause shifts of the frequencies of the involved modes.

In case all indices are equal, we obtain resonant terms of the form $(a_n^*)^2 a_n^2$ and $a_n^* a_n$. The contributions proportional to the number operator arise from commutators when rearranging other terms to the ordering $(a_n^*)^2 a_n^2$. Hence, the effects of the terms with all indices equal are twofold. We obtain a self-interaction of the modes which looks like the on-site interaction of a Bose-Hubbard model and shifts of the mode frequencies.

To summarize, the resonant contributions in $H_{xx}$ lead to cross-Kerr couplings between the $x$-modes, a self-interaction of the modes and mode frequency shifts. These are the obvious resonant contributions of $H_{xx}$. However, there is a plethora of non-resonant contributions. In order to make sure that we do not exclude any important contribution, it needs to be checked that these can be neglected.

In order to assess whether or not the non-resonant contributions can be neglected, we need to consider a specific set of parameters. Later, we will be interested in measuring the strength of the self-interaction of the zigzag mode close to the linear-to-zigzag transition because the self-interaction is a contribution of the fourth-order term in Eq. (3.35) that appears in the neighborhood of the structural transition. The emergence of this term and thus the self-interaction is a signature of the onset of the structural transition. A set of parameters where the crystal is still in the linear phase but the self-interaction has an appreciable value is reported in Tab. 3.2 for
a three-ion crystal of $^{40}\text{Ca}^+$. Note that the frequency $\omega_z$ is rounded and the exact value of the anisotropy we used was $\xi_z = 0.4159123$.

The procedure to analyze which contributions can be neglected is the following. For each combination $nmpq$ on the right hand side of Eq. (3.59) there are 16 operator terms $O_{nmpq,i}$, $i = 1, \ldots, 16$ consisting of different combinations of creation and annihilation operators. We denote their coefficients and time dependencies by $c_{nmpq,i}$ and $\omega_{nmpq,i}$, respectively. Then, we check if for all energy non-conserving terms, i.e. the terms with $\omega_{nmpq,i} \neq 0$, we have $c_{nmpq,i}/\omega_{nmpq,i} \ll 1$. If this is the case, they can be neglected in a rotating wave approximation. For the above parameters such an analysis shows that all energy non-conserving terms can be neglected. Denoting the zigzag mode operator in $x$ $a_3 \equiv a_{zz}$ the resonant part of $H_{xx}$ may be written as

$$H_{xx} = \frac{\hbar \Omega_{SI}^{zz}}{2} (a_{zz}^\dagger)^2 a_{zz}^2 + \hbar \Delta \omega_{zz} a_{zz}^\dagger a_{zz} + \hbar \Omega_{4,2}^{zz} a_{zz}^\dagger a_{zz}^\dagger a_2 + \frac{\hbar \Omega_{SI}^{zz}}{2} (a_2^\dagger)^2 a_2^2 + \hbar \Delta \omega_{zz} a_{zz}^\dagger a_2^\dagger a_2, \quad (3.60)$$

where we have introduced the self-interactions, frequency shifts and mode couplings

$$\Omega_{SI}^{zz} = 12 \times 3 \left( \frac{\zeta_0}{4\ell_z} \right)^2 \frac{D_{3333}^{(4)}}{\lambda_{zz}} \omega_z, \quad \Omega_{SI}^{x^2} = 12 \times 3 \left( \frac{\zeta_0}{4\ell_z} \right)^2 \frac{D_{2222}^{(4)}}{\lambda_{zz}^2} \omega_z, \quad (3.61)$$

$$\Omega_{4,2}^{zz} = 24 \times 3 \left( \frac{\zeta_0}{4\ell_z} \right)^2 \frac{D_{2222}^{(4)}}{\sqrt{\lambda_{zz}^2} \lambda_{zz}} \omega_z, \quad \Delta \omega_{zz} = \Omega_{SI}^{zz} + \frac{1}{2} \Omega_{4,2}^{zz}, \quad \Delta \omega_{zz} = \Omega_{SI}^{x^2} + \frac{1}{2} \Omega_{4,2}^{zz} \quad (3.62)$$

Here we introduced $\lambda_{zz} \equiv \lambda_{zz}$ to distinguish the eigenvalue associated to the $x$ zigzag mode. Later, when we investigate the dynamics of the zigzag mode near the linear-to-zigzag transition by 2D spectroscopy, the mode coupling $\Omega_{4,2}^{zz}$ will lead to dephasing of the zigzag mode for a thermal occupation of the rocking mode. Therefore, we call $\Omega_{4,2}^{zz}$ and other Kerr-type coupling strengths dephasing rates in the following. Note that for the parameters in Tab.3.2 $\Omega_{SI}^{zz}/2\pi \approx 25.5$kHz while $\Omega_{SI}^{x^2}/2\pi \approx 10$Hz. Hence, the self-interaction of the rocking mode is negligible.

More generally, all terms which do not involve the factor $1/\sqrt{\lambda_{zz}}$ are at least one order of magnitude smaller than the terms featuring this factor. This can be seen in Tab. 3.3 where we report the values of the normal mode frequencies and those of $1/\sqrt{\lambda_{nn,aa}}$ for the trapping parameters of Tab. 3.2. Accordingly, the effects of these terms are very weak and we neglect them for the remainder of our analysis.

Let us now turn to the remaining parts $H_{xy}$ and $H_{zz}$ from Eq. (3.58). Following the same steps we took to obtain the effective Hamiltonian for $H_{xx}$, we find that $H_{xy}$ can be approximated as

$$H_{xy} = a_{zz}^\dagger a_2 \left( \hbar \Delta \omega_{zz} + \sum_{n=2,3} \hbar \Omega_{4,2}^{yy} b_n^\dagger b_n + \sum_{n=2,3} \hbar \Delta \omega_{nn} b_n^\dagger b_n \right) \quad (3.64)$$
Table 3.3: Normal mode frequencies and magnitude of $1/\sqrt{\lambda_{n,\alpha}}$. The table lists the values of $\omega_{n,\alpha}$ for $N = 3^{40}\text{Ca}^+$ ions with the parameters in Tab. 3.2. The table also lists the corresponding values of $1/\sqrt{\lambda_{n,\alpha}}$ appearing in the fourth-order Hamiltonian $H^{(4)}$ in (3.54). The numerical values are rounded to the second digit.

<table>
<thead>
<tr>
<th>$\alpha \backslash n$</th>
<th>$\omega_{x,\alpha}/2\pi$</th>
<th>$\omega_{y,\alpha}/2\pi$</th>
<th>$\omega_{z,\alpha}/2\pi$</th>
<th>$1/\sqrt{\lambda_{1,\alpha}}$</th>
<th>$1/\sqrt{\lambda_{2,\alpha}}$</th>
<th>$1/\sqrt{\lambda_{3,\alpha}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$x$</td>
<td>3.101 MHz</td>
<td>2.370 MHz</td>
<td>0.132 MHz</td>
<td>0.64</td>
<td>0.84</td>
<td>15.16</td>
</tr>
<tr>
<td>$y$</td>
<td>5 MHz</td>
<td>4.583 MHz</td>
<td>3.924 MHz</td>
<td>0.40</td>
<td>0.44</td>
<td>0.51</td>
</tr>
<tr>
<td>$z$</td>
<td>2 MHz</td>
<td>3.464 MHz</td>
<td>4.817 MHz</td>
<td>1.0</td>
<td>0.58</td>
<td>0.42</td>
</tr>
</tbody>
</table>

where the dephasing rates and frequency shifts read

$$\Omega_{d,2}^{x} = 4 \times 6 \left(\frac{z_0}{4l_c}\right)^2 \frac{D_{332}^{(4)}}{\sqrt{\lambda_{zz}\lambda_{xy}}} \omega_{x}, \quad \Omega_{d,3}^{x} = 4 \times 6 \left(\frac{z_0}{4l_c}\right)^2 \frac{D_{333}^{(4)}}{\sqrt{\lambda_{zz}\lambda_{3y}}} \omega_{x},$$  \hspace{1cm} (3.65)

$$\Delta \omega_{n,x}^{y} = \frac{1}{2}(\Omega_{d,2}^{x} + \Omega_{d,3}^{y}), \quad \Delta \omega_{n,y}^{z} = \frac{\Omega_{d,2}^{x}}{2},$$  \hspace{1cm} (3.66)

Similarly, for $H_{xz}$ we find

$$H_{xz} = a_{xz}^\dagger a_{xz} \left(\hbar \Delta \omega_{n,x}^{y} + \sum_{n=2,3} \hbar \Omega_{d,n,\alpha}^{z} c_n^\dagger c_n + \sum_{n=2,3} \hbar \Delta \omega_{n,z} c_n^\dagger c_n, \right)$$  \hspace{1cm} (3.67)

where the dephasing rates $\Omega_{d,n}^{\alpha}$ are defined analogous to Eq. (3.65) multiplied by an additional factor of four and the frequency shifts $\Delta \omega_{n,\alpha}^{\alpha}$ and $\Delta \omega_{n,\alpha}$ are defined substituting $y$ by $z$ in Eq. (3.66).

We have compiled the value for the zigzag mode self-interaction, the mode frequency shifts and dephasing rates for the parameters of Tab. 3.2 in Tables 3.4 and 3.5. The frequency shift of the $x$ zigzag mode reported in Table 3.4 is the sum of the shifts in Eqs. (3.59), (3.64) and (3.67). It should be noted that the self-interaction and thus also the frequency shift of the $x$ zigzag mode increase fastest approaching the linear-to-zigzag transition since they scale as $1/\sqrt{\lambda_{zz}}$, while the mode couplings only scale as $1/\sqrt{\lambda_{zz}}$.

It turns out, though, that an analysis including only the fourth-order terms is incomplete. In Ref. [113] the observed mode shifts due to Kerr-type couplings could not be reproduced theoretically. In this reference only the fourth-order terms of the expansion of the Coulomb potential were included in the theoretical analysis. A more complete analysis [114] showed that the off-resonant third-order Hamiltonian causes energy shifts of the same order of magnitude as the fourth-order terms. Hence, for a complete analysis we need to include the Hamiltonian $H_{full}^{(3)}$ from (3.41) and compute the corrections to self-interactions, mode couplings and frequency shifts involving $x$ modes. The part of $H_{full}^{(3)}$ relevant for our analysis reads

$$H_{k}^{(3)} = 3 \frac{z_0}{4l_c} \hbar \omega_x \sum_{n,m,p} \frac{D_{nmp}^{(3)}}{\sqrt{\lambda_{nx}\lambda_{mx}\lambda_{px}}} (a_n + a_n^\dagger)(a_m + a_m^\dagger)(c_p + c_p^\dagger).$$  \hspace{1cm} (3.68)

The energy shifts induced by the off-resonant third-order terms are found in second-order perturbation theory [114]

$$\Delta E = \sum_{\langle n \rangle \neq \langle n' \rangle} \frac{|\langle n'|H_{k}^{(3)}\rangle \langle n \rangle|^2}{E_n - E_{n'}}.$$  \hspace{1cm} (3.69)

where $\langle n \rangle$ is a motional Fock state with energy $E_n$ and $\langle n' \rangle$ is any other motional Fock state with energy $E_{n'}$. Again we restrict our attention to terms involving the $x$ zigzag mode. The
we also neglect the self-interaction of the zigzag mode is the only self-interaction that appears in our further considerations. Except for the shift of the and 3.5. For the considered parameters we obtain an appreciable self-interaction and frequency diminishing the effects.

Table 3.4.: Shifts in the normal mode frequencies due third- and fourth-order effects of the Coulomb interaction for a crystal of three $^{40}$Ca$^+$ ions with trapping parameters as in Tab. 3.2. All quantities are given in kHz.

<table>
<thead>
<tr>
<th></th>
<th>$\Delta \omega_{\text{t},3}/2\pi$</th>
<th>$\Delta \omega_{\text{y},2}/2\pi$</th>
<th>$\Delta \omega_{\text{y},3}/2\pi$</th>
<th>$\Delta \omega_{\text{z},2}/2\pi$</th>
<th>$\Delta \omega_{\text{z},3}/2\pi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Third</td>
<td>-0.5008</td>
<td>-10.0850</td>
<td>0</td>
<td>0</td>
<td>0.5275</td>
</tr>
<tr>
<td>Fourth</td>
<td>0.4791</td>
<td>25.2874</td>
<td>0.0826</td>
<td>0.2894</td>
<td>-0.4371</td>
</tr>
<tr>
<td>Effective</td>
<td>-0.0217</td>
<td>15.2025</td>
<td>0.0826</td>
<td>0.2894</td>
<td>0.0905</td>
</tr>
</tbody>
</table>

Table 3.5.: Dephasing rates and self-interaction of the x zigzag mode due to third- and fourth-order effects of the Coulomb interaction for a crystal of three $^{40}$Ca$^+$ ions with trapping parameters as in Tab. 3.2. All quantities are given in kHz.

<table>
<thead>
<tr>
<th></th>
<th>$\Omega_{\text{d},3}/2\pi$</th>
<th>$(\Omega_{\text{SI}}/2)/2\pi$</th>
<th>$\Omega_{\text{d},2}/2\pi$</th>
<th>$\Omega_{\text{d},3}/2\pi$</th>
<th>$\Omega_{\text{d},2}/2\pi$</th>
<th>$\Omega_{\text{d},3}/2\pi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Third</td>
<td>-1.0487</td>
<td>-10.3467</td>
<td>0</td>
<td>0</td>
<td>1.0551</td>
<td>0.5171</td>
</tr>
<tr>
<td>Fourth</td>
<td>0.9582</td>
<td>12.9082</td>
<td>0.1652</td>
<td>0.5787</td>
<td>-0.8741</td>
<td>-1.8860</td>
</tr>
<tr>
<td>Effective</td>
<td>-0.0905</td>
<td>2.5615</td>
<td>0.1652</td>
<td>0.5787</td>
<td>0.1810</td>
<td>-1.3690</td>
</tr>
</tbody>
</table>

non-zero elements of $D_{\text{nnm}}^{(3)}$ that involve the x zigzag mode are $D_{233}^{(3)}$, $D_{123}^{(3)}$ and $D_{332}^{(3)}$. We note that there are no couplings to the modes in y-direction and as we have seen previously, there are also no couplings to the center-of-mass modes. Accordingly, the relevant quantum numbers are

$$n_x,2,n_x,3,n_z,2,n_z,3 \equiv n_x,n_z,n_{\text{str}},n_{\text{eg}},$$

(3.70)

where the subscripts stand for “tilt”, “zigzag”, “stretch” and “Egyptian”. This should make clearer which modes we are referring to. The state $|\{n\}\rangle \equiv |n_x,n_z,n_{\text{str}},n_{\text{eg}}\rangle$ is coupled to 14 different states by $H_x^{(3)}$ of Eq. (3.68). These are

$$|\{n'\}\rangle = |n_x \pm 1,n_z \pm 1,n_{\text{str}},n_{\text{eg}} \pm 1\rangle,$$

(3.71)

$$|\{n'\}\rangle = |n_x,n_z \pm 2,n_{\text{str}} \pm 1,n_{\text{eg}}\rangle,$$

(3.72)

$$|\{n'\}\rangle = |n_x,n_z,n_{\text{str}} \pm 1,n_{\text{eg}}\rangle.$$

(3.73)

Note that for the states $|\{n'\}\rangle$ where more than one $\pm$ appears all possible combinations of plus and minus signs are relevant. The numerical values for the corrections of the self-interaction, coupling rates and frequency shifts obtained from Eq. (3.69) for the parameters in Tab. 3.2 are given in Tables 3.4 and 3.5. When they are non-vanishing, the third-order corrections are of opposite sign than the fourth-order contributions diminishing the effects.

The effective self-interaction, dephasing rates and frequency shifts are obtained from summing the third- and fourth-order contributions and are displayed in the last rows of Tables 3.4 and 3.5. For the considered parameters we obtain an appreciable self-interaction and frequency shift of the x zigzag mode despite the cancellations due to the third-order contributions.

Before we close this section, let us briefly comment on the frequency shifts of the normal modes. Comparing the normal mode frequencies for the parameters in Tab. 3.2 reported in Tab. 3.3 with the frequency shifts due to higher-order corrections of the Coulomb potential in Tab. 3.4, we find that the relative frequency shifts are of the order $10^{-4}$ or less for all modes except for the x zigzag mode. Accordingly, we neglect these shifts in the following. Note that we also neglect the self-interaction of the x tilt mode, as we commented after Eq. (3.63). For ease of notation, we then write $\Omega_{\text{SI}}^{(3)} \equiv \Omega_{\text{SI}}$ in the following, since the self-interaction of the x zigzag mode is the only self-interaction that appears in our further considerations.
3.4. Population measurement for motional modes of trapped ion crystals

In summary, the appreciable effects of the fourth-order Hamiltonian in Eq. (3.54) for the parameters in Tab. 3.2 are a self-interaction and frequency shift of the $x$ zigzag mode as well as a Kerr-type coupling to other modes. The other effects in Eqs. (3.60), (3.64) and (3.67) are negligible. A thermal occupation of the modes coupled to the $x$ zigzag mode frequency and leads to effective dephasing for a thermal occupation of these modes. These considerations finally yield the effective fourth-order expansion Hamiltonian

$$H_{\text{eff}}^{(4)} = \frac{\hbar \Omega_{\text{SI}}}{2} (a_{zz}^\dagger)^2 a_{zz}^2 + \hbar \Delta \omega_{zz} a_{zz}^\dagger a_{zz} + \hbar \Omega_{d,n}^x a_{zz}^\dagger a_{zz} a_2 + a_{zz}^\dagger a_{zz} \sum_{n=2,3} \left( \hbar \Omega_{d,n}^x b_n^\dagger b_n + \hbar \Omega_{d,n}^z c_n^\dagger c_n \right).$$

(3.74)

Note again that $\Omega_{\text{SI}}, \Delta \omega_{zz}$ as well as the $\Omega_{d,n}^x, \alpha = x, y, z$ are the effective values in the last lines of Tables 3.4 and 3.5, which are obtained by adding the third- and fourth-order effects.

3.4. Population measurement for motional modes of trapped ion crystals

In the next section we present the 2D spectroscopy protocol that we propose for the investigation of nonlinear dynamics in crystals of trapped ions. The observable that we propose to measure at the end of the protocol is the mean occupation number of a motional mode, where $\hat{n} = a^\dagger a$ and $a$ is the destruction operator of the corresponding mode. In this section we present one method to determine the population of a motional mode of trapped ions. We will restrict our attention to the case of a single trapped ion. The method works in the same way for a normal mode.

Let us consider a single trapped ion whose internal levels can be described as a two-level system and the motion of the ion along one spatial direction. We assume that we are in the Lamb-Dicke regime for the corresponding motional mode and that the spin and motional degrees of freedom are in a product state initially

$$|\psi(0)\rangle = |g\rangle \sum_{n=0}^\infty c_n |n\rangle = \sum_{n=0}^\infty c_n |g, n\rangle,$$

(3.75)

where $c_n$ is the complex amplitude of the motional number state $|n\rangle$. Let us now assume that the system is driven by a laser tuned to the first red or blue sideband transition for a time $t$. The respective Hamiltonians are given in Eqs. (2.148) and (2.149) above. Interestingly, the dynamics of the red and blue sideband Hamiltonians can be solved analytically by means of a Laplace transform [39, 42] and the state at time $t$ takes the form

$$|\psi(t)\rangle = \sum_n c_n \left[ \cos \left( \frac{\Omega_{n,n+1} t}{2} \right) |g, n\rangle + \exp(i \phi) \sin \left( \frac{\Omega_{n,n+1} t}{2} \right) |e, n\rangle \right],$$

(3.76)

where $\phi$ is the laser phase. Now, a measurement of the observable $|e\rangle \langle e| \otimes \mathbb{1}$, where $\mathbb{1}$ is the unit operator of the motional mode, gives us the probability to find the system in state $|e\rangle$

$$p_e^{b/t}(t) = \sum_n |c_n|^2 \sin^2 \left( \frac{\Omega_{n,n+1} t}{2} \right) = \frac{1}{2} \left[ 1 - \sum_n p_n \cos \left( \Omega_{n,n+1} t \right) \right].$$

(3.77)

Measuring $p_e^{b}(t)$ for various times $t$, we obtain the initial populations $p_n = |c_n|^2$ by Fourier transforming $p_e^{b}(t)$ and can infer the mean occupation number from these.
A procedure especially well suited for thermal states is the following. Consider the probability to find the ion in the excited state \( p_r(t) \) for the red-sideband excitation. For a thermal initial state with mean occupation number \( \bar{n} \) the probability to find the system in the number state \( |n\rangle \) is given by

\[
p_n = \frac{\bar{n}^n}{(1+\bar{n})^{1+n}} \sin^2 \left( \frac{\Omega_{n,n-1} t}{2} \right)
\]

such that we have

\[
p_r(t) = \frac{\bar{n}}{1+\bar{n}} \sum_{n=1}^{\infty} \frac{\bar{n}^n}{(1+\bar{n})^{1+n}} \sin^2 \left( \frac{\Omega_{n+1,n} t}{2} \right)
\]

where we used \( \Omega_{n,n+1} = \Omega_{n+1,n} \) in the last step. Thus we find that the ratio between the excitation probabilities of the red and blue sideband relates to the mean occupation number \( \bar{n} \)

\[
R = \frac{p_r(t)}{p_b(t)} = \frac{\bar{n}}{1+\bar{n}}.
\]

From the above equation we obtain the mean occupation number as

\[
\bar{n} = \frac{R}{1-R}.
\]

Thus, the mean occupation number can be obtained from the ratio of the excitation probability on the red- and blue-sideband transitions for a thermal state.

### 3.5. Two-dimensional spectroscopy with trapped ions – protocol and theoretical analysis

In this section we present our 2D spectroscopy protocol for the investigation of anharmonic motional dynamics in crystals of trapped ions. The results presented in this section were published in [L1]. Accordingly, our presentation of the material follows along the same lines. Let us note that other approaches to apply 2D spectroscopy to trapped ions focusing on different aspects have been developed around the same time [106–108]. This section is organized as follows. We will first present the proposed 2D spectroscopy protocol for the investigation of nonlinear dynamics with trapped ions. In this context we will also discuss the concept of coherence transfer pathways that we have not introduced so far but which is of great importance for producing intelligible 2D spectra. Then, we proceed to show that the scheme yields a vanishing signal for purely harmonic time evolutions in the limit of weak excitation. While this section is devoted to the study of the protocol and its properties, the next section is reserved for a numerical study of possible implementations of the protocol.

#### 3.5.1. 2D spectroscopy protocol for the detection of nonlinear dynamics with trapped ions

The protocol we propose for 2D spectroscopy is of the general form of a multidimensional spectroscopy experiment as illustrated in Fig. 3.1 a). After preparation of the initial state this general form consists of a series of \( n \) electromagnetic pulses \( \hat{R}_k \) and subsequent free evolution periods \( t_k \) governed by a Liouvillian \( \hat{L}_k \), \( k = 1, \ldots, n \). The time evolution is then governed by Eq. (3.3). In the protocol we propose the pulses are small phase-controlled displacements of a motional mode

\[
\hat{R}_k \rho \equiv \hat{D}(\alpha_k) \rho \hat{D}(\alpha_k)^\dagger,
\]
where $D(\alpha_k)$ is the displacement operator introduced in Eq. (2.298) and $\alpha_k$ is a complex number
\[ \alpha_k = |\alpha_k| e^{i\phi_k}. \] (3.82)

The state at the end of the experimental protocol is then given by
\[ \rho(t_1, \ldots, t_n) = e^{\sum t_k \hat{D}_k} \rho. \] (3.83)

The observable to be measured at the end of the pulse sequence is the mean occupation number
\[ \bar{n} = \langle \hat{n} \rangle, \] where $\hat{n}$ is the number operator of the corresponding mode. One route to measure $\bar{n}$ for trapped ions was presented in Section 3.4. The signal for our protocol is thus given by
\[ s(t_1, \ldots, t_n) = \text{Tr}[\hat{n} \rho(t_1, \ldots, t_n)]. \] (3.84)

We should note that each of the displacements can be expanded as a series and contains all powers of the ladder operators acting on the state. In general, these different actions will lead to different time dependences during the free evolutions and thus different contributions in the spectrum. This can lead to overcrowded spectra. To alleviate this problem, we consider small displacements. Here, small means that we can approximate the displacement operators by expanding them to the first few orders in the $\alpha_k$. But even then there is a multitude of possible combinations of operators acting on the state during the pulses. It is therefore of vital importance that the phase of the displacements is controlled. Using \textit{phase cycling}, which we will discuss in the next section, one can select contributions to the final signal with a certain phase signature. In this way one can identify the operators acting during each pulse which allows us to understand the physical origin of the spectral peaks. Let us remark in closing that the first pulse only sets a reference for the phase of the subsequent pulses and accordingly we can set the phase of the first pulse to zero.

3.5.2. Phase cycling and coherence transfer pathways

Phase cycling is a procedure to postselect a specific contribution to a signal in a multidimensional spectroscopy experiment. In this section we will discuss phase cycling and the concept of coherence transfer pathways which is immediately connected to phase cycling. Here we adopt the concept of coherence transfer pathways from NMR and the presentation mostly follows the treatment in [30].

To begin let us consider the simplest 2D experiment for the protocol with trapped ions proposed in Eq. (3.83). It consists of two displacement pulses on a motional mode, each followed by a free evolution period. Let us assume for the moment that dissipative effects and dephasing do not play a role during the free evolution periods. The time evolution operator then reads
\[ U_0(t_1, t_2) = U_{\text{free}}(t_2) D(\alpha_2) U_{\text{free}}(t_1) D(\alpha_1). \] (3.85)

The state at the final time of the experiment is given by
\[ \rho(t_1, t_2) = U_0(t_1, t_2) \rho(0) U_0^\dagger(t_1, t_2), \] (3.86)
where $\rho(0)$ is the initial state. This brings us to the concept of coherence transfer pathways. Each of the displacement pulses in Eq. (3.86) can be expanded as a power series. For each displacement we pick only one operator term acting on the density matrix from each side. Note that this also includes the unit operator. Every possible combination of operators acting on the density matrix during the pulses together with the free evolutions forms a coherence transfer pathway. In our protocol only coherence transfer pathways that end up in a population contribute to the measured signal.
Figure 3.6.: Part a) shows a coherence transfer pathway for the simplest pulse sequence, Eq. (3.85), yielding a two-dimensional spectrum for the proposed protocol in Eq. (3.83). In the shown pathway during the first pulse \( a^\dagger \) acts from the left and the unit operator from the right. During the second pulse the unit operator acts from the left and \( a e^{-i\phi_2} \) from the right imprinting the phase \( -\phi_2 \) on the final population. Theoretically, \( |n+1\rangle\langle n+1| \) might be depleted during the evolution \( U(t_2) \) in part a) but we assume that this does not happen here. In part b) a coherence transfer pathway for an experiment consisting of four displacements and two free evolutions as in Eq. (3.92) below is shown. The phase \( \phi_2 - \phi_3 - \phi_4 \) is imprinted onto the final population. For both coherence transfer pathways population is transferred to observable population.

A simple example of a pathway for an experiment described by Eq. (3.86) is depicted in Fig. 3.6 a). For clarity only non-trivial actions on the state are displayed. Recall that the first displacement only sets a phase reference for the subsequent displacements and therefore its phase can be set to zero. In the illustrated sequence, we consider an action of \( a^\dagger \) from the left during the first displacement. The second displacement is now applied with a well defined phase so that \( \alpha_2 \) can be written as \( |\alpha_2|e^{i\phi_2} \). We consider the action of \( a \), which carries a factor \( e^{-i\phi_2} \), from the right during the second pulse. Thus, the displayed coherence transfer pathway yields a contribution to the final population with a phase \( e^{-i\phi_2} \). We say a phase \( -\phi_2 \) is imprinted on the final population.

Obviously, there are other pathways with different phases that also end up in a population contributing to the signal. Thus, the observable we measure in the end \( \langle \tilde{n} \rangle \) has contributions with different phase signatures \( e^{i\phi_2} \), where \( p \in \mathbb{Z} \). If we denote the probability to find the system in state \( |n\rangle \) in the final state of the experiment by \( c_n \), we may write the measured signal as

\[
s(t_1,t_2) = \langle \tilde{n} \rangle = \sum_n c_n n = \sum_n \sum_p c_{n,p} e^{ip\phi_2} = \sum_p e^{ip\phi_2} \sum_n c_{n,p} n = \sum_p s_p(t_1,t_2) e^{ip\phi_2}. \tag{3.87}
\]

Thus, we have separated the signal into contributions \( s_p(t_1,t_2) \) with different phase dependences \( e^{ip\phi_2} \). In order to make clear that the signal also depends on the value of \( \phi_2 \), we write the above equation as

\[
s(t_1,t_2,\phi_2) = \sum_p s_p(t_1,t_2) e^{ip\phi_2}. \tag{3.88}
\]

Note that the measurement result must be a real number and therefore we must have \( s_{-p}(t_1,t_2) = s_p^*(t_1,t_2) \). If we now want to obtain the contribution due to a certain coherence transfer pathway, e.g. the one illustrated in Fig. 3.6 a), we need to extract a certain contribution with phase \( e^{ip\phi_2} \) from the signal in Eq. (3.88). For the coherence transfer pathway in Fig. 3.6 a) this would be the \( q = -1 \) contribution. In order to be able to isolate this contribution, we apply phase cycling.

90
Phase cycling works as follows. One performs $N_{\phi_2}$ experiments varying the phase $\phi_2$ systematically such that in experiment $k$ it is given by

$$\phi_{2,k} = k \frac{2\pi}{N_{\phi_2}}, \quad k = 0, \ldots, N_{\phi_2} - 1. \quad (3.89)$$

Each of the experiments yields a signal of the form in Eq. (3.88). The contribution from the pathways with phase signature $q\phi_2$ is obtained from the data collected in the $N_{\phi_2}$ experiments by a discrete Fourier transform

$$s(t_1, t_2, q\phi_2) = \frac{1}{N_{\phi_2}} \sum_{k=0}^{N_{\phi_2}-1} s(t_1, t_2, \phi_{2,k}) e^{-iq\phi_{2,k}}$$

$$= \frac{1}{N_{\phi_2}} \sum_{k=0}^{N_{\phi_2}-1} \left[ \sum_p s_p(t_1, t_2) e^{ip\phi_{2,k}} \right] e^{-iq\phi_{2,k}}. \quad (3.90)$$

However, phase cycling does not only isolate the contribution with phase factor $q\phi_2$. It is clear from Eq. (3.90) that also all contributions with phase $(q + rN_{\phi_2})\phi_2$, where $r \in \mathbb{Z}$, are extracted. Hence, the signal we obtain after phase cycling is given by

$$s(t_1, t_2, q\phi_2) = s_q(t_1, t_2) + \sum_r s_{q+rN_{\phi_2}}(t_1, t_2). \quad (3.91)$$

This might seem problematic at first sight. However, the unwanted but selected contributions $s_{q+rN_{\phi_2}}$ are due to terms of higher orders in the $\alpha_k$ in the expansion of the displacements. If the moduli $|\alpha_k|$ are taken sufficiently small, only the first few terms of the expansion of the displacements are non-negligible and only the desired pathways contribute. If the $|\alpha_k|$ for some reason cannot be made smaller than a certain value, one can increase $N_{\phi_2}$ such that the contribution of the selected but unwanted pathways to the signal is negligible. To summarize, the condition that only the pathways with a desired phase signature contribute to the signal for a given $N_{\phi_2}$ defines what we mean by small displacements in our protocol. Apparently, what is considered “small” depends on the context. Finally, phase cycling is a nice asset to isolate only a number of contributions to the signal but it comes at the cost of increasing the number of experiments that have to be performed by a factor $N_{\phi_2}$.

Phase cycling can also be extended to sequences involving more than one phase. The pulse sequence we propose to investigate the third- and fourth-order effects of the Coulomb potential is not as simple as the one we considered in Eq. (3.85). It consists of four displacements and two free evolution periods. This means that we set the free evolution time after two pulses to zero. Again neglecting noise effects the time evolution operator under this sequence reads

$$U_0(t_1, t_3) = D(\alpha_1)U_{\text{free}}(t_3)D(\alpha_2)D(\alpha_3)D(\alpha_4)U_{\text{free}}(t_1)D(\alpha_1), \quad (3.92)$$

where we have set $t_2 = t_4 = 0$ and

$$\alpha_k = |\alpha_k|e^{i\phi_k}, \quad k = 2, 3, 4. \quad (3.93)$$

A coherence transfer pathway for the sequence in Eq. (3.92) is illustrated in part b) of Fig. 3.6. For the displayed pathway the phase $\phi_2 - \phi_3 - \phi_4$ is imprinted onto the final population $|n + 1\rangle (n + 1)$. We can isolate the part of the signal with this phase signature by performing phase cycling for each of the three phases independently. This means we need to perform $N_{\phi_i}$ experiments for each phase $\phi_i$ varying $\phi_i$ as in Eq. (3.89). Defining $q = (q_2, q_3, q_4)$ and $\phi_k = (\phi_{2,k}, \phi_{3,k}, \phi_{4,k})$, we obtain the signal with phase signature $e^{iq\phi}$ through

$$s(t_1, t_3, q\phi) = \frac{1}{N_{\phi_0}} \sum_{k_2} \sum_{k_3} \sum_{k_4} s(t_1, t_3, \phi_k) e^{-iq\phi_k}, \quad (3.94)$$
3. Two-dimensional spectroscopy for the study of Coulomb crystals

where \( N_{pc} = N_{\phi_1} \cdot N_{\phi_2} \cdot N_{\phi_3} \). The signal with phase signature \( \phi_2 - \phi_3 - \phi_4 \) is obtained for \( \mathbf{q} = (1, -1, -1) \). Note that now the number of experiments one needs to perform is increased by a factor \( N_{pc} \).

3.5.3. Proof of vanishing harmonic signal

In Eq. (3.92) we have already stated the pulse sequence we propose for the investigation of non-linear dynamics in crystals of trapped ions. In this section we want to show that this sequence, under the assumption that the displacements can be approximated by their first order expansion in the \( \alpha_k \), yields a vanishing signal for a purely harmonic time evolution during the free evolution periods.

We start by considering the dynamics of the addressed mode only and assume a unitary time evolution. According to Eq. (3.92) the time evolution operator of the sequence is given

\[
U_0(t_1,t_3) = D(\alpha_4)U_{\text{free}}(t_3)D(\alpha_3)D(\alpha_2)U_{\text{free}}(t_1)D(\alpha_1). \tag{3.95}
\]

We assume that the displacement operators can effectively be written as

\[
D(\alpha_k) = 1 + |\alpha_k| (e^{i\phi_k} a^\dagger - e^{-i\phi_k} a),
\]

where \( a \) is the annihilation operator of the addressed mode. The argument leading to the above approximation is that terms of higher order in \( |\alpha_k| \) are either canceled by phase cycling or yield a negligible contribution to the spectrum since the \( N_{\phi_k} \) are chosen large enough and \( |\alpha_k| \ll 1 \).

The signal at the end of the sequence is given by

\[
s(t_1,t_3) = \text{Tr}[\hat{n}U_0(t_1,t_3)\rho_0 U_0^\dagger(t_1,t_3)], \tag{3.97}
\]

where \( \hat{n} = a^\dagger a \) is the number operator of the displaced mode and we used \( t_2 = t_4 = 0 \). \( \rho_0 = \rho(0) \) is the initial state of the mode. Using the cyclic property of the trace we may write the above equation as

\[
s(t_1,t_3) = \text{Tr}[\rho_0 U_0^\dagger(t_1,t_3)\hat{n}U_0(t_1,t_3)]. \tag{3.98}
\]

We will now focus on the term \( U_0^\dagger(t_1,t_3)\hat{n}U_0(t_1,t_3) \) and split the time evolution operator \( U_0(t_1,t_3) \) in two parts \( U_0(t_1,t_3) = D(\alpha_3)U_1 \), where \( U_1 = U_{\text{free}}(t_3)D(\alpha_2)D(\alpha_1)U_{\text{free}}(t_1)D(\alpha_4) \). Let us assume that we select the terms with phase \( e^{-i\phi_k} \) by phase cycling. Expanding the displacement operator as in Eq. (3.96) and keeping only terms with phase \( e^{-i\phi_k} \), we obtain

\[
U_0^\dagger(t_1,t_3)\hat{n}U_0(t_1,t_3) = |\alpha_4| e^{-i\phi_4} U_1^\dagger [a, \hat{n}] U_1 = |\alpha_4| e^{-i\phi_4} U_1^\dagger a U_1. \tag{3.99}
\]

Next we write \( U_1 = U_{\text{free}}(t_3)U_2 \), where now \( U_2 = D(\alpha_3)D(\alpha_2)U_{\text{free}}(t_1)D(\alpha_1) \). For a harmonic time evolution, i.e. for a time evolution under a Hamiltonian at most quadratic in \( a \) and \( a^\dagger \), we have

\[
U_{\text{free}}^\dagger(t_3) a U_{\text{free}}(t_3) = f(a,a^\dagger), \tag{3.100}
\]

where \( f(a,a^\dagger) \) is a linear function of \( a \) and \( a^\dagger \). Inserting Eq. (3.100) into Eq. (3.99) yields

\[
U_0^\dagger(t_1,t_3)\hat{n}U_0(t_1,t_3) = |\alpha_4| e^{-i\phi_4} U_2^\dagger f(a,a^\dagger) U_2. \tag{3.101}
\]

We now write \( U_2 \) in Eq. (3.101) as \( U_2 = D(\alpha_3)U_3 \), where \( U_3 = D(\alpha_2)U_{\text{free}}(t_1)D(\alpha_1) \), and use the expansion in Eq. (3.96) for the displacement operator. Assuming we select the terms with phase \( e^{-i\phi_k} \) we obtain

\[
U_0^\dagger(t_1,t_3)\hat{n}U_0(t_1,t_3) = |\alpha_3| |\alpha_4| e^{-i(\phi_3 + \phi_4)} U_3^\dagger [a,f(a,a^\dagger)] U_3 = |\alpha_3| |\alpha_4| e^{-i(\phi_3 + \phi_4)} U_3^\dagger eU_3, \tag{3.102}
\]

\( 92 \)
3.6. Two-dimensional spectroscopy with trapped ions – simulations of experimental implementations

where \( c \in \mathbb{C} \) is a constant. Here, in the second equality we used that \( f(a,a^\dagger) \) is linear in the creation and annihilation operators such that \([a,f(a,a^\dagger)] = cI\) for some \( c \in \mathbb{C} \). We then write \( U_3 = D(\alpha_2)U_4 \), where \( U_4 = U_{\text{free}}(t_1)D(\alpha_1) \). With the expansion in Eq. (3.96) for the displacement operator and assuming that we select the terms with phase \( e^{i\phi_2} \), we finally obtain

\[
U_0^\dagger(t_1,t_3)nU_0(t_1,t_3) = -|\alpha_2||\alpha_3||\alpha_4|e^{i(\phi_2-\phi_3-\phi_4)}[a^\dagger,cI]U_4 = 0. \tag{3.103}
\]

Thus, under the assumption that the approximation Eq. (3.96) holds there will be no signal for a purely harmonic time evolution for the 2D spectroscopy protocol in Eq. (3.95). Here, we considered the coherence transfer pathways with a phase signature \( \phi_2 - \phi_3 - \phi_4 \). The calculation follows the same steps for coherence transfer pathways with different phase signatures. The key point is the fact that a harmonic time evolution maps creation and annihilation operators to linear combinations of creation and annihilation operators, which we used in Eq. (3.100).

The calculation above can be generalized to a system consisting of several modes evolving under a quadratic Hamiltonian. Then, in general the creation and destruction operators will be mapped to a linear superposition of creation and annihilation operators of all modes. The function \( f(a,a^\dagger) \) then becomes a function \( f(\{a_i,a_i^\dagger\}) \). Since the ladder operators of different modes commute the steps from Eq. (3.101) to Eq. (3.103) remain the same and we recover the result.

This reasoning can also be extended to the case where the system is in contact with some environment leading to a possibly non-unitary but linear time evolution. In this case one starts from the evolution of the full system including the environment whose time evolution is unitary and the same argument as above can be applied.

3.6. Two-dimensional spectroscopy with trapped ions – simulations of experimental implementations

After we have introduced the protocol for 2D spectroscopy with trapped ions and discussed some of its properties, we now proceed to show how it can be applied in practice. To this end we simulate two experiments for parameters which are realistic for a state-of-the-art trapped-ion experiment. The resulting spectra reveal nonlinear dynamics due to resonant terms in the third- and fourth-order expansion of the Coulomb potential, respectively. More generally, the simulated experiments show that 2D spectroscopy can be applied to trapped ions complementing standard spectroscopic methods in the trapped-ion toolbox. The first of the two experiments investigates the self-interaction of the zigzag mode of a crystal approaching the linear-to-zigzag transition, while the second simulated experiment witnesses resonant energy transfer between the normal modes. We include different sources of noise in the simulations showing that 2D spectroscopy provides us with information about the nature of the noise and that it can reveal the relevant information despite the noise. Finally, we show that the protocol is robust with respect to another source of experimental noise, which is laser phase fluctuations. Thereafter, we close this chapter with a discussion of future directions for 2D spectroscopy with trapped ions.

3.6.1. Signatures of the onset of a structural transition from 2D spectroscopy

The linear-to-zigzag structural transition is a paradigmatic example where nonlinear effects play a role in the dynamics of trapped ions. In particular, we have seen in Eq. (3.35) that close to the transition point the potential of the zigzag mode is approximately given by a fourth-order potential. A fourth-order expansion of the Coulomb potential in the small displacements of the ions
from equilibrium (see Section 3.3.3) confirmed that for a crystal in the linear phase an appreciable fourth-order contribution to the potential of the zigzag mode emerges when approaching the linear-to-zigzag transition. Neglecting energy non-conserving terms this fourth-order contribution yields a self-interaction of the zigzag mode and a shift of the mode frequency. In this section we will illustrate how 2D spectroscopy can be used to observe and quantify this self-interaction which arises close to the structural transition.

The smallest crystal that features a linear-to-zigzag transition is a three-ion crystal. In Section 3.3.3 we considered a three-ion crystal of $^{40}$Ca$^+$ ions in the linear phase approaching the linear-to-zigzag transition. We saw that in addition to the fourth-order effects also off-resonant contributions from the third-order expansion of the Coulomb potential can have appreciable effects. For the considered parameter regime the third-order effects actually diminished the fourth-order effects.

In particular, in Section 3.3.3 we assumed that the radial potential in one direction, $x$, is relaxed while the other potentials are kept constant. For the trapping potentials reported in Tab. 3.2 we found the effective Hamiltonian $H_{\text{eff}}^{(4)}$ in Eq. (3.74) which we now write as

$$H_{\text{eff}}^{(4)} = H_{\text{SI}} + H_d. \quad (3.104)$$

Here, the Hamiltonian $H_{\text{SI}}$ accounts for the self-interaction and frequency shift of the zigzag mode

$$H_{\text{SI}} = \frac{\hbar \Omega_{\text{SI}}}{2} (a_{xz}^\dagger a_{xz})^2 + \hbar \Delta \omega_{zz} a_{xz}^\dagger a_{xz} \quad (3.105)$$

and $H_d$ describes the couplings to other modes

$$H_d = a_{xz}^\dagger a_{xz} \left[ \frac{\hbar \Omega_{\text{d,zz}}}{2} a_{zz}^\dagger a_{zz} + \sum_{n=2,3} \left( \frac{\hbar \Omega_{\text{d,n}}}{2} b_n^\dagger b_n + \hbar \Omega_{\text{d,n}}^\dagger c_n^\dagger c_n \right) \right] \quad (3.106)$$

that causes dephasing of the zigzag mode for thermal populations of the other modes. Note that we refer to the $x$ zigzag mode when we write “zigzag mode” here and in the following. If we refer to the $y$ zigzag mode, we will state it explicitly. Let us recall for completeness that $a_{zz}$ and $a_{xz}^\dagger$ are the ladder operators for the zigzag mode while $b_n$, $b_n^\dagger$ and $c_n$, $c_n^\dagger$ are those for the $y$- and $z$-modes, respectively.

In order to determine the self-interaction of the zigzag mode we consider the 2D spectroscopy protocol of Eq. (3.92)

$$U_0(t_1, t_3) = D(\alpha_4)U_{\text{free}}(t_3)D(\alpha_3)D(\alpha_2)U_{\text{free}}(t_1)D(\alpha_1). \quad (3.107)$$

It consists of four small displacements on the zigzag mode and two free evolution periods. The displacements of the zigzag mode could be implemented, for example, by applying suitable fields to the trap electrodes or by optical dipole forces which can also be produced on optical transitions [117]. For the parameters of Tab. 3.2 the free evolution periods are governed by the Hamiltonian $H_{\text{eff}}^{(4)}$ in Eq. (3.104). At the end of the sequence the zigzag mode’s population is measured. The signal for an initial state $\rho_0$ is thus given by

$$s(t_1, t_3) = \text{Tr}\{\hat{n}_{zz} U_0(t_1, t_3) \rho_0 U_0^\dagger(t_1, t_3)\}, \quad (3.108)$$

where $\hat{n}_{zz}$ is the number operator of the zigzag mode.

For the simulation of an experiment we assume $|\alpha_4| = 0.25$ for all displacements and consider the signal due to coherence transfer pathways with the phase signature $\phi_2 - \phi_3 - \phi_4$. We estimate that the signal due to these pathways is filtered out by cycling each phase $N_{\phi_4} = 4$ times.
3.6. Two-dimensional spectroscopy with trapped ions – simulations of experimental implementations

Figure 3.7.: Examples of coherence transfer pathways carrying the phase signature $\phi_2 - \phi_3 - \phi_4$ for the pulse sequence in Eq. (3.107), which allow us to infer the self-interaction strength $\Omega_{\text{SI}}$ of the zigzag mode close to the linear-to-zigzag transition. For clarity we set $a_{zz} \equiv a$, $|n\rangle_{zz} \equiv |n\rangle$ in the figure. For the path in part a) the coherences oscillate with frequency $-(\omega_{zz} + n\Omega_{\text{SI}})$ during both free evolution periods giving rise to diagonal peaks in the spectrum. For the path in part b) the oscillation frequency during $t_3$ is shifted by $-\Omega_{\text{SI}}$ with respect to $t_1$. Accordingly, the peaks due to this pathway are shifted by $-\Omega_{\text{SI}}$ along the $\omega_3$-axis leading to off-diagonal peaks below the main diagonal. The distance between diagonal peaks along both frequency axes as well as the shift along the $\omega_3$-axis would allow us to determine $\Omega_{\text{SI}}$. Note that in the last displacement the action of the operator can always act from either side because both actions lead to a population.

We show two coherence transfer pathways with the phase signature $\phi_2 - \phi_3 - \phi_4$ in Fig. 3.7. The pathway in part a) of the figure shows a coherence transfer pathway which has the same time dependence $-(\omega_{zz} + n\Omega_{\text{SI}})$ during both free evolution periods $t_1$ and $t_3$. It will thus lead to peaks on the diagonal of the spectrum separated by $\Omega_{\text{SI}}$ along each of the frequency axes. From the separation of these diagonal peaks one could thus infer $\Omega_{\text{SI}}$. For the pathway in part b) of the figure the oscillation frequency during the second evolution period $t_3$ is shifted by $-\Omega_{\text{SI}}$ with respect to the oscillation frequency during $t_1$. Thus, we expect off-diagonal peaks. The shift of these peaks along the $\omega_3$-axis with respect to the diagonal of the spectrum also allows us to determine $\Omega_{\text{SI}}$. Note that there is only a contribution of the self-interaction $H_{\text{SI}}$ of the fourth-order Hamiltonian to the time evolution of the coherence transfer pathway in Fig. 3.7 a) for $n \geq 1$.

In order to confirm these considerations, we simulated an experiment for a crystal of three $^{40}\text{Ca}^+$ ions with the trapping potentials in Tab. 3.2. For these potentials the free evolution periods of the pulse sequence in Eq. (3.107) are governed by the effective Hamiltonian $H_{\text{eff}}^{(4)}$ in Eq. (3.104). The parameters relevant for the simulation are listed in Tab. 3.6 below. The chosen parameters should be sufficiently far from the linear-to-zigzag transition that the perturbative expansion of the Coulomb potential is valid and that the crystal remains in the linear configuration during the experiment. Note that we have not taken into account micromotion in our analysis which might alter the values of the parameters in Tab. 3.6 slightly [118]. This, however, does not affect the general concept we present.

The effective Hamiltonian in Eq. (3.104) incorporates coupling of the zigzag mode to five other modes by the Kerr-type interactions in $H_d$. The effective dephasing rates for the considered parameters are summarized in Tab. 3.5 above. The dephasing rates due to the Egyptian and $y$ zigzag modes are much stronger than those due to the stretch and the $x$ and $y$ tilt modes. Therefore, we include only the former two in our simulations. We assume that the modes are
initialized close to their motional ground states by laser cooling. For the chosen parameters we obtain a zigzag mode frequency $\omega_{zz}/2\pi \approx 132\text{kHz}$. Cooling should still be possible for this frequency. Alternatively, one might cool the mode for a stiffer potential in $x$ and then relax the potential adiabatically. We assume an initial product state with each of the modes in a thermal state as given in Eq. (2.367) with mean occupation numbers $\bar{n}_{zz} = 1$ and $\bar{n}_{yz} = \bar{n}_{eg} = 4$. This guarantees an appreciable population of levels with $n \geq 1$ for the zigzag mode. The motional Hilbert spaces are truncated including nine energy levels for the zigzag mode and 15 for the other two. This corresponds to including 99% and 97% of the respective populations. Finally, we consider maximal evolution times $t_{\text{max}} = 2\text{ms}$ and a time step $\Delta t_{1/3} = 2.5\mu s$. We estimate dephasing due to the thermal occupation of the $y$ zigzag and Egyptian modes to be the dominant source of noise and therefore neglect heating of the motional modes during the experiments. All parameters relevant for the simulation are summarized in Tab. 3.6.

The spectrum resulting from the simulated experiment is presented in Fig. 3.8. We can observe two dominant lines: one along the principal diagonal and one shifted below it. For the diagonal peaks the coherences oscillate with the same frequency during $t_1$ and $t_3$. The pathways illustrated in Fig. 3.7 a) are an example of a family of pathways that leads to such diagonal peaks. The coupling of the zigzag mode to the other modes through $H_d$ in Eq. (3.106) leads to effective static disorder that produces shifts of the zigzag mode frequency. These shifts can be taken to be constant during a single experimental run but fluctuate from one experimental run to the other. This inhomogeneous broadening leads to the ridge of peaks along the diagonal. We have seen this effect already in Fig. 3.4 above. Obviously, the same effect leads to the ridges along the off-diagonals. The pronounced line below the diagonal is shifted by an amount $-\Omega_{SI}$ along the $\omega_3$-axis with respect to the principal diagonal. The coherence transfer pathways given in Fig. 3.7 b) are an example for pathways that lead to these off-diagonal peaks. Here, the oscillation frequency of the coherences is shifted by $-\Omega_{SI}$ during the second free evolution period $t_3$. Hence, the shift of this line with respect to the main diagonal gives direct access to the self-interaction strength $\Omega_{SI}$.

The spectrum in Fig. 3.8 illustrates some advantages of 2D spectroscopy. By projecting the spectrum along one of the two frequency axes we obtain the information a 1D spectroscopy experiment with only one free evolution period would have yielded. These projections are shown along the $\omega_1$ and $\omega_3$ axes. Note that we took the full complex signal to calculate the projections and only computed the absolute value in the end. Obviously, we would not have obtained $\Omega_{SI}$ from these. The two-dimensional spectrum, however, allows us to access the value of $\Omega_{SI}$ despite the strong dephasing due to the other modes illustrating a clear benefit of 2D spectroscopy.

Let us remark again that the coherence transfer pathways in Fig. 3.7 without dephasing would produce a series of separated peaks. As we mentioned earlier, the blurring of the maxima along the diagonal is caused by dephasing due to thermal occupation of the other modes. This dephasing could be much reduced by assuming all other modes are ground-state cooled. The desired information would then be available along the diagonal maybe even in a 1D experiment.\footnote{The displayed figure was published in [L1]. Copyright (2015) by the American Physical Society.}
3.6. Two-dimensional spectroscopy with trapped ions – simulations of experimental implementations

Figure 3.8.: Central plot: 2D spectrum \( |S(\omega_1, \omega_3)| = |\mathcal{F}(s(t_1,t_3))| \) obtained for the four-pulse sequence of Eq. (3.107) probing the nonlinear dynamics of a linear crystal of three \(^{40}\text{Ca}^+\) ions in the neighborhood of the linear-zigzag transition. The Hamiltonian governing the free evolution periods is given in Eq. (3.104) and includes up to fourth-order terms in the Coulomb potential. The simulation parameters are summarized in Tab. 3.6. The peaks along the diagonal and the pronounced line below are due to coherence transfer pathways of the type displayed in Fig. 3.7 a) and b), respectively. The peaks are blurred in diagonal direction due to static dephasing caused by thermal populations of the spectator modes. The line below the main diagonal is shifted by \(-\Omega_{SI}\) along the \(\omega_3\)-axis and thus allows us to extract the self-interaction strength from the spectrum. Note that the origin of the spectrum was adjusted such that the diagonal lines are centered in the image. Small plots along \(\omega_1/3\): These are the spectra obtained by integrating the full spectrum along the other frequency direction. They would be obtained by a 1D experiment with only one free evolution period \( |S(\omega_1/3)| = |S(\omega_1/3,t_3/1 = 0)| \).

We considered a thermal occupation for the other modes for two reasons: The first is that the inhomogeneous broadening in the diagonal direction provides us with information about the mechanism of the noise. In the present case it shows that we are dealing with fluctuations in the mode frequency from one experimental run to the other in contrast to fluctuations during each experimental run. Second, all modes except for the center-of-mass modes contribute to the dephasing where, of course, some contributions are relatively small. Here we have seen, however, that already two modes can have quite an impact. Hence, in order to obtain sharp and well-separated resonances without dephasing in the spectrum, one would need to ground-state cool all of the modes. This is experimentally very demanding and also very time consuming for large crystals of trapped ions.

A little extra thought also allows us to find coherence transfer pathways that lead to the weaker lines in the 2D spectrum in Fig. 3.8. The second line below the diagonal is shifted by \(-2\Omega_{SI}\) along the \(\omega_3\)-axis with respect to the main diagonal. Part a) of Fig. 3.9 shows a family of pathways leading to peaks on this second line below the main diagonal. Note that the action of the operators during the second displacement is of order \(|\alpha|^3\) which leads to a weaker signal but shows that these contributions are not completely negligible for our choices of \(|\alpha_k|\) and \(N_{\phi_k}\). The line above the main diagonal is shifted by \(+\Omega_{SI}\) along the \(\omega_3\)-axis. An example for pathways...
leading to peaks along the line above the diagonal is given in Fig. 3.9 b).

The results we presented here are dependent on the capability to control the laser phase such that all pulses can be applied with a well-defined phase with respect to the previous pulses. In our simulations we assumed that the phases can be controlled arbitrarily well. In practice this is not the case. The laser phase will inevitably fluctuate in time. We address this issue in Section 3.6.3 below. For the moment we only state that based on our model for laser phase fluctuations we find that the loss in contrast is as little as 1% for the signal of the coherence transfer pathways we considered in Fig. 3.7. Consequently, also the spectrum in Fig. 3.8 will be largely unaffected by this type of noise.

### 3.6.2. Resonant energy transfer between normal modes due to third-order effects of the Coulomb potential investigated by 2D spectroscopy

In this section we will study nonlinear dynamics due to resonant third-order terms of the Coulomb potential by 2D spectroscopy. The effects can be observed using the same experimental setup and protocol as in the previous section. We again consider a linear crystal of three $^{40}\text{Ca}^+$ ions where the degeneracy between the radial modes is lifted. We assume that the potential in $y$ is sufficiently stiff that no resonances involving the radial $y$ modes occur. On the other hand we assume that the anisotropy between the trapping potentials in $x$ and $z$ is tuned to $\xi_{xz} = (\omega_z/\omega_x)^2 = 20/63$. For this anisotropy there is a resonant coupling between the $x$ zigzag mode and the axial stretch mode. This resonant coupling is described by the effective third-order Hamiltonian of Eq. (3.47)

$$H_{\text{res}}^{(3)} = \hbar \Omega_T \left[ a_{zz}^\dagger c_{\text{str}}^\dagger + (a_{zz}^\dagger)^2 c_{\text{str}} \right]$$

(3.109)

where $\Omega_T = 3\zeta_0 \omega_0 D_{33}^{(3)} / (4l_3 \sqrt{\lambda_{zz}^2 \lambda_{zz}'})$. Note again that we write $a_{zz}$ and $c_{\text{str}}$ for the destruction operators of the $x$ zigzag and stretch modes. As in the previous section we refer to the $x$ zigzag mode simply as “zigzag mode”. If we refer to the $y$ zigzag mode, we will state it explicitly. For the regime we consider, all other terms in the third-order Hamiltonian, Eq. (3.41), are negligible.
Table 3.7.: Simulation parameters for the 2D spectrum probing third-order effects of the Coulomb potential displayed in Fig. 3.10.

<table>
<thead>
<tr>
<th>$\omega_c/2\pi$</th>
<th>$\omega_r/2\pi$</th>
<th>$\omega_{t}/2\pi$</th>
<th>$\Omega_T/2\pi$</th>
<th>$\Delta t_{1/3}$</th>
<th>$\bar{n}_t$</th>
<th>$N_h$</th>
</tr>
</thead>
<tbody>
<tr>
<td>2MHz</td>
<td>3.55MHz</td>
<td>5MHz</td>
<td>5.9kHz</td>
<td>2000 $\mu$s</td>
<td>10.6 $\mu$s</td>
<td>0.25</td>
</tr>
</tbody>
</table>

as well as the effects due to the fourth-order Hamiltonian, Eq. (3.54). We also note again that a third-order Hamiltonian can only be an approximation for small excitation numbers. In general, fourth-order terms have to be taken into account to guarantee stability.

In order to investigate the interaction in Eq. (3.109) with 2D spectroscopy, we rely on the pulse sequence in Eq. (3.107) we used to investigate the self-interaction of the zigzag mode in the previous section

$$U_0(t_1,t_3) = D(\alpha_1)U_{\text{free}}(t_3)D(\alpha_3)D(\alpha_2)U_{\text{free}}(t_1)D(\alpha_1).$$  \hspace{1cm} (3.110)

Also in the present case we assume that the displacements are applied to the zigzag mode and a measurement of the zigzag modes population completes the measurement cycle. The signal at the end of the sequence is therefore given by Eq. (3.97) with $\hat{n} = \bar{n}_{zz}$. Again we choose $|\alpha_1| = 0.25$ and analyze the contribution to the signal due to pathways with phase signature $\phi_2 - \phi_3 - \phi_4$, where each of the phases is cycled $N_{th} = 4$ times. Furthermore, we consider $\omega_r/2\pi = 2$ MHz which leads to $\omega_{t}/2\pi = 3.55$ MHz and $\Omega_T/2\pi = 5.9$ kHz for the considered anisotropy. The maximal times of the free evolution remain $t_{1/3}^{\text{max}} = 2$ ms but with a reduced time step $\Delta t_{1/3} = 10.6 \mu s$ for the interrogation. The parameters are summarized in Table 3.7.

For the simulation we assume an initial product state for the two modes with each of the modes in a thermal state, Eq. (2.367), with mean occupation numbers $\bar{n}_{zz} = 0.7$ and $\bar{n}_{\text{str}} = 0.2$, respectively. We neglect dephasing due to coupling to other modes in this simulation but now include heating of the ions in the trap as a source of decoherence. The main reason to neglect dephasing in this simulations is to illustrate the effect of heating on 2D spectra. On the other hand, the dephasing rates in the resonant terms of fourth-order Hamiltonian are small for the considered parameters and by assuming all modes are initialized close to their ground states we can safely neglect this type of dephasing.

The dynamics of an oscillator coupled to a thermal reservoir is described by the master equation (2.366) above [69]. Thus, the time evolution of the system during the free evolution periods is given by

$$\dot{\rho} = -\frac{i}{\hbar} [H^{(3)}_\text{osc}, \rho] + \sum_{n=\text{str,zz}} \left( 2\kappa_n (\bar{n}_{\text{eq},n} + 1)[a_n \rho a_n^+ - \frac{1}{2} \{a_n^+, a_n, \rho\}] + 2\kappa_n \bar{n}_{\text{eq},n} [a_n^+ \rho a_n - \frac{1}{2} \{a_n a_n^+, \rho\}] \right).$$ \hspace{1cm} (3.111)

In order to solve the dynamics, we need to set the heating rates $2\kappa_n$ and equilibrium mean occupation numbers $\bar{n}_{\text{eq},n}$ of the modes. We assume that the modes are coupled to a room temperature reservoir with $T_c = 300$ K. The equilibrium mean occupation numbers are then given by [69]

$$\bar{n}_{\text{eq},n} = \frac{1}{e^{\frac{\hbar}{2} \kappa_n} - 1}.$$ \hspace{1cm} (3.112)

Now, it remains to determine the rates $2\kappa_n$. If a single mode is subject to a dissipator of the form on the right hand side of Eq. (3.111), the mean occupation number obeys [69]

$$\bar{n}_n(t) = \bar{n}_{0,n} e^{-2\kappa t} + \bar{n}_{\text{eq},n}(1 - e^{-2\kappa t}).$$ \hspace{1cm} (3.113)
Two-dimensional spectroscopy for the study of Coulomb crystals

Figure 3.10.: a) 2D spectrum due to the pulse sequence in Eq. (3.110) probing third-order effects of the Coulomb potential. During the free evolution periods the evolution is governed by Eq. (3.111) including resonant third-order terms in the Coulomb potential and heating of the modes. The simulation parameters are reported in Tab. 3.7 and the text. A strong peak at $\omega_1 = \omega_3 = -\omega_{zz}$ was removed from the spectrum to enhance the contrast. The off-diagonal peaks witness resonant energy transfer between the zigzag and stretch modes and eigenvalues of $H_{\text{res}}^{(3)}$ for states with small occupation numbers of the modes can be identified in the spectrum. Homogeneous broadening of the peaks, i.e. broadening of the peaks along the vertical and horizontal frequency axes, due to the heating of the modes is clearly visible. b) Same as a) with the peaks in the upper right quadrant labeled. The frequency coordinates of the labeled peaks can be identified, see Eq. (3.116) below. The frequency coordinates of the remaining peaks can be inferred from these values.\footnote{The displayed figures were published in [L1]. Copyright (2015) by the American Physical Society.}

where $\bar{n}_{0,n}$ is the initial occupation number of the mode. Starting from the ground state or close to it, the heating observed in ion traps usually shows a linear increase in the mean phonon number $\dot{\bar{n}}(t) = \dot{\bar{n}} = \text{const}$ \cite{45}. Linearizing Eq. (3.113) in the limit $2\kappa_n t \ll 1$ we find

$$2\kappa_n = -\frac{\dot{\bar{n}}}{\bar{n}_{\text{eq},n} - \bar{n}_{0,n}} \tag{3.114}$$

Thus, specifying experimentally plausible rates $\dot{\bar{n}}_{zz}$ and $\dot{\bar{n}}_{\text{str}}$, all parameters for the model are fixed.

We set $\dot{\bar{n}}_{zz} = 0.2 \text{s}^{-1}$ and $\dot{\bar{n}}_{\text{str}} = 0.1 \text{s}^{-1}$, which seems to be a rather conservative estimate for macroscopic Paul traps \cite{95,119}, and truncate the motional Hilbert spaces at $n_{zz,\text{max}} = 9$ and $n_{\text{str, max}} = 6$ in our simulations, which leaves out a fraction of $10^{-4}$ of the initial populations.

The spectrum resulting for the pulse sequence Eq. (3.110), where the free evolution periods are described by Eq. (3.111), for the parameters in Table 3.7 is shown in Fig. 3.10 a).

A very bright peak at $(-\omega_{zz}, -\omega_{zz})$ has been subtracted from the spectrum to increase the contrast of the figure. Above and below the central peak there are two bright peaks. These originate from coherence transfer pathways starting in the ground state and are the most pronounced since the modes are initialized close to the ground state. We will identify the peaks appearing in
3.6. Two-dimensional spectroscopy with trapped ions – simulations of experimental implementations

Figure 3.11.: Coherence transfer pathways leading to a signal in the spectrum in Fig. 3.10. States are in the form $|n_{zz}, n_{str}\rangle$. The figure shows coherence transfer pathways that start in the motional ground state. The pathway in part a) leads to the central peak labeled a) in Fig. 3.10 b) with frequency coordinates $(-\omega_{zz}, -\omega_{zz})$. The pathway in part b) leads to the two bright peaks above and below the central peak with coordinates $(-\omega_{zz}, -\omega_{zz} \pm \sqrt{2}\Omega_T)$. The peak involving $+\sqrt{2}\Omega_T$ is labeled b) in Fig. 3.10 b).

the spectrum below. Before we get to this, we should pay attention to a different aspect of the spectrum. The peaks show homogeneous broadening, i.e. broadening along the two frequency axes, due to the heating. This broadening is due to fluctuations during each experimental run. As we have seen in Fig. 3.8, effective static dephasing due to couplings to other modes where the mode frequency is stable over one experimental run but differs from run to run leads to inhomogeneous broadening along the diagonal. Let us therefore note again that the form of the 2D spectrum provides us with information about the nature of the noise in the system.

Let us now proceed to identify the peaks in the spectrum. In Fig. 3.10 b) we show the spectrum of part a) with the peaks in the upper right quadrant marked from a)-f). Note that the remaining peaks in the spectrum can be related by reflections with respect to the origin. Hence, identifying the peaks a)-f) suffices to infer the coordinates of the remaining peaks.

We start by considering peaks a) and b), which are the most pronounced. Since both modes are initialized close to the ground state, we start by considering coherence transfer pathway starting in the ground state. It is then easy to see that the coherence transfer pathways displayed in Fig. 3.11 lead to the peaks at a) and b) as well as b)’s counterpart below the center of the spectrum. Both pathways have a time dependence $-\omega_{zz}$ during the first free evolution period $t_1$. Pathway a) also has a time dependence $-\omega_{zz}$ during the second free evolution period, while pathway b) has time dependences $-\omega_{zz} \pm \sqrt{2}\Omega_T$ during $t_3$. The two different time dependences during the second free evolution period of pathway b) can be understood considering Table 3.1. The state $|n_{zz} = 2, n_{str} = 0\rangle$ can be expanded in terms of the eigenstates of $H_{\text{res}}^{(3)}$ with eigenvalues $\pm \sqrt{2}\Omega_T$

$$|n_{zz} = 2, n_{str} = 0\rangle = \frac{1}{\sqrt{2}}(|\psi_+\sqrt{2}\Omega_T\rangle - |\psi_-\sqrt{2}\Omega_T\rangle), \quad (3.115)$$

where $|\psi_{\pm\sqrt{2}\Omega_T}\rangle = \frac{1}{\sqrt{2}}(0, 1) \pm |2, 0\rangle$. The states $|n_{zz} = 2, n_{str} = 0\rangle$ and $|n_{zz} = 0, n_{str} = 1\rangle$ are the states with the smallest occupation number where transfer of excitations between the modes happens. Thus, the peak b) and its counterpart directly witness transfer of excitations between the modes.

Peaks a) and b) were identified by considering the possible coherence transfer pathways that start in the motional ground state and end in a population. Thus, in order to identify the remaining peaks one has to consider initial motional states which contain excitations. Together with Tab. 3.1 one is then able to work out the pathways with the corresponding time dependences and can identify the physical origin of the peaks appearing in the spectrum. Below we list the peaks
a)−f) with their corresponding frequency coordinates

\[ \begin{align*}
\text{a)} & : (−\omega_{xx}, −\omega_{zz}) , \\
\text{b)} & : (−\omega_{xx}, −\omega_{zz} + \sqrt{2}\Omega_T) , \\
\text{c)} & : (−\omega_{xx} + (\sqrt{6} − \sqrt{2})\Omega_T, −\omega_{zz} + (\sqrt{6} − \sqrt{2})\Omega_T) , \\
\text{d)} & : (−\omega_{xx} + \sqrt{2}\Omega_T, −\omega_{zz} + \sqrt{2}\Omega_T) , \\
\text{e)} & : (−\omega_{xx} + (\sqrt{6} − \sqrt{2})\Omega_T, \omega_{xx} + (4 − \sqrt{6})\Omega_T) , \\
\text{f)} & : (−\omega_{xx} + (\sqrt{6} + \sqrt{2})\Omega_T, −\omega_{zz} + (\sqrt{6} + \sqrt{2})\Omega_T) .
\end{align*} \tag{3.116} \]

Accordingly, one can infer a few of the eigenvalues of \( H^{(3)}_{\text{res}} \) summarized in Tab. 3.1 from the spectrum.

### 3.6.3. Impact of laser phase fluctuations on phase cycling

The usefulness of the 2D spectra we presented in the previous sections depended critically on our ability to perform phase cycling. Phase cycling prevented the spectra from being overcrowded and thus incomprehensible. It also allowed us to identify the physical origin of the peaks appearing in the spectra by associating them to certain coherence transfer pathways. Phase cycling, in turn, depends on our ability to apply pulses with well-controlled phases. Actually, in the two previous sections we assumed that the phases of the laser-induced displacements can be controlled with arbitrary precision. This is not true in practice since a laser will exhibit phase fluctuations in the course of time. In this section we will analyze how harmful laser phase fluctuations are for the proposed protocol of 2D spectroscopy and calculate the loss in contrast we expect for the experiments we simulated.

We start our considerations with the simplest protocol for 2D spectroscopy consisting of a sequence of only two pulses. In this experiment the only relevant phase is the phase \( \phi_2 \) of the second laser pulse. According to Eq. (3.88) the recorded signal at the end of the sequence can be written as

\[ s(t_1, t_2, \phi_2) = \sum_p s_p(t_1, t_2) e^{ip\phi_2} . \tag{3.117} \]

In practice the signal is the mean of a series of measurements. In every experiment the phase \( \phi_2 \) fluctuates and at the time the second pulse is applied, it shows some small deviation from the desired value. In other words, the phase \( \phi_2 \) becomes a random variable and Eq. (3.117) becomes

\[ s(t_1, t_2, \phi_2) = \sum_p \langle s_p(t_1, t_2) e^{ip\phi_2} \rangle_{\text{st}} \tag{3.118} \]

where \( \langle \ldots \rangle_{\text{st}} \) denotes the stochastic average.

We now want to determine the effect of the laser phase fluctuations on the signal. To this end we need to specify a model for the laser phase fluctuations. We start by noting that the \( s_p(t_1, t_2) \) are independent of \( \phi_2 \). They are only determined by the initial populations, the moduli \( |\alpha_k| \) of the displacements and the action of the annihilation and creation operators. We further assume that the phase fluctuation are small on the experimental time scale and that accordingly we can write the phase of the \( j \)th experimental run as \( \phi_{2,j} = \phi_2 + \Delta\phi_{2,j} \), where \( |\Delta\phi_{2,j}| \ll 1 \). For the number of phase cycles we considered above the phases are of order 1 except for the first phase which is zero. Hence, \( |\Delta\phi_{2,j}| \ll 1 \) can be considered small. This choice is motivated by the results of [120] where laser phase drifts were analyzed. There, a drift of \( 2\pi \) in the laser phase was observed on a timescale \( \tau_d \approx 10s \), while the time scale for the experiments we consider here...
is of the order \( \tau_{\text{exp}} \approx 5 \text{ ms} \). Following these considerations we can write Eq. (3.118) as

\[
s(t_1, t_2, \phi_2) = \sum_p e^{i p \phi_2} s_p(t_1, t_2) \langle e^{i p \Delta \phi_2} \rangle_{\text{st}},
\]  

(3.119)

where \( \Delta \phi_2 \) is the random variable describing the fluctuations in the phase around the desired value \( \phi_2 \). We assume that \( \Delta \phi_2 \) can be modeled as a Wiener process [121]. A Wiener process \( X(t) \) is a Gaussian stochastic process that obeys the stochastic differential equation

\[
\frac{dX(t)}{dt} = \sqrt{c} N(t),
\]  

(3.120)

where \( c > 0 \) is called the diffusion constant and \( N(t) \) is the Gaussian white noise process. A stochastic process is a random variable whose distribution parametrically depends on the time \( t \). The Wiener process is a Gaussian random variable and therefore at every time \( t \) it represents a Gaussian random variable \( X(t) \) characterized by its first and second moments [121]

\[
\langle X(t) \rangle = X(t_0), \quad \text{Var}[X(t)] = c(t - t_0) \quad (t \geq t_0).
\]  

(3.121)

Note that in the following we will assume \( t_0 = 0 \). The covariance of the Wiener process for times \( s, t \) is given by [69]

\[
\text{Cov}[X(s), X(t)] = c \cdot \min(s, t).
\]  

(3.122)

In order to model the phase fluctuations as a Wiener process, we need to set the initial value and find an appropriate value for the diffusion constant. We set the initial value \( X(0) = 0 \) such that also the mean of the Wiener process is zero. For the diffusion constant we use the result of [120] that the laser phase drifts by \( 2\pi \) over an interval of \( \sim 10 \text{ s} \). We set the drift of \( 2\pi \) as the standard deviation \( \sigma \) of the process at \( t = 10 \text{ s} \). The diffusion constant is then given by solving \( \sigma = \sqrt{c t} \) for \( c \). With \( \sigma = 2\pi \) and \( t = 10 \text{ s} \) we find

\[
c = \left(4\pi^2 / 10\right) \text{s}^{-1}.
\]  

(3.123)

The value of \( \Delta \phi_2 \) is then given by the value of the stochastic process at the time the displacement pulse is applied. Here we have \( \Delta \phi_2 = X(t_1) \) because the pulse with phase \( \phi_2 \) is applied directly after the free evolution for \( t_1 \).

If we now want to evaluate Eq. (3.119) with \( \Delta \phi_2 \) being a Wiener process \( X(t) \), we have to take into account that the expansion of the exponential for order \( k \geq 1 \) involves the \( k \)th moment of \( X(t_1) \). Since \( X(t_1) \) is a Gaussian random variable with zero mean, all odd moments vanish. The even moments can be written in terms of the variance in Eq. (3.121)

\[
\text{Var}[X(t)] = ct.
\]  

(3.124)

In particular, using the well-known result that for a standard normal distributed \( \Delta \phi_2 \) we have \( \langle \Delta \phi_2^{2n} \rangle_{\text{st}} = (2n - 1)!! \text{Var}[\Delta \phi_2]^n \), where \( !! \) is the double factorial, we obtain

\[
\langle e^{i p \Delta \phi_2} \rangle_{\text{st}} = e^{-\frac{1}{2} p^2 \text{Var}[X(t)]}.
\]  

(3.125)

Using this result Eq. (3.119) becomes

\[
s(t_1, t_2, \phi_2) = \sum_p e^{i p \phi_2} s_p(t_1, t_2) e^{-\frac{1}{2} p^2 \text{Var}[X(t)]}.
\]  

(3.126)

Inserting the value for \( c \) from Eq. (3.123) and \( t = \tau_{\text{exp}} = 5 \text{ ms} \), we find \( \text{Var}[X(t)] \approx 0.02 \). Note that this is a conservative estimate since \( t_1 \) will be smaller than \( \tau_{\text{exp}} \). Hence, one can approximate
the exponential containing the phase fluctuations in Eq. (3.126) to second order for not too large values of $p$

\[
s(t_1, t_2, \phi_2) \approx \sum_p e^{ip\phi_2} s_p(t_1, t_2) \left(1 - \frac{1}{2} p^2 \text{Var}[\Delta \phi_2]\right) = \sum_p e^{ip\phi_2} s_p(t_1, t_2) \left(1 - \frac{1}{2} p^2 c t_1\right).
\]

(3.127)

For $c$ from Eq. (3.123) and $t_1 = 5$ ms we obtain corrections of about 1% for terms with $p = 1$ and 4% for $p = 2$. For these values there is basically no difference between the predictions of Eqs. (3.126) and (3.127). We will use this result for the four pulse sequence that we discuss in the following. In brief, we have seen that the phase fluctuations are not too harmful for the two pulse sequence.

After this instructive example, let us consider the four pulse sequence that we considered in the previous section. Generalizing Eq. (3.117) to the four pulse experiment with $t_2 = t_4 = 0$, we may write the signal as

\[
s(t_1, t_3, \phi) = \sum_p s_p(t_1, t_3) e^{ip\phi} = \sum_{p_2, p_3, p_4} s_{p_2, p_3, p_4}(t_1, t_3) e^{ip_2\phi_2} e^{ip_3\phi_3} e^{ip_4\phi_4}.
\]

(3.128)

Following our previous considerations we model the phases as stochastic variables $\phi_k \rightarrow \phi_k + \Delta \phi_k$, $k = 2, 3, 4$ with fluctuations $\Delta \phi_k$. Note, however, that the fluctuations of the different phases $\phi_k$ are independent. They are samples of the same stochastic process at different instances of time. With this substitution Eq. (3.128) becomes

\[
s(t_1, t_3, \phi) = \sum_p e^{ip\phi} s_p(t_1, t_3) \langle e^{ip_2\Delta \phi_2} e^{ip_3\Delta \phi_3} e^{ip_4\Delta \phi_4}\rangle_s,
\]

(3.129)

where we used that the $s_p(t_1, t_3)$ are independent of the fluctuations $\Delta \phi_k$. Motivated by the good agreement of Eqs. (3.126) and (3.127), we expand the exponentials in the above equation to second order, which is justified if the fluctuations are not too large. Using $\langle \Delta \phi_k \rangle_s = 0$ we obtain

\[
s(t_1, t_3, \phi) \approx \sum_p e^{ip\phi} s_p(t_1, t_3) \left[1 - \frac{3}{2} \sum_{k=2}^4 p_k^2 \Delta \phi_k^2 - \sum_{k=2}^4 \sum_{k'=k+1}^4 p_k p_{k'} \Delta \phi_k \Delta \phi_{k'}\right].
\]

(3.130)

Using $\Delta \phi_2 = \Delta \phi_3 = X(t_1)$ and $\Delta \phi_4 = X(t_1 + t_3)$ together with the covariance property Eq. (3.122) of the Wiener process finally yields

\[
s(t_1, t_3, \phi) \approx \sum_p e^{ip\phi} s_p(t_1, t_3) \left[1 - \frac{3}{2} c(p_2 + p_3 + p_4)^2 t_1 - \frac{1}{2} c p_{t_3}^2\right] = \sum_p e^{ip\phi} s_p(t_1, t_3) \left[1 - \Delta s_p(t_1, t_3)\right].
\]

(3.131)

With the help of Eq. (3.131) we can now estimate the loss in contrast for the four pulse experiment. For a conservative estimate we set $t_1 = t_3 = 2.5$ ms. We have summarized values for $\Delta s_p(t_1, t_3)$ for pathways whose contributions scale with $|\alpha|^4$ and $|\alpha|^6$ in Tab. 3.8. For the pathway with phase signature $\phi_2 - \phi_3 - \phi_4$, i.e., $(p_2, p_3, p_4) = (1, -1, -1)$, we estimate a loss of contrast of only 1%. For the other considered pathways the losses in contrast lie in the range 1-5%. Coherence transfer pathways with contributions $\propto |\alpha|^8$ or higher are very weak due to the smallness of $|\alpha|$ and are unlikely to contribute to the signal. Hence, laser phase fluctuations should not form an obstacle for the implementation of the proposed protocol for 2D spectroscopy with trapped ions.
Table 3.8: Expected loss in signal due to laser phase fluctuations. The table lists the loss in signal according to Eq. (3.131) for different pathways that can contribute to a 2D spectrum obtained with the pulse sequence in Eq. (3.107). This pulse sequence was used for the simulated 2D spectra in Figures 3.8 and 3.10.

<table>
<thead>
<tr>
<th>$p_2$</th>
<th>$p_3$</th>
<th>$p_4$</th>
<th>$\Delta \phi(t_1,t_3)$</th>
</tr>
</thead>
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</tr>
<tr>
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<td>-1</td>
<td>2.5%</td>
</tr>
<tr>
<td>1</td>
<td>-1</td>
<td>-2</td>
<td>4.0%</td>
</tr>
<tr>
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<td>-2</td>
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</tr>
<tr>
<td>-1</td>
<td>-1</td>
<td>-1</td>
<td>5.0%</td>
</tr>
</tbody>
</table>

3.7. Conclusions and outlook

In this chapter we have developed an extension of 2D spectroscopy for the study of nonlinear dynamics in crystals of trapped ions. We have illustrated the applicability of the method by simulating two experiments that can be realized in state-of-the-art setups of trapped ions. In particular, in the simulated experiments we showed how 2D spectroscopy can be used to detect signatures of the onset of a structural phase transition and resonant energy transfer between normal modes. In both cases 2D spectroscopy detects resonant interactions due to nonlinear corrections to the harmonic approximation of the Coulomb potential.

The scheme we developed has several advantages. In the case of small displacements, it does not produce a signal for purely harmonic time evolutions. This is a desirable feature because the harmonic contributions to the spectrum are typically much stronger than those due to the nonlinear parts of the time evolution.

The proposed protocol also allows us to separate signals that would appear superposed in a 1D spectroscopy experiment. It thus allows us to extract more information about the investigated system with 2D spectroscopy than would be available in a 1D spectroscopy experiment. Furthermore, the protocol allows us to characterize noise in the system. Effective static disorder, where frequencies are constant during one experimental run but fluctuate between runs, leads to inhomogeneous broadening, here broadening of the peaks in the diagonal direction of the spectrum. Fluctuations leading to dephasing or heating during each experimental run lead to homogeneous broadening, i.e. broadening of the peaks along the two frequency axes. A feature that is appealing from the experimental side is that the protocol does not need ground state cooling, which can become a real asset for the study of larger crystals.

A drawback of 2D spectroscopy is that it needs $N^2$ data points for a spectrum with $N$ points along each frequency axis, while a 1D spectrum with $N$ points only needs $N$ data points. The fact that the 2D experiment needs a factor $N$ more data points is not so problematic in the first place. However, for a trapped-ion experiment each data point is the mean of a series of measurements, typically of the order of 100, which makes it desirable to generate as few data points as possible. This problem can be remedied by employing techniques from the field of matrix completion. These facilitate a significant reduction in the number of measurements that need to be performed to obtain a 2D spectrum, ideally of the order of $N \log N$ instead of $N^2$ [122].

An interesting future direction for 2D spectroscopy with trapped ions would be to apply these concepts to the study of larger ion crystals. Here, the attention has not to be restricted to linear crystals, 2D spectroscopy could also be useful for the study of planar crystals in Penning traps [62,63] or 3D crystals in linear Paul traps [123]. It would be interesting to see whether and not unlikely that nonlinear effects play a role for the dynamics of these systems.

Another very interesting future direction of 2D spectroscopy with trapped ions would be the
investigation of interacting spin systems which are encoded in the internal degrees of freedom of the ions. The study of spin-spin interactions is a prominent field of 2D spectroscopy in NMR and it seems a natural extension to study these in systems of trapped ions. Nowadays, spin-spin interactions are routinely implemented both in linear and planar crystals incorporating tens \[23\] to hundreds \[62\] of ions, respectively. A topology of interactions in such systems is one example where 2D spectroscopy could be useful.

We should note, though, that NMR often relies on the fact that the nuclear spin frequencies are non-degenerate, because the chemical shifts of the spins depend on their location. In trapped ion experiments, the transition frequencies of all ions of the same species are in principle the same. One would thus have to introduce a separation of the spin frequencies “manually”. This could be done, for example, by introducing a magnetic field gradient along the trap axis in a linear trap. Interestingly, one scheme to induce spin-spin interactions between the internal degrees of freedom of trapped ions uses this configuration \[58, 59\]. The ions are placed in a static magnetic field gradient to generate interactions between the internal, i.e. spin, degrees of freedom of the ions. In this configuration all spins have a characteristic transition frequency and a COSY inspired protocol to study the interactions between ions could be applied. While this scheme seems to be especially well-suited for an application of 2D spectroscopy to study spin-spin interactions in crystals of trapped ions, the application of such a protocol should in general also be feasible for other spin-spin coupling schemes such as a Mølmer-Sørensen interaction \[86, 124\]. However, here one has to take care that resolving the individual spin frequencies does not hamper the interactions.

More generally, 2D spectroscopy could be applied to study the electronic or vibrational dynamics of many-body models that are simulated with trapped ions. This could also involve models where the spin and motional degrees of freedom are coupled. For example, if one is able to engineer models of interacting spins coupled to dissipative environments, as they occur in the dynamics of photosynthetic pigment protein complexes \[125\], trapped ions can be used to simulate 2D spectra of these systems. This is of great interest because these spectra provide information about the mechanisms underlying energy transport in these complexes and are very hard to simulate on classical computers \[126\].

In summary, we have seen that 2D spectroscopy is a versatile diagnostic tool that complements the available trapped-ion toolbox and has exciting perspectives for future applications with trapped ions.
4. Simulations of the dynamics of spin-boson models with trapped ions

The spin-boson model is a paradigmatic model of an open quantum system. It considers a single spin coupled to an environment made up of an infinite set of harmonic oscillators. The model is used in numerous contexts ranging from solid state physics [127, 128] and the analysis of chemical reactions [129] to the modeling of photosynthetic pigment protein complexes [125, 130]. Due to its wide range of applicability the model has been intensively studied and is still under consideration in research today.

Interestingly enough, the effect of thermal oscillator environments on the spin can be fully described by a single scalar function called the spectral density, also spectral function, of the environment [127, 128, 131]. However, despite its alleged simplicity no closed solution of the spin-boson model is known.

In some regions of parameter space analytical solutions for the dynamics and thermodynamical properties of the model are known. In most cases, however, numerical methods are needed to solve the dynamics or to extract thermodynamical properties of the model. Actually, a whole zoo of numerical methods that are used to tackle the spin-boson model exists. There are approaches based on the numerical renormalization group (NRG) [132], the time-dependent density matrix renormalization group (t-DMRG) [133, 134], the quasi-adiabatic path integral propagator (QUAPI) [135] or path integral Monte Carlo techniques [136, 137] to name a few examples. Another technique that has been applied to the spin-boson model are the so-called hierarchical equations of motion (HEOM) [138, 139].

All of the aforementioned techniques were successfully applied to describe the dynamics of spin-boson models. Yet, all numerical approaches are limited to certain regions of parameter space. Usually, the crucial feature that limits numerical methods is the environment’s memory time. In particular, all of the methods are challenged by environments with long memory times. A class of interesting examples where environments with long memory times are ubiquitous is the dynamics of photosynthetic pigment protein complexes [140]. A better understanding of the mechanisms of energy transport in these complexes is not only interesting in its own right but might also enable the development of new devices inspired by the design principles of Nature, such as highly efficient solar cells.

Photosynthetic pigment protein complexes are often modeled by spin-boson models consisting of several interacting spins coupled to dissipative environments [125]. Environments in molecules or photosynthetic pigment protein complexes are typically characterized by spectral densities that possess sharp peaks at certain frequencies over a broad background [140]. Because of the sharp peaks, these spectral densities are referred to as structured. Usually each of the sharp peaks corresponds a long-lived vibrational mode in the environment. If the system of interacting spins exhibits resonances with the long-lived modes, the effective memory time of the environment becomes long. In contrast to these structured environments, there are the unstructured environments such as the Ohmic as well as sub- and super-Ohmic environments which are characterized by shorter memory times [127].

Analytical approaches for these multi-spin-boson models with structured environments are rare while numerical methods are severely challenged by the complexity of the models. Yet, for the aforementioned reasons simulating the dynamics of photosynthetic pigment protein com-
plexes, or more generally multi-spin-boson models, is highly desirable. Since simulations on classical computers are exceedingly hard, these systems are an appealing candidate for quantum simulation. Building a quantum simulator with a high level of control and flexibility for such systems could be an instance where quantum simulation can outperform classical computation and achieve truly new insights.

Here, we are interested in finding out if and how one can build a quantum simulator of the dynamics of spin-boson models with trapped ions as a first step towards a quantum simulator of the dynamics of photosynthetic pigment protein complexes. The present chapter summarizes the results.

The basic idea for the construction of a trapped-ion quantum simulator for the dynamics of spin-boson models is that damping broadens the resonance of modes in frequency space to Lorentzians. Larger damping causes broader resonances and combining several damped modes one can theoretically construct arbitrary spectral densities. As we have seen in Chapter 2, all the needed ingredients are available with trapped ions. The internal degrees of freedom can be used to encode spins and the motional degrees of freedom can act as the modes of the environment. Cooling of the modes and spin-motion coupling are standard techniques and thus we see that trapped ions offer all the prerequisites to build such a simulator.

We should note that there is a previous proposal to build an ion trap simulator of spin-boson models [34]. In the cited work, the idea is to gather large ion crystals incorporating as many as 50-100 ions. The many normal modes of the crystal are then used to model the environment, however without the always-on cooling that we propose to use, yielding a discrete set of modes in the environment. In that way, one obtains mesoscopic environments for the spin. The large number of ions brings about difficulties in the state preparation for the modes, since they call for shallow axial potentials with relatively low frequencies. The level of control needed to implement the simulation proposed in [34] has not yet been demonstrated for such large crystals.

The protocol we describe below offers the advantage that one can simulate non-trivial physics already with a few ions. A remarkable feature of the protocol is that we obtain continuous spectral densities, that would usually need an infinite number of oscillators, due to the cooling. This is interesting because one usually considers continuous spectral densities in the condensed phase, where the environment is made up of a macroscopic number of particles. Thus, our protocol opens the perspectives for simulations of spin-boson models with spectral densities as they occur in the condensed phase. In particular, we will see that the protocol is especially well-suited for structured environments with long-lived modes, which is the type of spectral densities that appears for molecules and photosynthetic pigment protein complexes.

In order to illustrate our protocol, we will provide a detailed study of the simplest instance of a simulation of the spin-boson model with trapped ions, which is a spin coupled to a Lorentzian spectral density. This simulation only needs two ions. The Lorentzian spectral density is an instance of a structured environment and thus can yield long environmental memory times. This is the regime where the Markovian assumption used in the derivation of Markovian master equations breaks down. We will actually show that the dynamics of the models we study here are truly non-Markovian by computing two quantitative measures of non-Markovianity.

Note that the main results of this section were published in [L2]. Naturally, we follow the presentation of the material in [L2]. Finally, in order to construct a trapped-ion simulator of spin-boson models, we have to put together different pieces which are presented in the remainder of this chapter. First, we need to familiarize with the spin-boson model. Several analytical and numerical approaches to the problem are based on the path integral formulation of quantum mechanics. Therefore, after introducing the spin-boson model, we will briefly review the path integral formalism and then see how it can be applied to the study of open quantum systems. In this context, we will see how the
4.1. Spin-boson model and spectral densities

In the first section of this chapter we will briefly introduce the spin-boson model and the concept of the spectral density. After we have presented the path integral approach and the idea of an influence functional in the next sections, we will return to the spin-boson model and the concept of the spectral density. In particular, we will see how the statement that the spectral density alone determines the influence of the environment arises. In this section, however, we will only summarize some result from the literature.

4.1.1. Spin-boson model

The spin-boson model is an archetypical model of an open quantum system. The spin constitutes the open quantum system and its environment is modeled, as customary in open quantum systems, by an infinite set of harmonic oscillators. The Hamiltonian of the full system is usually taken to read [127]

$$H_{sb} = \frac{\varepsilon}{2} \sigma^z - \frac{\hbar \Delta}{2} \sigma^x + \frac{1}{2} \sum_{n=1}^{\infty} \left[ \frac{p_n^2}{m_n} + m_n \omega_n^2 x_n^2 - \gamma_n q_0 \sigma^z x_n \right]$$

where $\sigma^z = |e\rangle \langle e| - |g\rangle \langle g|$ and $\sigma^x = |e\rangle \langle g| + |g\rangle \langle e|$ are the usual Pauli matrices, $\varepsilon$ is the energy splitting of the eigenstates of $\sigma^z$ and $\hbar \Delta$ is their coupling also called tunnel element. The $p_n$ and $x_n$ denote the canonical momenta and coordinates of the environmental modes with associated frequency $\omega_n$. $q_0$ is some characteristic length scale and $\gamma_n$ describes the coupling of mode $n$ to the spin. Introducing the creation and annihilation operators of the harmonic oscillators of the environment according to

$$x_n = \sqrt{\frac{\hbar}{2m_n \omega_n}} (a_n + a_n^\dagger),$$

$$p_n = i \sqrt{\frac{\hbar m_n \omega_n}{2}} (a_n^\dagger - a_n),$$

so that $a_n$ and $a_n^\dagger$ denote the annihilation and creation operators of mode $n$, we can write the spin-mode coupling as

$$\hbar \lambda_n = \gamma_n q_0 \sqrt{\frac{\hbar}{2m_n \omega_n}}.$$

The spin-boson Hamiltonian can then be written as

$$H_{sb} = \frac{\varepsilon}{2} \sigma^z - \frac{\hbar \Delta}{2} \sigma^x - \frac{1}{2} \sum_{n=1}^{\infty} \hbar \lambda_n (a_n + a_n^\dagger) + \sum_{n=1}^{\infty} \hbar \omega_n a_n^\dagger a_n.$$
4. Simulations of the dynamics of spin-boson models with trapped ions

Figure 4.1.: Spin-boson model. Part a) of the figure, derived from [128], shows a double well system where the ground states of the left and right wells, $|L\rangle$ and $|R\rangle$, are separated by an energy $\varepsilon$. The first excited states in both wells are separated by some energy $\sim \hbar \omega_0$ from the respective ground states. The height of the potential barrier in the middle is $V_0$. Since we are dealing with a quantum system, tunneling with a rate $\Delta$ between the two wells is possible indicated by the dashed line. If $k_BT, \varepsilon, \hbar \Delta \ll \hbar \omega_0 \ll V_0$, we can reduce the system to a two level system [127]. The situation is depicted in part b). The figure also illustrates the coupling to an environment of harmonic oscillators where the spin couples to oscillator $\alpha$ with strength $\lambda_\alpha$.

where we omit the ground state energies of the oscillators.

Thus, the situation we are considering is a spin with an energy splitting $\varepsilon$ between the eigenstates of $\sigma^z |g\rangle$ and $|e\rangle$ that are coupled with strength $\hbar \Delta$. The spin interacts with an environment modeled by an infinite set of modes in the $\sigma^z$-basis. Note that it is common to denote the coupling between the spin states by $\Delta$ in the spin-boson literature [127, 128], which should not be confused with the detunings denoted $\Delta$ in the context of trapped ions in Chapter 2.

Let us try to motivate the choice of the spin-boson Hamiltonian in Eq. (4.5). Actually, the open system does not necessarily need to be a two-state system for the spin-boson Hamiltonian to be applicable [127, 128]. Consider, for example, a system that is described by a single generalized coordinate $q$ with an associated potential energy $V(q)$ which features two separate minima. The situation is depicted in Fig. 4.1 a).

We assume that the energy difference $\varepsilon$ between the two minima of the potential is small as compared to the energy gap to the first excited states which we assume to be of magnitude $\sim \hbar \omega_0 \ll V_0$ in both wells. Now, if the temperature is so low that $k_BT \ll \hbar \omega_0$, then the only relevant states will be the two ground states and the dynamics is confined to a two-dimensional space. Since we are dealing with a quantum mechanical system, we have to take into account the possibility that the system tunnels from one of the minima to the other. Thus, the system should be properly described by the spin part $(\varepsilon/2) \sigma^z - (\hbar \Delta/2) \sigma^x$ of $H_{sb}$ in Eq. (4.5).

In the condensed phase the system will be in contact with its environment which could be the phonons in a solid, for example. This situation is sketched in part b) of Fig. 4.1. It is common to model the environment as a bath of harmonic oscillators. If the spin interacts with a bath different from the phonons in a solid or the electromagnetic field, then the environment is in most cases actually not composed of harmonic oscillators. The choice to model environments as harmonic oscillators is due to the central limit theorem. For a very large number of fluctuating external forces, for example caused by the surrounding particles in the condensed phase, the joint effect of all influences will be a fluctuating force with Gaussian statistics on the system of interest [131]. This is also what the infinite set of harmonic oscillators causes on a system of interest with the advantage that it is much easier to treat theoretically. The free Hamiltonian of the oscillator bath
4.1. Spin-boson model and spectral densities

together with the coupling to the spin complete the Hamiltonian in Eq. (4.5). The coupling term is of course a specific choice but the common one in the spin-boson literature [127]. It can be interpreted as a dipole–local field coupling [127].

There is a variety of physical systems that can be described by a spin-boson model such as the tunneling of defects in crystalline solids, the tunneling of light particles in metals or chemical reactions involving electron transfer [127–129]. For the dynamics in biological systems the model in Eq. (4.5) is extended to include more spins that might also mutually interact and have a common or individual baths [125].

4.1.2. Spectral densities

As we will see later on, the quantity that determines the influence of the environment on the spin dynamics is the spectral density. It is usually denoted \( J(\omega) \) and there are different definitions used in the literature. We will adopt the convention that for the Hamiltonian in Eq. (4.5) the spectral density is given by

\[
J(\omega) = \pi \sum_n \lambda_n^2 \delta(\omega - \omega_n),
\]

(4.6)

where \( \delta \) is the Dirac \( \delta \)-function and can be interpreted as a density of states. Accordingly, the spectral density has units of frequency. Note that \( J(\omega) \) used here corresponds to \( G(\omega) \) in [127] with an additional factor of \( \pi \).

In solid state physics, the spectral densities are often taken to be of the form [128, 132]

\[
J(\omega) = 2\pi \alpha \omega^s f(\omega, \omega_c),
\]

(4.7)

where \( \alpha \) is a constant and \( \omega_c \) a cutoff frequency much larger than all other frequencies involved in the problem. \( f(\omega, \omega_c) \) is a cutoff function and can take different forms. Popular choices are the exponential cutoff \( f(\omega, \omega_c) = e^{-\omega / \omega_c} \) or the hard cutoff \( f(\omega, \omega_c) = \Theta(\omega - \omega_c) \). Depending on the exponent \( s \) the above spectral densities are called “Ohmic” \( (s = 1) \), “sub-Ohmic” \( (s < 1) \) and “super-Ohmic” \( (s > 1) \).

The dynamics of the spin under the influence of the environment depends on the shape of the spectral density and, as stated earlier, is not known in closed form for a general environment. The above cases, however, have been studied intensively and the behavior of the spin dynamics is known in many cases. Especially, the Ohmic case has gained a lot of attention and is amenable to analytic treatment in some parts of parameter space [127, 128]. Let us collect some known results for the dynamics of \( \langle \sigma_z(t) \rangle \) in environments characterized by different spectral densities.

For the Ohmic cases the constant in Eq. (4.7) becomes dimensionless and depending on its value, there are three different dynamical regimes at zero temperature [127,128]. Let us consider an unbiased spin, i.e. \( \epsilon = 0 \), in the scaling limit \( \omega_c \rightarrow \infty \), where all other quantities remain finite, for an initially localized state \( \langle \sigma_z(0) \rangle = \pm 1 \). For \( 0 < \alpha < 1/2 \), one obtains damped oscillations with a transition to incoherent (exponential) relaxation at \( \alpha = 1/2 \). At \( \alpha = 1 \), there is a further transition where the spin remains in its initial state called the localization transition. For finite temperatures there is a crossover from damped oscillations to overdamped oscillations, i.e. exponential relaxation, above a critical temperature [127]. In the subohmic case, NRG methods predict a localized phase at zero temperature where the critical value of \( \alpha \) depends on the tunnel element \( \Delta \) [132]. In the super-Ohmic case with \( s > 2 \), one obtains damped oscillations with a transition to overdamped oscillations above a critical temperature as in the Ohmic case [127]. Obviously, there are many more parameter regimes to be explored.

The collection of results above shows that the of behavior \( \langle \sigma_z(t) \rangle \) strongly depends on the properties of the environment or an interplay of the parameters of the spin and the environment.
4. Simulations of the dynamics of spin-boson models with trapped ions

This should clearly illustrate that it is hard to predict the dynamical behavior for a general environment and that a simulator for the dynamics of such a system would be very interesting. In fact, this is even more so when we consider spin-boson models including several coupled spins.

4.2. Path integrals in quantum mechanics

Before we proceed to consider the spin-boson model further, we take a little detour and briefly review the path integral formalism [141] which constitutes an alternative approach to quantum mechanics. Path integrals are widely used in the study of open quantum systems [69, 127, 142] but also find application in many other areas of physics such as quantum field theory [143, 144]. In the path integral formalism, the effect of harmonic oscillator environments on an open quantum system can be described using so-called influence functionals. Influence functionals form the basis for our further approach to the spin-boson model and we introduce them in Sections 4.3 and 4.4 below. However, before we get to this, we need to familiarize ourselves with the path integral formalism.

4.2.1. The quantum mechanical propagator as a path integral

In standard quantum mechanics, the quantity that determines the time evolution of a system is the Hamiltonian as it governs the Schrödinger equation. Classical mechanics, on the other hand, can be formulated equivalently using the Hamilton function of a system or the Lagrangian and the associated action of a system, which leads to the Euler-Lagrange equations. In the path integral formulation of quantum mechanics it is again the action that determines the dynamics of a system. In this section, we will briefly sketch an intuitive approach to the path integral formulation of quantum mechanics. We follow the treatment of [145]. References for mathematical details regarding the presentation can be found in [146].

Let us start by considering a classical particle moving from point \( x_0 \) at time \( t_0 \) to some point \( x_f \) at \( t_f \) in some potential \( V(x,t) \). The classical path \( x(t) \) taken by the particle is the one for which the classical action

\[
S[x(t)] = \int_{t_0}^{t_f} L(x,\dot{x},t)dt
\]

has an extremum [48]. Here, \( L(x,\dot{x},t) = T - V \) is the Lagrangian of the particle, where \( T \) is the kinetic energy. We note that the classical action is a functional, that is a rule that maps an entire function to a single number. To make this distinction clear we have written the argument of the action in angular brackets and shall do so for the remainder of this chapter.

In quantum mechanics we are many times interested in the question what is the amplitude that a system starting in some state \( |\psi_0\rangle \) at \( t_0 \) ends up in some state \( |\phi_f\rangle \) at some later time \( t_f \). Using the usual Hamiltonian formalism the transition amplitude reads

\[
\langle \phi_f | \psi(t_f) \rangle = \langle \phi_f | e^{-\frac{i}{\hbar}H(t_f-t_0)} |\psi_0\rangle ,
\]

where \( H \) is the Hamiltonian of the system under consideration and for simplicity we assumed it to be time independent. Let us consider the time evolution of the initial state

\[
|\psi(t_f)\rangle = e^{-\frac{i}{\hbar}H(t_f-t_0)} |\psi_0\rangle .
\]

Moving to the coordinate representation and using the completeness relation \( \int_{-\infty}^{\infty} dx_0 |x_0\rangle \langle x_0| = 1 \), we obtain

\[
\psi(x_f,t_f) = \int_{-\infty}^{\infty} dx_0 \langle x_f | e^{-\frac{i}{\hbar}H(t_f-t_0)} |x_0\rangle \psi(x_0,t_0) = \int_{-\infty}^{\infty} dx_0 K(x_f,t_f;x_0,t_0) \psi(x_0,t_0),
\]
where we have used $\psi(x_i, t_i) = \langle x_i | \psi(t_i) \rangle$. In the last equality we have introduced a general propagator $K(x_f, t_f; x_0, t_0)$ taking the system from position $x_0$ at time $t_0$ to $x_f$ at $t_f$. Comparing the second and third expressions we find the equality
\[ K(x_f, t_f; x_0, t_0) = \langle x_f | e^{-\frac{i}{\hbar}H(t_f-t_0)} | x_0 \rangle. \tag{4.12} \]

From the above equation, we immediately find the following product rule for the propagator [141]
\[ K(x_f, t_f; x_0, t_0) = \int_{0}^{\infty} dx_t K(x_f, t_f; x_t, t_t)K(x_t, t_t; x_0, t_0) \tag{4.13} \]
by dividing the time interval $[0, t_f]$ in two intervals $[0, t_t]$ and $[t_t, t_f]$ and inserting a completeness relation at time $t_t$. If we think of a particle traveling in one dimension, we can physically interpret this identity as the fact that the particle has to be at some point $x_i$ at any chosen time $t_i$.

We will now proceed to obtain a path integral representation for the propagator $K(x_f, t_f; x_0, t_0)$. Let us assume we are considering a system whose Hamiltonian is of the form
\[ \hat{H} = \hat{\rho} \hat{\pi} = \frac{\hat{\rho}^2}{2m} + V(\hat{x}), \tag{4.14} \]
where $\hat{\rho}$ is the particle's momentum operator and $m$ its mass. Note that we restrict our attention to the non-relativistic case and time-independent potentials. In the following we will make use of the identity [49]
\[ \langle x | p \rangle = \frac{1}{\sqrt{2\pi\hbar}} e^{\frac{ipx}{\hbar}} \tag{4.15} \]
for the product between a position eigenstate $\hat{x}|x\rangle = x|x\rangle$ and a momentum eigenstate $\hat{\rho}|p\rangle = p|p\rangle$.

Dividing the time interval $t_f-t_0 = N\Delta t$ into $N$ equal intervals of length $\Delta t$ and applying Eq. (4.13) $N-1$ times, we may write the propagator as
\[ K(x_f, t_f; x_0, t_0) = \int dx_{N-1} \ldots \int dx_1 K(x_N, t_N; x_{N-1}, t_{N-1}) \ldots K(x_1, t_1; x_0, t_0), \tag{4.16} \]
where $x_N = x_f$ and $t_N = t_f$ and $t_i = t_0 + i\Delta t$. Using the completeness relation of the momentum states, we may write
\[ K(x_{i+1}, t_i + \Delta t; x_i, t_i) = \langle x_{i+1} | e^{-\frac{i}{\hbar}H(\hat{\rho}, \hat{\pi})\Delta t} | x_i \rangle = \int_{-\infty}^{\infty} dp_i \langle x_{i+1} | p_i \rangle \langle p_i | e^{-\frac{i}{\hbar}H(\hat{\rho}, \hat{\pi})\Delta t} | x_i \rangle. \tag{4.17} \]

In order to proceed, we assume that $\Delta t$ is sufficiently small that it is justified to consider only the first order expansion of $e^{-\frac{i}{\hbar}H(\hat{\rho}, \hat{\pi})\Delta t}$. We obtain
\[
K(x_{i+1}, t_i + \Delta t; x_i, t_i) \approx \int dp_i \langle x_{i+1} | p_i \rangle \langle p_i | (\mathbb{I} - \frac{i}{\hbar}H(\hat{\rho}, \hat{\pi})\Delta t) | x_i \rangle \\
= \int dp_i \langle x_{i+1} | p_i \rangle \langle p_i | (\mathbb{I} - \frac{i}{\hbar}H(\rho, x_i)) \Delta t | x_i \rangle \\
\approx \int dp_i \langle x_{i+1} | p_i \rangle \langle p_i | e^{-\frac{i}{\hbar}H(\rho, x_i)\Delta t} | x_i \rangle \\
= \frac{1}{2\pi\hbar} \int dp_i e^{i\frac{p_i(x_{i+1} - x_i) - H(p_i, x_i)\Delta t}{\hbar}}.
\tag{4.18}
\]
where we used Eq. (4.15) twice in the last step. Substituting this result back into Eq. (4.16), we obtain
\[
K(x_f, t_f; x_0, t_0) \\
= \int dx_N \ldots \int dx_1 \int \frac{dp_{N-1}}{2\pi\hbar} \ldots \int \frac{dp_0}{2\pi\hbar} \exp \left\{ \frac{i}{\hbar} \Delta t \sum_{i=0}^{N-1} \left[ \frac{p_i(x_{i+1} - x_i)}{\Delta t} - H(p_i, x_i) \right] \right\}. \tag{4.19}
\]
Note that the appearing \( p_i \) and \( x_i \) should be understood as \( p(t_i) \) and \( x(t_i) \). Using the explicit form of the Hamiltonian in Eq. (4.14), we have \( H(p_i,x_i) = p_i^2/(2m) + V(x_i) \) and we can write the exponent in the above equation as

\[
\frac{p_i(x_{i+1} - x_i)}{\Delta t} - H(p_i, x_i) = -\frac{1}{2m} \left( p_i - m \frac{x_{i+1} - x_i}{\Delta t} \right)^2 + \frac{m}{2} \left( \frac{x_{i+1} - x_i}{\Delta t} \right)^2 - V(x_i). \tag{4.20}
\]

We simplify the above equation by performing the integrals over the momenta \( p_i \) by substitution and using the identity \( \int_{-\infty}^{\infty} dx e^{-c x^2} = \sqrt{\pi/c} \) for \( \text{Re}(c) > 0 \). To ensure convergence of the integrals one can add a factor \(-\varepsilon p_i^2; \varepsilon > 0\) to the exponent and let \( \varepsilon \to 0 \) in the end. After performing the integrals, we obtain

\[
K(x_f,t_f;x_0,t_0) = \left( \frac{m}{2 \pi i \hbar^2 \Delta t} \right)^{N/2} \int dx_{N-1} \cdots \int dx_1 \exp \left\{ \frac{i}{\hbar} \Delta t \sum_{i=0}^{N-1} \left[ \frac{m}{2} \left( \frac{x_{i+1} - x_i}{\Delta t} \right)^2 - V(x_i) \right] \right\}. \tag{4.21}
\]

Now we consider the limit \( N \to \infty \) and \( \Delta t \to 0 \) while keeping \( N \Delta t = t_f - t_0 \) fixed. The exponent of the propagator then tends to

\[
\frac{i}{\hbar} \int_{t_0}^{t_f} dt \left( \frac{m}{2} x^2(t) - V(x(t)) \right) = \frac{i}{\hbar} \int_{t_0}^{t_f} dt L(x, \dot{x}, t) = \frac{i}{\hbar} S[x(t)]. \tag{4.22}
\]

Thus, we obtain the action in the exponent.

In the propagator in Eq. (4.21), we divide the path into steps \( x_0, x_1, \ldots, x_N \) and integrate over all of the possible intermediate positions \( x_1, x_2, \ldots, x_{N-1} \) between the end points \( x_0 \) and \( x_N \). Accordingly, we are summing over all possible paths \( x(t) \) connecting \( x_0 \) and \( x_N \) where each of them contributes with a weight \( e^{\frac{i}{\hbar} S[x(t)]} \). The common short hand notation for the propagator in Eq. (4.21) is [141]

\[
K(x_f,t_f;x_0,t_0) = \int_{x(t_0)=x_0}^{x(t_f)=x_f} Dx \, e^{\frac{i}{\hbar} S[x(t)]}. \tag{4.23}
\]

Here \( \int_{x(t_0)=x_0}^{x(t_f)=x_f} Dx \) is the integral over all paths \( x(t) \) connecting \( x_0 \) and \( x_f \) and should be understood as a more concise expression for Eq. (4.21) in the limit \( N \to \infty \). The above object is what we understand as a path integral. The idea is that the physical path taken emerges from the interference of all possible paths where every path contributes with a phase given by its action [141].

There are some mathematical subtleties with the above formulation because \( \exp \left[ \frac{i}{\hbar} S[x(t)] \right] \) is not a proper measure in a mathematical sense and the existence of the limit in Eq. (4.21) is not guaranteed. This problem can be overcome by a so-called Wick rotation which leads to Euclidean path integrals where this problem can be alleviated [144]. Here, we shall not be concerned with these subtleties and content ourselves with the observation that path integrals work and are commonly used, e.g. for the description of dissipative quantum systems [127, 142]. The time evolution of an initial state \( |\psi_0\rangle \) with wave function \( \psi(x_0, t_0) \) in the path integral formalism is obtained by combining Eqs. (4.11) and (4.23)

\[
\psi(x_f, t_f) = \int_{-\infty}^{\infty} dx_0 \int_{x(t_0)=x_0}^{x(t_f)=x_f} Dx \, e^{\frac{i}{\hbar} S[x(t)]} \psi(x_0, t_0). \tag{4.24}
\]

This result can be generalized to the time evolution of a density matrix [127]

\[
\rho(x_f, x'_f, t_f) = \int_{-\infty}^{\infty} dx_0 dx'_0 K(x_f, t_f; x_0, t_0) \rho(x_0, x'_0, t_0) K^*(x'_f, t_f; x'_0, t_0) \\
= \int_{-\infty}^{\infty} dx dx' \int_{x(t_0)=x_0}^{x(t_f)=x_f} Dx \int_{x'(t_0)=x'_0}^{x'(t_f)=x'_f} Dx' \, e^{\frac{i}{\hbar} \left[ S[x(t)] - S[x'(t)] \right]} \rho(x_0, x'_0, t_0). \tag{4.25}
\]
It is understood that integrations over the spatial coordinates always extend over the whole real line and we will therefore omit the limits for them from now on. Also, we shall use the shorthand notation \( \int_{x(t_0)=x_0}^{x(t_f)=x_f} \mathcal{D}x = \int_{x_0}^{x_f} \mathcal{D}x \) if convenient.

### 4.2.2. Two-time correlation functions with path integrals

In this section we will establish how two-time correlation functions can be calculated within the path integral formalism. Let us consider a system with a general, again for simplicity time independent, Hamiltonian \( \hat{H} \). The treatment follows those in [145, 147].

In order to derive expressions for the two-time correlation functions, it will turn out to be useful to work in the Heisenberg picture where \( \hat{x}(t) = e^{i\hat{H}t} \hat{x} e^{-i\hat{H}t} \) with \( \hat{x} \) the Schrödinger picture position operator. We will use operators hat in this section for clarity. Let us also define the eigenstates \( |x, t \rangle \) of the Heisenberg picture operators

\[
|x, t \rangle = e^{i\hat{H}t} |x \rangle \quad \text{such that} \quad \hat{x}(t) |x, t \rangle = x |x, t \rangle. \tag{4.26}
\]

The Heisenberg operator eigenstates form a basis for every \( t \), i.e. \( \int \langle x, t | x, t \rangle \, dx = 1 \). Note that we can also write the propagator of Eqs. (4.12) and (4.23) in terms of the Heisenberg operator eigenstates as

\[
K(x_f, t_f; x_0, t_0) = \int_{x(t_0)=x_0}^{x(t_f)=x_f} \mathcal{D}x e^{iS[x]} = \langle x_f, t_f | x_0, t_0 \rangle. \tag{4.27}
\]

With these results at hand, our goal is to obtain an expression for the two-time correlation function \( \langle \hat{x}(t_2) \hat{x}(t_1) \rangle_\beta \), where we assume \( t_2 > t_1 \) and \( (\ldots)_\beta \) is the expectation value with respect to some thermal state at inverse temperature \( \beta = 1/(k_B T) \) where \( k_B \) is the Boltzmann constant. Expanding the thermal density operator in the Heisenberg eigenstates \( \rho_\beta = \int dx_0 dx_0' \rho(x_0, x_0', t_0) |x_0, t_0 \rangle \langle x_0', t_0| \), we have

\[
\langle \hat{x}(t_2) \hat{x}(t_1) \rangle_\beta = \text{Tr} \left[ \int dx_0 dx_0' \rho(x_0, x_0', t_0) \hat{x}(t_2) \hat{x}(t_1) |x_0, t_0 \rangle \langle x_0', t_0| \right]. \tag{4.28}
\]

Using the completeness of the Heisenberg eigenstates we obtain

\[
\langle \hat{x}(t_2) \hat{x}(t_1) \rangle_\beta = \text{Tr} \left[ \int dx_0 dx_0' dx_1 dx_2 \rho(x_0, x_0', t_0) \hat{x}(t_2) |x_2, t_2 \rangle \langle x_2, t_2| \hat{x}(t_1) |x_1, t_1 \rangle \langle x_1, t_1| x_0, t_0 \rangle \langle x_0', t_0| \right], \tag{4.29}
\]

where we have used Eq. (4.26) from the second to the third line. Performing the trace yields

\[
\langle \hat{x}(t_2) \hat{x}(t_1) \rangle_\beta = \int dx_f \ldots dx_2 \rho(x_0, x_0', t_0) x_1 x_2 \langle x_f, t_f | x_2, t_2 \rangle \langle x_2, t_2| x_1, t_1 \rangle \langle x_1, t_1| x_0, t_0 \rangle \langle x_0', t_0| x_f, t_f \rangle. \tag{4.30}
\]

Recalling Eq. (4.27) we can write the scalar products \( \langle x_2, t_2 | x_1, t_1 \rangle = \int_{x(t_1)=x_1}^{x(t_2)=x_2} \mathcal{D}x e^{iS[x]} \) in terms of propagators. The two time correlation function then becomes

\[
\langle \hat{x}(t_2) \hat{x}(t_1) \rangle_\beta = \int dx_f \ldots dx_2 \rho(x_0, x_0', t_0) \int dx_1 dx_2 x_1 x_2 \int_{x(t_0)=x_0}^{x(t_2)=x_2} \mathcal{D}x e^{iS[x]} \int_{x(t_0)=x_0}^{x(t_1)=x_1} \mathcal{D}x e^{iS[x]}, \tag{4.31}
\]

\[
\int_{x(t_2)=x_2}^{x(t_f)=x_f} \mathcal{D}x e^{iS[x]} \int_{x(t_1)=x_1}^{x(t_2)=x_2} \mathcal{D}x e^{iS[x]} \int_{x(t_0)=x_0}^{x(t_1)=x_1} \mathcal{D}x e^{iS[x]} \int_{x'(t_0)=x'_0}^{x(t_f)=x_f} \mathcal{D}x' e^{-iS[x']}. \]
Thus, we have a succession of three path integrals taking the coordinate $x(t)$ from $x_0$ at time $t_0$ to $x_f$ at time $t_f$ passing through the points $x_1$ at time $t_1$ and $x_2$ at time $t_2$. Since the coordinates $x_1$ and $x_2$ are arbitrary and we integrate over $x_1$ and $x_2$, the path integral includes all paths connecting $x_0$ at time $t_0$ to $x_f$ at time $t_f$ [147]. Hence, we may write a single path integral for $x(t)$ with initial and final points $x_0$ and $x_f$, respectively. We obtain the factors $x_1$ and $x_2$ above, i.e. the expectation values of $\hat{x}(t)$ at these times, by taking the factors of $x_1(t_1)$ and $x_2(t_2)$ into the path integral [145, 147]

\[
\int dx_1 dx_2 x_1 x_2 \int_{x(t_1)}^{x(t_2)} dx \mathcal{D}x e^{\frac{i}{\hbar} S[x] + \int x(t_1) = x_1 \mathcal{D}x e^{\frac{i}{\hbar} S[x]}} \int_{x(t_0)}^{x(t_2)} dx \mathcal{D}x e^{\frac{i}{\hbar} S[x]}
\]

\[
= \int_{x(0)=x_0}^{x(t_f)=x_f} \mathcal{D}xx(t_1)x(t_2) e^{\frac{i}{\hbar} S[x]}. \quad (4.32)
\]

With this we obtain the final result

\[
\langle \hat{x}(t_2),\hat{x}(t_1) \rangle_B = \int dx_f dx_0 dx'_0 \int_{x(t_0)=x_0}^{x(t_f)=x_f} \mathcal{D}xx(t_1)x(t_2) e^{\frac{i}{\hbar} S[x]} \int_{x(t_0)=0}^{x(t_f)=x_f} \mathcal{D}x'e^{-\frac{i}{\hbar} S[x']} \rho(x_0,x'_0,t_0).
\]

(4.33)

Following the same reasoning as above and using the invariance of the trace under cyclic permutation, we obtain the following expressions for the commutator and anticommutator of $\hat{x}$ at different times $t_2 > t_1$ which will be of use to us later

\[
\langle \{\hat{x}(t_2),\hat{x}(t_1)\} \rangle_B = \int dx_f ... \mathcal{D}x'e^{\frac{i}{\hbar}(S[x]-S[x'])} \rho(x_0,x'_0) \left[ x(t_2)x(t_1) - x'(t_2)x'(t_1) \right], \quad (4.34)
\]

\[
\langle \{\hat{x}(t_2),\hat{x}(t_1)\} \rangle_B = \int dx_f ... \mathcal{D}x'e^{\frac{i}{\hbar}(S[x]-S[x'])} \rho(x_0,x'_0) \left[ x(t_2)x(t_1) - x'(t_2)x(t_1) \right], \quad (4.35)
\]

and

\[
\langle \{\hat{x}(t_2),\hat{x}(t_1)\} \rangle_B = \int dx_f ... \mathcal{D}x'e^{\frac{i}{\hbar}(S[x]-S[x'])} \rho(x_0,x'_0) \left[ x(t_2)x(t_1) + x'(t_2)x'(t_1) \right], \quad (4.36)
\]

\[
\langle \{\hat{x}(t_2),\hat{x}(t_1)\} \rangle_B = \int dx_f ... \mathcal{D}x'e^{\frac{i}{\hbar}(S[x]-S[x'])} \rho(x_0,x'_0) \left[ x(t_2)x(t_1) + x'(t_2)x(t_1) \right]. \quad (4.37)
\]

In the above expressions we have used that $\rho(x_0,x'_0,t_0) = \rho(x_0,x'_0)$ for a thermal state with respect to $H$. Let us finally note that by performing a calculation completely analogous to the one presented above we obtain the following expression for the expectation value of $x$

\[
\langle \hat{x}(t_1) \rangle_B = \int dx_f ... \mathcal{D}x'e^{\frac{i}{\hbar}(S[x]-S[x'])} x(t_1) \rho(x_0,x'_0), \quad (4.38)
\]

\[
\langle \hat{x}(t_1) \rangle_B = \int dx_f ... \mathcal{D}x'e^{\frac{i}{\hbar}(S[x]-S[x'])} x'(t_1) \rho(x_0,x'_0). \quad (4.39)
\]

### 4.3. Path integrals for open quantum systems – Influence functionals

In this section we will introduce the concept of influence functionals characterizing the influence of an environment on the dynamics of a system under consideration. We will follow the treatment of [131], which relies on the assumption of an initial product state between system and environment. We note that extensions of the method in order to facilitate the treatment of arbitrary initial conditions exist [127, 142].

Let us consider a system composed of two subsystems which we will designate as principal system and environment. We denote the coordinate of the principal system by $q$ while we call
Accordingly, the state of the principal system \( \rho \) reads
\[
\rho(q_f, x_f, q'_f, x'_f, t_f) = \int dq_0 dq_0' dq_0 dq_0' dx_0 \int_{q_0}^{q_f} dq \int_{q_0}^{q_f'} dq' \int_{x_0}^{x_f} dx \int_{x_0'}^{x_f'} dx' \rho(q_0, q_0', x_0, x_0', t_0).
\]
(4.41)

Here, \( S[q, x] \) denotes the action of the full system as specified previously and therefore we have to sum over all possible paths of both \( q \) and \( x \). However, we are only interested in the principal system alone and therefore want to trace out the environment. In the representation we are using, the trace operation becomes
\[
\text{Tr}_E \rho = \int dx_f dx'_f \delta(x_f - x'_f) \rho(q_f, x_f, q'_f, x'_f, t_f) = \int dx_f \rho(q_f, x_f, q'_f, x_f, t_f).
\]
(4.42)

Accordingly, the state of the principal system \( \rho_S \) at time \( t_f \) is given by
\[
\rho_S(q_f, q'_f, t_f) = \int dx_f dq_0 dq_0 dx_0 \int_{q_0}^{q_f} dq \int_{q_0}^{q_f'} dq' \int_{x_0}^{x_f} dx \int_{x_0'}^{x_f'} dx' e^{i[S(q, x) - S(q', x')]} \rho(q_0, x_0, q_0', x_0', t_0).
\]
(4.43)

We will now assume that system and environment are initially in some product state
\[
\rho(q_0, x_0, q_0', x_0', t_0) = \rho_S(q_0, q_0', t_0) \rho_E(x_0, x_0', t_0).
\]
(4.44)

Inserting this initial state and using the decomposition of the action in Eq. (4.40), we can write Eq. (4.43) as
\[
\rho_S(q_f, q'_f, t_f) = \int dq_0 dq_0 dx_0 \int_{q_0}^{q_f} dq \int_{q_0}^{q_f'} dq' e^{i[S_0(q) - S_0(q')]} \rho_S(q_0, q_0', t_0) \left[ \int dx_f dx_0 dx_0' \int_{x_0}^{x_f} dx \int_{x_0'}^{x_f'} dx' e^{i[S(q, x) + S_E[x] - S(q', x') - S_E[x']]} \rho_E(x_0, x_0', t_0) \right].
\]
(4.45)

Here, we have collected all terms involving functions other than \( q \) and \( q' \) in the brackets. The resulting term contains all the dynamics of the environment. Since the order of integration is in principle arbitrary, we can perform the integrals over the \( x \) variables first and the resulting expression will only depend on \( q \) and \( q' \). This expression is called the influence functional \( F[q, q'] \) of the environment and captures all effects of the environment on the dynamics of the principal system [131]. Here, the influence functional reads
\[
F[q, q'] = \int dx_f dx_0 dx_0' \int_{x_0}^{x_f} dx \int_{x_0'}^{x_f'} dx' e^{i[S(q, x) + S_E[x] - S(q', x') - S_E[x']]} \rho_E(x_0, x_0', t_0).
\]
(4.46)
4. Simulations of the dynamics of spin-boson models with trapped ions

With the influence functional, the time evolution of the principal system’s density matrix can be written more concisely as

$$\rho_S(q_f, q'_f, t_f) = \int dq'_0 dq_0 \int d\delta q' \int d\delta q \rho(q_0, q'_0, t_0) \cdot F[q, q'] \cdot e^{i\tilde{S}_0[q] - \tilde{S}_0[q']}.$$ (4.47)

Accordingly, two environments that have the same influence functional produce the same influence on the dynamics of the principal system, even if they are physically different. The most important properties of influence functionals are discussed in [131]. In the following, we will make use of the following property of influence functionals:

If different, statistically and dynamically independent systems $x_k$ interact with the principal system each with influence functional $F_k[q, q']$, then the combined influence functional is given by

$$F[q, q'] = \Pi_k F_k[q, q'].$$ (4.48)

Accordingly, the influence functional in Eq. (4.46) is easily generalized to environments consisting of many, possibly infinitely many, constituents.

4.4. Influence functionals for harmonic oscillator environments

The path integral formalism is appealing for the description of open quantum systems because the influence functional for an environment of independent, lossless harmonic oscillators can be solved analytically. Note that this, however, does not mean that the dynamics of the principal system can be solved analytically with the same ease. In this section, we introduce the spectral density for oscillator environments and how it relates to the two-time correlation functions of the oscillators comprising the environment. Before we consider an oscillator bath with infinitely many oscillators, we start by considering a principal system coupled to a single lossless harmonic oscillator.

4.4.1. Influence functionals and spectral density for oscillator environments

We consider a principal system described by a generalized coordinate $q$ interacting with an environment consisting of a single harmonic oscillator with mass $m_n$, coordinate $x_n$ and frequency $\omega_n$. Let us suppose that the interaction with the environment is described by the interaction potential

$$V(q, x_n) = -\gamma_n q x_n,$$ (4.49)

where $\gamma_n$ is a coupling constant. $\gamma_n$ could, in principle, be time dependent but we take it to be time independent here. The action of the whole system takes the form in Eq. (4.40) where

$$S_E[x_n] = \int_{t_0}^{t_f} dt \left[ \frac{1}{2} m_n \dot{x}_n^2 - \frac{1}{2} m_n \omega_n^2 x_n^2 \right],$$ (4.50)

$$S_I[q, x_n] = \int_{t_0}^{t_f} dt \gamma_n q x_n.$$ (4.51)

Combining the two contributions to the action we obtain the Lagrangian

$$L = \frac{1}{2} m_n \dot{x}_n^2 - \frac{1}{2} m_n \omega_n^2 x_n^2 + \gamma_n q x_n,$$ (4.52)

for the oscillator $x_n$. This is the Lagrangian of a driven harmonic oscillator. The path integral propagator for the driven harmonic oscillator in a thermal state can be solved analytically [127, 146]. Thus, for an initial product state of principal system and oscillator, the influence
where the function \( \tilde{L}(t) \) reads

\[
\tilde{L}(t) = \sum_n \frac{\gamma_n^2}{2 m_n \omega_n} \left[ \coth \left( \frac{\hbar \beta \omega_n}{2} \right) \cos(\omega_n t) - i \sin(\omega_n t) \right].
\]

(4.56)

\( \tilde{L}(t) \) is sometimes also called the correlation function of the environment and we shall see in the following section why this is so. It is interesting to note that the influence of the whole, possibly infinite, environment is contained in a single scalar function \( \tilde{L}(t) \).

Let us now define the spectral density

\[
\tilde{J}(\omega) = \pi \sum_n \frac{\gamma_n^2}{2 m_n \omega_n} \delta(\omega - \omega_n).
\]

(4.57)

The function \( \tilde{L}(t) \) can now be written in terms of the spectral density

\[
\tilde{L}(t) = \frac{1}{\pi} \int_0^\infty d\omega \tilde{J}(\omega) \left[ \coth \left( \frac{\hbar \beta \omega}{2} \right) \cos(\omega t) - i \sin(\omega t) \right].
\]

(4.58)

In this sense, we can state that the influence of a thermal oscillator environment on the principal system is determined by the spectral density of the environment alone.

As we stated previously in Section 2.4.2, loss can only truly be modeled by an environment which contains a continuum of oscillators. Actually, one can show that if the principal system is also a harmonic oscillator, there will be revivals of the state of the full system after a finite recurrence time for a finite number of oscillators in the environment [146]. For a sufficiently large but still finite number of oscillators this so-called Poincaré recurrence time becomes practically infinite [127, 146].

In practice, therefore, one usually considers that the frequencies in Eq. (4.57) are so closely spaced that the spectral density becomes a continuous function of \( \omega \) [127, 128], which we have already anticipated in Eq. (4.7) above. If we consider a system in the condensed phase where the open system is surrounded by a practically infinite number of particles (more than \( 10^{20} \)), this assumption seems fair. For a continuous environment the function \( \tilde{L}(t) \) still has the form Eq. (4.58) with the only difference that the spectral density is now a continuous function of \( \omega \).
Let us finally note that in this section we have considered a principal system described by a generalized coordinate \( q \). If \( q \) is really an extended coordinate, the coupling constant \( \gamma_n \) has unit of energy over length squared and accordingly the unit of the spectral density \( \tilde{J}(\omega) \) is frequency over distance squared. If we now consider the principal system to be a spin such that \( q(t) = \pm \frac{1}{2} \), we substitute \( q(t) \) by \( \frac{q_0}{2} \sigma_z \) to obtain the interaction between spin and modes, see the Hamiltonian in Eq. (4.5). This leads to an additional factor \( q_0^2 \) in \( \tilde{J}(\omega) \). For the spin-boson model we also absorb the factor \( \frac{1}{\hbar} \) in the influence functional, Eq. (4.55), into the spectral density such that using Eq. (4.4) the spectral density takes the form

\[
J(\omega) = \pi \sum_n \lambda_n^2 \delta(\omega - \omega_n), \tag{4.59}
\]

which we already introduced in Eq. (4.6) above. The spectral density \( J(\omega) \) has units of frequency. Finally, the influence functional, Eq. (4.55), for the spin-boson model takes the form

\[
F[q, q'] = \exp \left\{ - \int_{t_0}^{t_f} dt_1 \int_{t_0}^{t_1} dt_2 [q(t_1) - q'(t_1)] [q(t_2) L(t_1 - t_2) - q'(t_2) L^*(t_1 - t_2)] \right\}, \tag{4.60}
\]

where the function \( \tilde{L}(t) \) is substituted by

\[
L(t) = \frac{q_0^2}{\hbar} \tilde{L}(t) \tag{4.61}
\]
determined by \( J(\omega) \)

\[
L(t) = \frac{1}{\pi} \int_0^{\infty} d\omega J(\omega) \left[ \coth \left( \frac{\hbar \beta \omega}{2} \right) \cos(\omega t) + i \sin(\omega t) \right]. \tag{4.62}
\]

### 4.4.2. The spectral density, two-time correlation functions and the fluctuation-dissipation theorem

In the derivation of master equations in the Born-Markov approximation in Section 2.4.1 we have seen that the Lindblad rates are essentially given by a Fourier transform of the two-time correlation functions of environmental operators obtained under the free evolution of the environment.

In the path integral approach the environmental correlation function \( \tilde{L}(t) \) from Eq. (4.56) determines the influence of the oscillator environment on the principal system. For the interaction in Eq. (4.49) it is determined by the coordinate correlation function of the environmental oscillators. In particular, for this interaction we have [127]

\[
\tilde{L}(t) = \sum_n \frac{\gamma_n^2}{\hbar} \langle x_n(t)x_n(0) \rangle_B. \tag{4.63}
\]

In this section we want to show how one can arrive at this result starting from the general expression for the influence functional of an environment that is a linear system in [131]. Once we have done this, in the second part of this section we try and elucidate that the validity of the fluctuation-dissipation theorem enables us to write \( \tilde{L}(t) \) as in Eq. (4.58), i.e. as a function of the spectral density alone.
The spectral density and two-time correlation functions

In case the external influence on the principal system is due to a linear system or a definite or fluctuating classical force, the most general form of the influence functional is given by [131]

\[
F[q, q'] = \exp \left\{ \int_0^t dt C_1(t) [q(t) - q'(t)] - \int_0^t dt \int_0^t ds A_1(t - s) [q(t) - q'(t)] [q(s) - q'(s)] - i \int_0^t dt \int_0^t ds B_1(t - s) [q(t) - q'(t)] [q(s) + q'(s)] \right\}.
\]

(4.64)

Accordingly, the influence functional for these types of environments is generally determined by three functions \(A_1(t), B_1(t)\) and \(C_1(t)\). Note that the harmonic oscillator is an instance of a linear system. Comparing with Eq. (4.55), we find that for oscillator environments \(A_1(t) = \text{Re}[\mathcal{L}(t)/\hbar]\) and \(B_1(t) = \text{Im}[\mathcal{L}(t)/\hbar]\) which are determined by the spectral density, while \(C_1(t) = 0\).

Let us now define the coordinates \(q_+ \equiv q(t) + q'(t)\) and \(q_- \equiv q(t) - q'(t)\) and write the general influence functional in Eq. (4.64) in terms of the new coordinates

\[
F[q_+, q_-] = \exp \left\{ \int_0^t dt C_1(t) q_- (t) - \int_0^t dt \int_0^t ds A_1(t - s) q_- (t) q_- (s) - i \int_0^t dt \int_0^t ds B_1(t - s) q_- (t) q_+ (s) \right\}.
\]

(4.65)

In order to obtain the functions \(A_1(t), B_1(t)\) and \(C_1(t)\) we now take functional derivatives of the above expression with respect to \(q_\pm\). One needs to find the expressions for

\[
\frac{\delta F[q_+, q_-]}{\delta q_- (t_1)}, \frac{\delta F[q_+, q_-]}{\delta q_- (t_2)}, \frac{\delta F[q_+, q_-]}{\delta q_+ (t_1)}, \frac{\delta F[q_+, q_-]}{\delta q_+ (t_2)}.
\]

(4.66)

The resulting general expressions are quite involved but we find the following identities

\[
\left. \frac{\delta F[q_+, q_-]}{\delta q_- (t_1)} \right|_{q_- = q_+ = 0} = iC_1(t_1),
\]

(4.67)

\[
\left. \frac{\delta^2 F[q_+, q_-]}{\delta q_- (t_2) \delta q_- (t_1)} \right|_{q_- = q_+ = 0} = -A_1(t_2 - t_1) - C_1(t_2) C_1(t_1),
\]

(4.68)

\[
\left. \frac{\delta^2 F[q_+, q_-]}{\delta q_- (t_2) \delta q_+ (t_1)} \right|_{q_- = q_+ = 0} = -i\Theta(t_2 - t_1) B_1(t_2 - t_1).
\]

(4.69)

Thus, we have found a recipe to obtain the functions \(A_1(t), B_1(t)\) and \(C_1(t)\) in terms of functional derivatives of the influence functional.

The simplest case we can consider now is, again, an environment made up of a single harmonic oscillator. Inserting the action for an environment of a single harmonic oscillator from Eqs. (4.50), (4.51) into the general form of the influence functional in Eq. (4.46) yields the influence functional

\[
F[q, q'] = \int dx dx' dx_0 \int_{x_0}^{x_f} \mathcal{D}x_n \int_{x_n}^{x'} \mathcal{D}x'_n e^{i[S_i[q, x_n] + S_0[x_n] - S_i[q', x'_n] - S_0(x_n)]} \rho_E(x_0, x_0')
\]

(4.70)

with \(S_0[x_n] = \int_0^t dt \left( \frac{1}{2} m_n x_n^2 - \frac{1}{2} m_n \omega_n^2 x_n^2 \right)\) and \(S_i[q, x_n] = \int_0^t dt \gamma_0 q x_n\). Note that we assume that
the environment is in a thermal equilibrium state here. Defining \( x_\pm = x_n \pm x'_n \) we find

\[
\frac{\delta F[q_+, q_-]}{\delta q_-(t_1)} \bigg|_{q_- = q_+ = 0} = \frac{i\gamma}{\hbar} \int dx_f \ldots \langle x'_n e^{\frac{i}{\hbar}(S_0| x_0 \rangle - S_0| x'_0 \rangle) x_+ (t_1) \rangle \rho_E(x_0, x'_0),
\]

(4.71)

\[
\frac{\delta^2 F[q_+, q_-]}{\delta q_-(t_2) \delta q_-(t_1)} \bigg|_{q_- = q_+ = 0} = -\frac{\gamma^2}{2\hbar^2} \int dx_f \ldots \langle x'_n e^{\frac{i}{\hbar}(S_0| x_0 \rangle - S_0| x'_0 \rangle) x_+ (t_1) x_+ (t_2) \rangle \rho_E(x_0, x'_0),
\]

(4.72)

\[
\frac{\delta^2 F[q_+, q_-]}{\delta q_-(t_2) \delta q_+(t_1)} \bigg|_{q_- = q_+ = 0} = -\frac{\gamma^2}{2\hbar^2} \int dx_f \ldots \langle x'_n e^{\frac{i}{\hbar}(S_0| x_0 \rangle - S_0| x'_0 \rangle) x_- (t_1) x_+ (t_2) \rangle \rho_E(x_0, x'_0),
\]

(4.73)

where it is understood that \( \int dx_f \ldots \) contains the same integrals as the right-hand side of Eq. (4.70). Writing the \( x_\pm \) in terms of \( x_n \) and \( x'_n \) and comparing the resulting expressions with Eqs. (4.34)-(4.39), we find

\[
\frac{\delta F[q_+, q_-]}{\delta q_-(t_1)} \bigg|_{q_- = q_+ = 0} = i\gamma \langle x_n(t_1) \rangle \beta,
\]

(4.74)

\[
\frac{\delta^2 F[q_+, q_-]}{\delta q_-(t_2) \delta q_-(t_1)} \bigg|_{q_- = q_+ = 0} = -\frac{\gamma^2}{2\hbar^2} \langle \{ x_n(t_2), x_n(t_1) \} \rangle \beta,
\]

(4.75)

\[
\frac{\delta^2 F[q_+, q_-]}{\delta q_-(t_2) \delta q_+(t_1)} \bigg|_{q_- = q_+ = 0} = -\frac{\gamma^2}{2\hbar^2} \{ x_n(t_2), x_n(t_1) \} \beta.
\]

(4.76)

Accordingly, the quantities determining the influence of the environment on the principal system are the expectation value of \( x_n \) and its two-time correlation function with respect to the evolution of the environment alone. Comparing the expressions (4.67)-(4.69) and (4.74)-(4.76) we obtain the following expressions for the functions \( A_1(t_1), B_1(t_1) \) and \( C_1(t) \)

\[
C_1(t_1) = \frac{\gamma}{\hbar} \langle x_n(t_1) \rangle \beta,
\]

(4.77)

\[
A_1(|t_2 - t_1|) = \frac{\gamma^2}{2\hbar^2} \langle \{ x_n(t_2 - t_1), x_n(0) \} \rangle \beta - \frac{\gamma^2}{\hbar^2} \langle x_n(0) \rangle \beta,
\]

(4.78)

\[
B_1(t_2 - t_1) = \frac{\gamma^2}{2\hbar^2} \langle \{ x_n(t_2 - t_1), x_n(0) \} \rangle \beta.
\]

(4.79)

Here, we have assumed that the generator of the environment’s time evolution is time independent such that the thermal expectation value \( \langle x_n(t_1) \rangle \beta = \langle x_n(0) \rangle \beta \) is independent of time while \( \langle \{ x_n(t_2), x_n(t_1) \} \beta = \langle \{ x_n(t_2 - t_1), x_n(0) \} \beta \) and the same holds for the anti commutator.

Let us check the consistency of the results in Eqs. (4.77)-(4.79) with the results of the previous paragraph. Denoting the oscillator’s annihilation and creation operators \( a_n \) and \( a_n^\dagger \), we can write its Hamiltonian \( H_{\text{osc}} = \hbar \omega_n a_n^\dagger a_n \) omitting the ground state energy. The thermal state of a harmonic oscillator at inverse temperature \( \beta \) was given in Eq. (2.367). We have

\[
\rho_\beta = e^{-\beta H_{\text{osc}}}/\text{Tr}e^{-\beta H_{\text{osc}}} = \sum_{k=0}^\infty \frac{\bar{n}(\omega_n)^k}{(1 + \bar{n}(\omega_n))^{1+k}} |k\rangle \langle k|,
\]

(4.80)

where

\[
\bar{n}(\omega_n) = \frac{1}{e^{\beta \omega_n} - 1}.
\]

(4.81)

The Heisenberg operator \( x_n(t) \) reads

\[
x_n(t) = \sqrt{\frac{\hbar}{2m_n \omega_n}} (a_ne^{-i\omega_n t} + a_n^\dagger e^{i\omega_n t}).
\]

(4.82)
Using Eqs. (4.80) and (4.82) we find $C_1(t) = 0$ and

$$A_1(|t|) = \frac{\gamma_n^2}{2\hbar m_n\omega_n}(2\bar{n}(\omega_n) + 1)\cos(\omega_n t) = \frac{\gamma_n^2}{2\hbar m_n\omega_n} \coth\left(\frac{\hbar \beta \omega_n}{2}\right) \cos(\omega_n t),$$

$$B_1(t) = -\frac{\gamma_n^2}{2\hbar m_n\omega_n} \sin(\omega_n t).$$

(4.83)

(4.84)

Inserting the results for $A_1(t), B_1(t), C_1(t)$ into Eq. (4.64) we recover the influence functional in Eq. (4.53) with the environmental correlation function $\tilde{L}_n(t)$ in Eq. (4.54). Furthermore, comparing the influence functional in Eq. (4.53) with the general influence functional in Eq. (4.64) we find

$$\tilde{L}_n(t) = \hbar A_1(t) + iB_1(t_1) = \frac{\gamma_n^2}{\hbar} \langle x_n(t)x_n(0) \rangle_{\beta}.$$  

(4.85)

We can easily generalize the above result to the influence functional of an environment of independent harmonic oscillators using the property Eq. (4.48) of influence functionals. Assuming all oscillators in the reservoir are at the same temperature, we recover the influence functional Eq. (4.55) with $\tilde{L}(t)$ given in Eq. (4.56). Generalizing Eq. (4.85) to this case yields

$$\tilde{L}(t) = \sum_n \frac{\gamma_n^2}{\hbar} \langle x_n(t)x_n(0) \rangle_{\beta},$$

(4.86)

and we thus recover Eq. (4.63) from the beginning of this section. The correlation function of the environment $\tilde{L}(t)$ is determined by the coordinate correlation functions of the environmental oscillators weighted by their couplings to the principle system. Accordingly, the two-time correlation functions of the environmental coordinate determine the influence functional.

For the case of an environment consisting of many oscillators it is common to define the collective coordinate $X = \sum_n \gamma_n x_n$ such that $\tilde{L}(t)$ can be written as

$$\tilde{L}(t) = \frac{1}{\hbar} \langle X(t)X(0) \rangle_{\beta} = \frac{1}{\hbar} \sum_n \gamma_n x_n(t) \sum_m \gamma_m x_m(0) \rangle_{\beta} = \sum_n \frac{\gamma_n^2}{\hbar} \langle x_n(t)x_n(0) \rangle_{\beta},$$

(4.87)

where we used that the oscillators are independent and $\langle x_n(0) \rangle_{\beta} = 0$ in the last step.

**The spectral density and the fluctuation dissipation theorem**

Let us now turn to the spectral representation of the functions $A_1(t)$ and $B_1(t)$ from Eqs. (4.78) and (4.79). To this end we will use a few results of linear response theory. Instructive accounts of linear response theory can be found for example in [127, 148]. Let us consider the coordinate correlation function of a harmonic oscillator in a thermal state at inverse temperature $\beta$

$$C_{xx}(t) = \langle x_n(t)x_n(0) \rangle_{\beta} - \langle x_n(t) \rangle_{\beta} \langle x_n(0) \rangle_{\beta}. $$

(4.88)

We can decompose $C_{xx}(t)$ as

$$C_{xx}(t) = S(t) + iA(t),$$

(4.89)

where

$$S(t) = \frac{1}{2} \langle \{x_n(t), x_n(0) \} \rangle_{\beta} - \langle x_n(t) \rangle_{\beta} \langle x_n(0) \rangle_{\beta},$$

(4.90)

$$A(t) = \frac{i}{2} \langle [x_n(t), x_n(0)] \rangle_{\beta}. $$

(4.91)

Realizing that the thermal expectation value $\langle x_n(t) \rangle_{\beta} = \langle x_n(0) \rangle_{\beta}$ and comparing with Eq. (4.78), we see that the function $A_1(t)$ is proportional to $S(t)$: $A_1(t) = (\gamma_n^2/\hbar^2)S(t)$. Similarly $B_1(t)$ is...
proportional to $A(t)$: $B_1(t) = -(\gamma n^2/h^2)A(t)$. Thus, in order to avoid the prefactors, we can consider $S(t)$ and $A(t)$ instead of $A_1(t)$ and $B_1(t)$. We will also assume $\langle x_n(t)\rangle_\beta = \langle x_n(0)\rangle_\beta = 0$ in the following.

Let us first realize that the real part $S(t) = S(-t)$ is a symmetric function of time while the imaginary part $A(t) = -A(-t)$ is an antisymmetric function of time. Furthermore, the function $A(t)$ is proportional to the quantum mechanical response function $\chi_n(t)$ of a harmonic oscillator driven by an external force coupling to its coordinate [127]

$$\chi_n(t) = \frac{1}{\hbar} \Theta(t)[x_n(t), x_n(0)]_\beta = -\frac{\gamma}{\hbar} \Theta(t)A(t). \quad (4.92)$$

For a linear system the system’s response is completely determined by the linear response function. Recall that the harmonic oscillator is a linear system. The Fourier transform of the response function is a complex function called the dynamical susceptibility

$$\chi_n(\omega) = \chi_n^\prime(\omega) + i\chi_n^\prime\prime(\omega) = \int_0^\infty \! dr \chi_n(t)e^{i\omega t}. \quad (4.93)$$

The imaginary part of the dynamical susceptibility is an odd function $\chi_n(\omega) = -\chi_n(-\omega)$ and the power dissipated by the external force is proportional to it [148]. Moreover, we may write $A(t)$ in terms of the imaginary part of the dynamical susceptibility [127]

$$A(t) = \frac{\hbar}{2\pi i} \int_{-\infty}^{\infty} \! d\omega \chi_n^\prime(\omega)e^{-i\omega t} = -\frac{\hbar}{2\pi} \int_{-\infty}^{\infty} \! d\omega \chi_n^\prime\prime(\omega)\sin(\omega t), \quad (4.94)$$

where we used that $\chi(\omega)$ is an odd function in the last step.

Let us now consider the Fourier transform of the real part $S(t)$

$$S(\omega) = \int_{-\infty}^{\infty} \! dt S(t)e^{i\omega t}. \quad (4.95)$$

The fluctuation spectrum $S(\omega)$ is connected to the imaginary part of the dynamical susceptibility $\chi_n^\prime\prime(\omega)$ by the fluctuation-dissipation theorem (FDT) [127]

$$S(\omega) = \hbar \coth \left( \frac{\beta \hbar \omega}{2} \right) \chi_n^\prime\prime(\omega). \quad (4.96)$$

Inverting the Fourier transform in Eq. (4.95) and using Eq. (4.94), we obtain the frequency space representation of $C_{xx}(t)$. Using the fluctuation-dissipation theorem and that $\chi_n^\prime(\omega)$ is an odd function, we arrive at

$$C_{xx}(t) = \langle x_n(t)x_n(0)\rangle_\beta = \frac{\hbar}{\pi} \int_0^\infty \! d\omega \chi_n^\prime(\omega) \left[ \coth \left( \frac{\hbar \beta \omega}{2} \right) \cos(\omega t) - i\sin(\omega t) \right]. \quad (4.97)$$

According to Eq. (4.85) we obtain the function $\tilde{L}_n(t)$ for a single oscillator from the above equation by multiplying by $\gamma_n^2/\hbar$

$$\tilde{L}_n(t) = \frac{\gamma_n^2}{\pi} \int_0^\infty \! d\omega \chi_n^\prime\prime(\omega) \left[ \coth \left( \frac{\hbar \beta \omega}{2} \right) \cos(\omega t) - i\sin(\omega t) \right]. \quad (4.98)$$

For the free harmonic oscillator, $\chi_n^\prime\prime(\omega)$ actually reads [127]

$$\chi_n^\prime\prime(\omega) = \frac{\pi}{2m_n\omega_n} \left( \delta(\omega - \omega_n) - \delta(\omega + \omega_n) \right). \quad (4.99)$$
4.5. Effective spectral densities of damped harmonic oscillators

Substituting $\chi''_n(\omega)$ into Eq. (4.98) we recover $\tilde{L}_n(t)$ from Eq. (4.54). For a bath of independent oscillators, Eq. (4.98) becomes

$$\tilde{L}(t) = \frac{1}{\pi} \int_0^\infty d\omega \sum_n \gamma_n^2 \chi''_n(\omega) \left[ \coth \left( \frac{\hbar \beta \omega}{2} \right) \cos(\omega t) - i \sin(\omega t) \right]$$

(4.100)

such that with Eq. (4.99) we recover Eq. (4.56). Note that $\tilde{L}(t)$ can be written in terms of a single spectral density

$$\tilde{J}(\omega) = \Theta(\omega) \sum_n \gamma_n^2 \chi''_n(\omega)$$

(4.101)

as in Eq. (4.58). Inserting the susceptibility from Eq. (4.99), indeed we obtain the spectral density $\tilde{J}(\omega)$ of Eq. (4.57).

Let us briefly recapitulate the results of this section. First, we have seen in Eqs. (4.63) and (4.86) that the environmental correlation function $\tilde{L}(t)$, and thus also $L(t)$ from Eq. (4.62), which determines the influence functional of an oscillator environment coupled to a principal system through the oscillator coordinates, is determined by the two-time correlation function of the oscillator’s coordinate. Second, we have seen that the validity of the fluctuation-dissipation theorem ensures that we can write the function $\tilde{L}(t)$ in terms of the single spectral density $\tilde{J}(\omega)$ as in Eq. (4.58).

4.5. Effective spectral densities of damped harmonic oscillators

So far, we have considered influence functionals for environments comprised by free harmonic oscillators. We have seen that the influence of these environments on the principal system is completely determined by the spectral density or, equivalently, by the environmental correlation function $\tilde{L}(t)$, Eq. (4.58), which turns into the function $L(t)$, Eq. (4.62), for spin-boson models. A further observation that we could make is that each of the free oscillators has a $\delta$ contribution to the spectral density. Thus, one needs theoretically an infinite or in practice a very large number of oscillators to produce a continuous spectral density.

In [129], it is shown that an oscillator damped by an oscillator bath with Ohmic spectral density yields a Lorentzian spectral density. The effective spectral density is given by the normal modes of the coupled system oscillator plus reservoir. The width, height and location of the Lorentzian depend on the damping rate, coupling and frequency of the damped harmonic oscillator. Hence, if one could implement such a system experimentally, width, height and location of the Lorentzian could be tunable in an experiment. Moreover, combining several damped oscillators with controllable parameters, one can theoretically tailor arbitrary spectral densities. This might, however, require a large number of modes. Interestingly, already with a few modes non-trivial spectral densities can be tailored [149]. We want to apply this idea to crystals of trapped ions. Cooling of trapped ions is, however, described by a Lindblad master equation and it is not clear that the result of [129] holds for this description of cooling.

Therefore, we will study the influence functional and spectral densities generated by damped oscillators with different damping models in this section. In particular, we will consider an oscillator damped by a bath with Ohmic spectral density and the damped oscillator in Lindblad description. We will see that the result of [129] approximately holds for appropriate parameters when damping is described by a Lindblad equation.

Let us note here that damped harmonic oscillators in Lindblad description have been proposed for the simulation of non-Markovian dynamics in oscillator systems [150, 151] and also in the theory of pseudomodes to describe quantum optical processes, especially the interaction of an atom with a structured reservoir in a cavity [152, 153].
4. Simulations of the dynamics of spin-boson models with trapped ions

4.5.1. Two-time correlation functions of damped oscillators

In this section we will study the two-time coordinate correlation function of the damped oscillator in Lindblad description, which we also call “Lindblad oscillator”, and the coordinate correlation function of an oscillator damped by an Ohmic bath (“Ohmic oscillator”). Although in most cases one characterizes an environment by its spectral density, here we start by comparing the coordinate correlation functions of harmonic oscillators in these two models of damping. As we have seen above, the two-time correlation function and the spectral density equivalently specify the influence of the environment on the system. It turns out, however, that it is easier to establish a connection between the effective environments generated by the two models of damping in the time domain. The frequency domain will be considered in the next paragraph. Note that the presentation follows closely the supplemental material of the publication [L2].

Let us first motivate why we consider the correlation functions of damped oscillators. We have seen in the previous section that the influence of an oscillator environment on a principal system is determined by the coordinate correlation functions of the environmental oscillators for a coupling of the form in Eq. (4.51). Now, if we consider an environment consisting of a single damped oscillator with the same coupling, we could follow the reasoning of the previous section. Assuming the damping of the oscillator is caused by an oscillator bath we could insert an influence functional $G[x,x']$ to model the damping of the oscillator in Eq. (4.70). This would then also appear in Eqs. (4.71)-(4.73) such that the function $L(t)$ determining the influence of the damped oscillator on the principal system is the coordinate correlation function of the damped harmonic oscillator.

Now, the final stage of cooling of the normal modes of a trapped-ion crystal in many cases is well described by a Lindblad master equation (see Section 2.6). Accordingly, the Lindblad description of damping will be our starting point. In order to characterize the influence of the Lindblad oscillator on a spin, we have to consider its reservoir correlation function $L(t)$ from Eq. (4.61). Combining Eqs. (4.61) and (4.85), we write $L(t)$ as

$$L(t) = q_0^2 \gamma^2 \hbar^2 \langle x_n(t)x_n(0) \rangle \bar{\rho} = L'(t) + iL''(t). \quad (4.102)$$

We omit the sum here because we are considering only a single oscillator. For ease of notation we omit the index $n$ in the following and consider $\langle x(t)x(0) \rangle \bar{\rho}$ avoiding the prefactors in $L(t)$.

We recall from Eq. (4.89) that we may decompose the coordinate correlation function as

$$\langle x(t)x(0) \rangle \bar{\rho} = S(t) + iA(t), \quad (4.103)$$

where $S(t)$ and $A(t)$ are defined in Eqs. (4.90) and (4.91), respectively,

$$S(t) = \frac{1}{2} \langle \{x(t),x(0)\} \rangle \bar{\rho}, \quad (4.104) \quad A(t) = \frac{1}{2} \langle [x(t),x(0)] \rangle \bar{\rho}. \quad (4.105)$$

Here, we already anticipated that $\langle x(t) \rangle \bar{\rho} = 0$ for the Lindblad oscillator in a thermal state. The equation of motion of the Lindblad oscillator is given in Eq. (2.366) and in the Schrödinger picture may be written as

$$\dot{\rho} = \mathbb{L}_k \rho = -i[\omega_m a^\dagger a, \rho] + D_{k,\bar{\rho}} \rho, \quad (4.106)$$

where the subscript lc stands for “laser cooling”, $a$ and $a^\dagger$ are the ladder operators of the considered oscillator and $\omega_m$ its frequency. We assume that $\omega_m$ incorporates possible renormalizations (Lamb shifts) of the mode frequency due to the damping. The dissipator is given by

$$D_{k,\bar{\rho}} \rho = \kappa (\bar{n} + 1)[a^\dagger a \bar{\rho} - a^\dagger a^\dagger \bar{\rho}] + \kappa \bar{n} [a^\dagger \rho a - a a^\dagger \rho] + \text{H.c.} \quad (4.107)$$
4.5. Effective spectral densities of damped harmonic oscillators

We denote the coordinate correlation function of the Lindblad oscillator by

\[ \langle x(t)x(0) \rangle_{\beta,L} = S_{L}(t) + iA_{L}(t). \]  

(4.108)

We can readily obtain \( S_{L}(t) \) and \( A_{L}(t) \) using the quantum regression theorem introduced in Section 2.4.3. With the help of the identities

\[ \mathcal{L}_{ic}(\rho_{\beta}a^\dagger) = ( -\kappa - i\omega_{m})\rho_{\beta}a^\dagger, \]  

(4.109)

\[ \mathcal{L}_{ic}(a^\dagger\rho_{\beta}) = ( -\kappa - i\omega_{m})a^\dagger\rho_{\beta}, \]  

(4.110)

and

\[ \mathcal{L}_{ic}(\rho_{\beta}a) = ( -\kappa + i\omega_{m})\rho_{\beta}a, \]  

(4.111)

\[ \mathcal{L}_{ic}(a\rho_{\beta}) = ( -\kappa + i\omega_{m})a\rho_{\beta}, \]  

(4.112)

where \( \rho_{\beta} \) is a thermal state of the oscillator as in Eq. (4.80), we obtain

\[ S_{L}(t) = \frac{\hbar}{2m\omega_{m}} \coth \left( \frac{\beta \hbar \omega_{m}}{2} \right) \cos(\omega_{m}|t|)e^{-\kappa|t|} \]  

(4.113)

and

\[ A_{L}(t) = -\frac{\hbar}{2m\omega_{m}} \sin(\omega_{m}|t|)e^{-\kappa|t|}, \]  

(4.114)

where \( m \) is the mass of the oscillator. Let us recall that a necessary condition for the derivation of the Lindblad equation with dissipator in Eq. (4.107) is

\[ \kappa \ll \omega_{m}, \]  

(4.115)

which is required for the secular approximation. Thus, this is the regime we consider in the following.

In order to proceed, let us note that inserting \( A_{L}(t) \) into Eq. (4.92) we find that the response function of the Lindblad oscillator is actually the same as that of the classical damped harmonic oscillator obeying the equation of motion

\[ m\ddot{x} + \eta\dot{x} + m\omega_{0}^{2}x = F_{ext}(t). \]  

(4.116)

Here, \( m \) is the oscillator’s mass, the friction coefficient \( \eta \) determines the damping of the oscillator \( \kappa = \eta/(2m) \) and \( F_{ext}(t) \) is some external force. The spectral density leading to such an equation of motion is the Ohmic spectral density [129]

\[ \tilde{J}_{\Omega}(\omega) = \eta \omega e^{-\omega/\omega_{c}}. \]  

(4.117)

Hence, the Lindblad oscillator has the same response function as an oscillator damped by a reservoir with Ohmic spectral density. In the light of these considerations, it seems obvious that we should compare the correlation function \( L_{c}(t) \) of the Lindblad oscillator with that of the oscillator damped by an Ohmic bath. Note that indeed it is shown in [129] that an oscillator damped by a strict Ohmic spectral density, i.e. an Ohmic spectral density with infinite cutoff frequency \( \omega_{c} \), leads to an effective Lorentzian spectral density.

Let us, therefore, move on to the harmonic oscillator damped by a thermal oscillator bath with Ohmic spectral density. For a strictly Ohmic spectral density the coordinate correlation function \( \langle x(t)x(0) \rangle_{\beta} \) can be evaluated analytically [127,146,154]. We denote the free oscillation frequency of the oscillator by \( \Omega \) while we denote the damping rate on the oscillator’s coordinate
by \( \kappa_{\text{ohm}} \). The free oscillation frequency of the oscillator is reduced by the damping. In the underdamped regime \( \kappa_{\text{ohm}} < \Omega \) we have [127, 146]

\[
\omega_t = \sqrt{\Omega^2 - \kappa_{\text{ohm}}^2}.
\] (4.118)

Since according to Eq. (4.115) we are bound to the regime \( \kappa \ll \omega_m \) for the Lindblad oscillator, we expect that it suffices to consider the Ohmic oscillator in the underdamped regime. We write the coordinate correlation function of the Ohmic oscillator as

\[
\langle x(t)x(0) \rangle_{\beta, \omega} = S_O(t) + iA_O(t).
\] (4.119)

The real part of the coordinate correlation function \( S_O(t) \) is composed of two contributions [127]

\[
S_O(t) = S_1(t) + S_2(t)
\] (4.120)

given by

\[
S_1(t) = \frac{\hbar}{2m\omega_t} \left[ \frac{\sinh(\beta\omega_t)}{\cosh(\beta\omega_t) - \cos(\beta\kappa_{\text{ohm}})} \cos(\omega_t t) \right. \\
+ \left. \frac{\sin(\beta\kappa_{\text{ohm}})}{\cosh(\beta\omega_t) - \cos(\beta\kappa_{\text{ohm}})} \sin(\omega_t |t|) \right] e^{-\kappa_{\text{ohm}} |t|}
\] (4.121)

and

\[
S_2(t) = -\frac{4\kappa_{\text{ohm}}}{m\beta} \sum_{n=1}^{\infty} \frac{v_n e^{-v_n |t|}}{(\Omega^2 + v_n^2)^2 - 4\kappa_{\text{ohm}}^2 v_n^2}.
\] (4.122)

Here we have introduced the so-called Matsubara frequencies

\[
v_n = \frac{2\pi}{\hbar \beta} n.
\] (4.123)

We already anticipated above that the form of the imaginary part \( A_O(t) \) is the same as that of \( A_L(t) \) in Eq. (4.114) above. Indeed, we have [127, 146]

\[
A_O(t) = -\frac{\hbar}{2m\omega_t} \sin(\omega_t t) e^{-\kappa_{\text{ohm}} |t|}.
\] (4.124)

It is readily seen that the imaginary parts of the coordinate correlation functions \( A_O(t) \) and \( A_L(t) \) from Eqs. (4.124) and (4.114) coincide for \( \omega_t = \omega_m \) and \( \kappa_{\text{ohm}} = \kappa \). Accordingly, we set \( \omega_t = \omega_m \) and \( \kappa_{\text{ohm}} = \kappa \). However, also with this substitution, the symmetric parts \( S_0(t) \) and \( S_L(t) \) do not coincide. Hence, we will investigate if the functions \( S_O(t) \) and \( S_L(t) \) can be made to coincide approximately.

Obviously, \( S_L(t) \) cannot reproduce the contribution \( S_2(t) \) in \( S_O(t) \) as well as the \( \sin(\hbar \beta \kappa) \) in \( S_1(t) \) which yields a contribution that is out of phase with \( S_L(t) \). In order to achieve that the two functions coincide, we need to be able to neglect these two contributions.

Let us start with \( S_2(t) \). The decay of \( S_2(t) \) is governed by the Matsubara frequencies, the smallest of which is \( v_1 \). \( S_L(t) \) only features a single decay rate \( \kappa \) which also appears in \( S_1(t) \) and consequently, \( S_L(t) \) cannot reproduce the decay in \( S_2(t) \). In order to be able to neglect \( S_2(t) \) we need it to decay much quicker than \( S_L(t) \) and \( S_1(t) \), which leads us to demand

\[
v_1 \gg \kappa.
\] (4.125)

In this regime \( S_2(t) \), only causes corrections to \( S_1(t) \) on a very short time scale and the influence of \( S_2(t) \) can be neglected if we are interested in the dynamics on not too short time scales. If,
moreover, \( S_2(0) \ll S_1(0) \), we can neglect \( S_2(t) \) completely [127]. Inserting the definition of the Matsubara frequencies into Eq. (4.125) we can recast it into the form

\[
\frac{k_b \beta}{2\pi} = \frac{\kappa}{\nu_1} \ll 1.
\] (4.126)

The above equation tells us that higher temperatures are more favorable if we want \( S_2(t) \) to decay quickly. For a given cooling rate \( \kappa \) it provides us with a rough lower bound on the temperature where we can neglect \( S_2(t) \). If the condition in Eq. (4.126) is satisfied, it ensures that the smallest Matsubara frequency is much larger than the decay rate \( \kappa \) so that we can neglect \( S_2(t) \) on the timescales of interest. The decay of correlations is then given by \( S_1(t) \).

Note that the condition in Eq. (4.126) is usually fulfilled unless we are dealing with very low temperatures. For very low temperatures it is well-known that the Matsubara frequencies in \( S_2(t) \) dictate the timescale for the decay of correlations [74, 127, 154]. The Matsubara frequencies then become very small and in the limit of \( T \to 0 \) they approximately yield a continuum of frequencies. The asymptotic decay rate of \( S_2(t) \) in this case shows a characteristic polynomial decay [74, 127, 154]

\[
\lim_{t \to \infty} S(t) \sim \frac{1}{t^2}.
\] (4.127)

Let us now assume that we are in the regime Eq. (4.126) and can disregard \( S_2(t) \). We now want to find the regime where

\[
S_1(t) \approx S_1(t).
\] (4.128)

Note that in addition to Eq. (4.126) we also consider \( \kappa \ll \omega_m \) because of Eq. (4.115) above. A first order expansion of the sine and cosine terms in \( S_1(t) \) in the small parameter \( \beta h \kappa \ll 1 \) yields

\[
S_1(t) \approx \frac{\hbar}{2m \omega_m} \left[ \frac{\sinh(\beta \hbar \omega_m)}{\cosh(\beta \hbar \omega_m) - 1} \cos(\omega_m t) \right. \\
+ \left. \frac{\beta h \kappa}{\cosh(\beta \hbar \omega_m) - 1} \sin(\omega_m t) \right] e^{-\kappa |t|}
\] (4.129)

\[
\approx \frac{\hbar}{2m \omega_m} \sinh(\beta \hbar \omega_m) \left( \frac{1}{\cosh(\beta \hbar \omega_m) - 1} \right) \cos(\omega_m t) e^{-\kappa |t|}.
\]

Here we have used \( \beta \hbar \kappa \ll \sinh(\hbar \beta \omega_m) \) in the last step, which should hold in the regime \( \kappa \ll \omega_m \). Applying the identity \( \coth \frac{x}{2} = \sinh(x)/(\cosh x - 1) \) to the last equation yields

\[
S_1(t) = \frac{\hbar}{2m \omega_m} \coth \left( \frac{\beta \hbar \omega_m}{2} \right) \cos(\omega_m t) e^{-\kappa |t|}.
\] (4.130)

Now, if the reservoir of the Ohmic and Lindblad oscillators are at the same temperature, we have

\[
S_1(t) = S_L(t).
\] (4.131)

Summarizing the above considerations, we have established a regime where the coordinate correlation functions of the Lindblad oscillator and the Ohmic oscillator coincide. We expect very good agreement between the two correlation functions

\[
\langle x(t)x(0) \rangle_{\beta,L} \approx \langle x(t)x(0) \rangle_{\beta,O},
\] (4.132)

if conditions (4.115) and (4.126) are fulfilled and if the reservoirs are at the same inverse temperature \( \beta \).

In particular, we have seen that the Lindblad oscillator whose correlation functions we computed using the quantum regression theorem can only be identified with the oscillator damped
by a reservoir with Ohmic spectral density in a restricted parameter regime. This is connected to some problems of the quantum regression which we shall allude to in the next section. For now, we note that the regime we found above is actually the parameter regime where references [74, 154] estimate that the quantum regression theorem can yield quantitatively correct results for the two-time correlation functions of the damped harmonic oscillator.

Finally, inserting Eqs. (4.113) and (4.114) into Eq. (4.102) and using Eq. (4.4), we obtain the reservoir correlation function of the Lindblad oscillator

\[ L_L(t) = L'_L(t) + iL''_L(t) \]  
(4.133)

with real and imaginary parts

\[ L'_L(t) = \lambda^2 \coth \left( \frac{\beta \hbar \omega_m}{2} \right) \cos(\omega_m t) e^{-\kappa |t|} \]  
(4.134)

and

\[ L''_L(t) = -\lambda^2 \sin(\omega_m t) e^{-\kappa |t|}. \]  
(4.135)

For the Ohmic oscillator with the help of Eqs. (4.121)-(4.124) we obtain the reservoir correlation function

\[ L_O(t) = L'_O(t) + iL''_O(t) = L_1(t) + L_2(t) + iL''_O(t), \]  
(4.136)

where

\[ L_1(t) = \lambda^2 \left[ \frac{\sinh(\beta \hbar \omega_m)}{\cosh(\beta \hbar \omega_m) - \cos(\beta \hbar \kappa)} \cos(\omega_m t) \right. \]

\[ + \left. \frac{\sin(\beta \hbar \kappa)}{\cosh(\beta \hbar \omega_m) - \cos(\beta \hbar \kappa)} \sin(\omega_m |t|) \right] e^{-\kappa |t|}, \]  
(4.137)

\[ L_2(t) = -\lambda^2 \frac{8\kappa \omega_m}{\hbar \beta} \sum_{n=1}^{\infty} \frac{\nu_n e^{-\nu_n |t|}}{(\Omega^2 + \nu_n^2)^2 - 4\kappa^2 \nu_n^2} \]

and

\[ L''_O(t) = -\lambda^2 \sin(\omega_m t) e^{-\kappa |t|}. \]  
(4.138)

Before we move on to a more quantitative comparison of \( L_1(t) \) and \( L_O(t) \), we remark that in the context of ion-trap experiments thermal states of the bosonic normal modes are usually characterized by their mean occupation number \( \bar{n} \) rather than a temperature or inverse temperature. We can associate an effective temperature \( T_{\text{eff}} \) to a mode of frequency \( \omega_m \) with mean occupation number \( \bar{n}(\omega_m) \) solving Eq. (4.81) for the temperature, which yields

\[ T_{\text{eff}} = \frac{\hbar \omega_m}{k_B \log(1 + 1/\bar{n}(\omega_m))}. \]  
(4.139)

With the above equation we can also write the condition in Eq. (4.26) in terms of \( \bar{n}(\omega_m) \) yielding

\[ \frac{\log \left( 1 + \frac{1}{\bar{n}(\omega_m)} \right)}{2 \pi} \frac{\kappa}{\omega_m} \ll 1. \]  
(4.140)

In order to underpin the result that the reservoir correlation functions of the damped oscillator in the Lindblad description \( L_L(t) \) and that of an oscillator damped by an Ohmic bath \( L_O(t) \) coincide if the conditions in Eqs. (4.115) and (4.126) are fulfilled, we present a numerical comparison of the real parts \( L'_L(t) \), Eq. (4.134), and \( L''_O(t) = L_1(t) + L_2(t) \), Eq. (4.137). Recall that the imaginary parts are identical.
Figure 4.2 shows $L'_O(t)$ (solid lines) including the first $10^4$ Matsubara frequencies and $L_L(t)$ (dashed-line and crosses) for $\omega_m/2\pi = 100$kHz, $\kappa/2\pi = 1.25$kHz and $\bar{n} (\omega_m) = 0.025$ – parameters that are realistic for an ion trap experiment. Note that this occupation number corresponds to an inverse temperature $\hbar\beta = 5.91 \cdot 10^{-6}$s. Part a) of the figure shows the short time behavior while part b) shows intermediate time scales. Indeed, we obtain excellent agreement between the two functions on both time scales.

![Figure 4.2.](image)

Figure 4.2.: The figure shows the real parts $L'_O(t) = L_1(t) + L_2(t)$ (blue solid lines) and $L'_L(t)$ (dashed-dot line and crosses) of the reservoir correlation functions for the Ohmic and Lindblad oscillators from Eqs. (4.137) and (4.134). $L'_O(t)$ includes the first $10^4$ Matsubara frequencies. The parameters are $\omega_m/2\pi = 100$kHz, $\kappa/2\pi = 1.25$kHz and $\bar{n} (\omega_m) = 0.025$ which corresponds to $\hbar\beta = 5.91 \cdot 10^{-6}$s. Panel a) shows the time evolution for short times, while panel b) illustrates the intermediate time behavior.

Before we close this section, we present a small survey of the quality of the match between the correlation functions for different values of $\kappa$ and $\bar{n}(\omega_m)$ while keeping the mode frequency $\omega_m/2\pi = 100$kHz fixed. We choose to quantify the quality of the match by the distance

$$d = \frac{1}{\lambda \pi} \left| \int_0^\infty dt [L_O(t) - L_L(t)] \right|$$

(4.141)

between the functions $L_O(t)$ and $L_L(t)$. Actually, $d$ can be evaluated analytically and yields

$$d = \left| c_0 \frac{\kappa}{\kappa^2 + \omega_m^2} + c_m \frac{\omega_m}{\kappa^2 + \omega_m^2} - \frac{8 \kappa \omega_m}{\hbar \beta} \sum_{n=1}^{\infty} \frac{1}{(\omega_m^2 + \kappa^2 + \nu_n^2 - 4 \kappa^2 \nu_n^2)} \right|$$

(4.142)

where we introduced

$$c_0 = \frac{\sinh(\beta \hbar \omega_m)}{\cosh(\beta \hbar \omega_m) - \cos(\hbar \beta \kappa)} - \coth\left(\frac{\hbar \beta \omega_m}{2}\right),$$

$$c_m = \frac{\sin(\beta \hbar \kappa)}{\cosh(\beta \hbar \omega_m) - \cos(\hbar \beta \kappa)}.$$

In Fig. 4.3 we plot the values of $d$ for different cooling rates $\kappa$ and mean occupation numbers $\bar{n}(\omega_m)$. Note that the $z$-axis is $-\log d$ such that higher bars indicate smaller values of $d$. We can make two observations: For fixed $\bar{n}(\omega_m)$ larger values of $\kappa$ increase the difference $d$ between $L_O(t)$ and $L_L(t)$. On the other hand, for a fixed cooling rate the distance between the two functions has a minimum for intermediate values of $\bar{n}(\omega)$. 

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4.5. Effective spectral densities of damped harmonic oscillators

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131
4. Simulations of the dynamics of spin-boson models with trapped ions

Both effects can be understood considering Eqs. (4.134) and (4.137). Firstly, for fixed temperature increasing $\kappa$ increases the ratio $\kappa/\nu_1$ and makes it less favorable. Increasing $\kappa$ also increases the term $\sin(\hbar \beta \kappa)$ in $L_1(t)$. For fixed cooling rate $\kappa$ we have the following situation: in order to neglect $L_2(t)$ we favor higher temperatures due to Eq. (4.126). However, in order to suppress the sine component in $L_1(t)$ lower temperatures are more favorable. Accordingly there is an intermediate temperature regime where we obtain the best match between $L_O(t)$ and $L_L(t)$.

Figure 4.3.: The figure depicts the distance $d$ from Eqs. (4.141) and (4.142) between the correlation functions $L_O(t)$ and $L_L(t)$. We include the first $10^4$ Matsubara frequencies in the evaluation of $L_O(t)$. The distance is shown for different values of $\kappa$ and $n(\omega_m)$ for fixed mode frequency $\omega_m/2\pi = 100$kHz. Note that higher bars correspond to smaller values of $d$.

4.5.2. Effective spectral density of Lindblad and Ohmic damped oscillator

In the previous paragraph, we have seen that the Lindblad oscillator and the harmonic oscillator damped by a reservoir with Ohmic spectral density yield the same coordinate correlation function in an appropriate parameter regime. Let us now consider the spectral densities associated to the damped oscillators.

We start by considering the oscillator damped by a thermal oscillator environment with Ohmic spectral density. This model of damping is investigated in [129]. The Ohmic spectral density in this work is taken to read

$$\tilde{J}_O = \eta \omega e^{-\omega/c^\omega}. \quad (4.143)$$

The final result is obtained in the limit $\omega_c \to \infty$ of a strict Ohmic spectral density. It is shown in [129] that in this limit an effective spectral density $J_{\text{eff,ohm}}(\omega)$ can be associated to the damped oscillator. Adapting to our notation conventions, the effective spectral density of the damped harmonic oscillator takes the form

$$J_{\text{eff}}(\omega) = \frac{q^2_0}{\hbar} J_{\text{eff,ohm}}(\omega) = \frac{q^2_0 \gamma^2}{\hbar m} \frac{2\lambda}{(\Omega^2 - \omega^2)^2 + 4\omega^2 \kappa^2}. \quad (4.144)$$

As before $\kappa$ is the damping on the coordinate of the oscillator due to the oscillator bath, $\Omega$ the
4.5. Effective spectral densities of damped harmonic oscillators

The frequency representation of $L$ where

$$\omega_m = \sqrt{\Omega^2 - \kappa^2}. \quad (4.145)$$

Using $\omega_m$ and Eq. (4.4) we can write the effective spectral density in Eq. (4.144) as

$$J_{\text{eff}}(\omega) = \lambda^2 \left[ \frac{\kappa}{\kappa^2 + (\omega - \omega_m)^2} - \frac{\kappa}{\kappa^2 + (\omega + \omega_m)^2} \right]. \quad (4.146)$$

We call the effective spectral density $J_{\text{eff}}(\omega)$ Lorentzian although it consists of the difference of two Lorentzians centered at $\pm \omega_m$. Inserting the above spectral density into Eq. (4.62), we obtain

$$L(t) = \frac{1}{\pi} \int_0^\infty d\omega J_{\text{eff}}(\omega) \left[ \coth \left( \frac{\hbar \beta \omega}{2} \right) \cos(\omega t) - i \sin(\omega t) \right], \quad (4.147)$$

and one recovers $L_O(t)$ from Eq. (4.136) by contour integration. Actually, the integral to be solved is the same as the one used to find the position correlation function of the damped harmonic oscillator from the imaginary part of its dynamical susceptibility for strict Ohmic damping as presented in e.g. [127, 154].

Let us now turn to the effective spectral density of the Lindblad oscillator. The Fourier representation of $L_L(t)$ is obtained by considering its real and imaginary parts, Eqs. (4.134) and (4.135), separately. Since the imaginary part $L''_L(t)$ coincides with $L''_O(t)$, Eq. (4.138), their spectral representations are the same. However, the real parts $L'_L(t)$ and $L'_O(t)$, Eq. (4.137), differ as we have seen in the previous section. Indeed, one finds that the spectral representation of $L'_L(t)$ is not given by $J_{\text{eff}}(\omega)$ but can be written as

$$L'_L(t) = \frac{1}{\pi} \int_0^\infty d\omega J'_{\text{eff}}(\omega) \coth \left( \frac{\hbar \beta \omega_m}{2} \right) \cos(\omega t), \quad (4.148)$$

where

$$J'_{\text{eff}}(\omega) = \lambda^2 \frac{\coth \left( \frac{\hbar \beta \omega_m}{2} \right)}{\coth \left( \frac{\hbar \beta \omega}{2} \right)} \left[ \frac{\kappa}{\kappa^2 + (\omega - \omega_m)^2} + \frac{\kappa}{\kappa^2 + (\omega + \omega_m)^2} \right]. \quad (4.149)$$

The frequency representation of $L_L(t)$ then takes the form

$$L_L(t) = \frac{1}{\pi} \int_0^\infty d\omega \left[ J'_{\text{eff}}(\omega) \coth \left( \frac{\hbar \beta \omega}{2} \right) \cos(\omega t) - i J_{\text{eff}}(\omega) \sin(\omega t) \right]. \quad (4.150)$$

In general $J'_{\text{eff}}(\omega) \neq J_{\text{eff}}(\omega)$ and hence $L_L(t)$ cannot be written in terms of a single spectral density as we expect for an oscillator reservoir correlation function from Eq. (4.62). As we pointed out at the end of Section 4.4.2, the fluctuation-dissipation theorem (FDT) allows us to write the coordinate correlation function and thus the reservoir correlation function in terms of a single quantity – the spectral density. In fact, it is known that the quantum regression theorem is, in general, not compatible with the FDT and the so-called KMS condition which is used in the derivation of the FDT [74]. Therefore, we cannot expect that the identity Eq. (4.62) also holds for the regression theorem results. Yet, from our considerations in the previous section we expect that for appropriate parameters, i.e. when we satisfy Eqs. (4.115) and (4.126), we have

$$J'_{\text{eff}}(\omega) \approx J_{\text{eff}}(\omega) \quad (4.151)$$
4. Simulations of the dynamics of spin-boson models with trapped ions

such that $L_L(t)$ can be written in the form of Eq. (4.62) approximately as

$$L_L(t) \approx \frac{1}{\pi} \int_0^\infty d\omega J_{\text{eff}}(\omega) \left[ \coth \left( \frac{\hbar \beta \omega}{2} \right) \cos(\omega t) - i \sin(\omega t) \right].$$

(4.152)

In order to illustrate that indeed we obtain very good agreement between $J'_{\text{eff}}(\omega)$ and $J_{\text{eff}}(\omega)$, we compare the left and right hand sides of Eq. (4.151) in Fig. 4.4. We use the parameters that yielded a very good agreement between $L_L(t)$ and $L_O(t)$ in the previous section, namely $\omega_m/2\pi = 100\text{kHz}$, $\kappa/2\pi = 1.25\text{kHz}$ and $\bar{n}(\omega_m) = 0.025$ which corresponds to $\hbar \beta = 5.91 \cdot 10^{-6}\text{s}$. In panel a) of the figure we show $J_{\text{eff}}(\omega)$ and $J'_{\text{eff}}(\omega)$ for small frequencies while part b) shows the behavior for intermediate frequencies and around the resonance $\omega_m/2\pi = 100\text{kHz}$. As expected we obtain very good agreement.

Finally, in Fig. 4.5 we plot the relative error

$$\varepsilon_J = \frac{|J'_{\text{eff}}(\omega) - J_{\text{eff}}(\omega)|}{J_{\text{eff}}(\omega)}$$

(4.153)

of the two functions. The error is remarkably small in the range of interest $\omega/2\pi \in [0, 150]\text{kHz}$. For higher frequencies the error increases because the spectral density $J_{\text{eff}}(\omega)$ tends to zero more rapidly than the effective one $J'_{\text{eff}}(\omega)$. However, since both contributions are small the effect of this difference should be negligible, especially because usually we are not interested in the high frequency features of the spectral density.

Summarizing the results of this section: for appropriate parameters, i.e. parameters satisfying Eqs. (4.115) and (4.126), to a very good approximation we obtain the same reservoir correlation functions for a harmonic oscillator damped by a thermal oscillator bath with Ohmic spectral density and an oscillator where damping is described by a Lindblad equation. For these parameters we also obtain very good agreement between the effective spectral densities for the two descriptions of damping. The parameters where the identification is possible are in the regime where the quantum regression theorem is supposed to yield quantitatively correct two-time correlation functions for the damped harmonic oscillator [74, 154].

The interesting consequence of this is the following. The oscillator coupled to a bath with Ohmic spectral density includes an infinite number of oscillators and therefore the oscillator plus bath system constitutes an environment consisting of a macroscopic number of oscillators.
4.6. Experimental simulations of the spin-boson model using trapped ions

With the results that we gathered in the previous sections of this chapter we are now in a position to develop the scheme for the simulation of spin-boson models with trapped ions. We will briefly collect the crucial points of the previous sections. Then, we present the simulation procedure and benchmark it with a numerically exact simulation for the simplest model that we can simulate: a spin coupled to a bath with Lorentzian spectral density. Thereafter, we will show how the necessary Hamiltonian is obtained in an ion trap experiment and we will illustrate the practicality of the protocol with the simulation of an experiment with parameters that can be achieved in state-of-the-art experiments. The discussion of the impact of experimental noise is deferred to the next section.

Let us start by recalling that the spin-boson model is described by the Hamiltonian

\[
H_{sb} = \frac{\epsilon}{2} \sigma^z - \frac{\hbar \Delta}{2} \sigma^x - \frac{1}{2} \sum_{n=1}^{\infty} \hbar \lambda_n (a_n + a_n^\dagger) + \sum_{n=1}^{\infty} \hbar \omega_n a_n^\dagger a_n, \tag{4.154}
\]

where \( \epsilon \) is the energy difference of the eigenstates of \( \sigma^z = |e\rangle \langle e| - |g\rangle \langle g| \) and they are coupled with strength \( \hbar \Delta \). The spin interacts with the environmental oscillators of frequency \( \omega_n \) with coupling strength \( \hbar \lambda_n \). For an initial product state of the spin and the oscillator environment where the environment is in a thermal state, the propagator of the full system can be written in...
the path integral formalism as
\[
G(t,0) = \int_{q_0}^{q_f} \mathcal{D}q \int_{q'_0}^{q'f} \mathcal{D}q' e^{\frac{i}{\hbar} \{ S_0[q] - S_0[q'] \}} F[q,q'].
\] (4.155)

Here \(\int_{q_0}^{q_f} \mathcal{D}q\) denotes the path integral over all spin state trajectories that connect \(q(0) = q_0\) and \(q(t) = q_f\) and \(S_0[q]\) is the action of the free spin evolution. The same holds for the primed coordinate. \(F[q,q']\) is the influence functional given by
\[
F[q,q'] = \exp \left\{ -\int_0^t dt_1 \int_0^{t_1} dt_2 [q(t_2) - q'(t_1)] [L(t_1 - t_2)q(t_2) - L'(t_1 - t_2)q'(t_2)] \right\},
\] (4.156)
where
\[
L(t) = \frac{1}{\hbar^2} \langle X(t)X(0) \rangle^\beta
\] (4.157)
is the reservoir correlation function with
\[
X(t) = \sum_n \hbar \lambda_n [a_n(t) + a_n^\dagger(t)].
\] (4.158)

The influence functional is equivalently determined by the spectral density \(J(\omega)\) as the reservoir correlation function may be written as
\[
L(t) = \frac{1}{\pi} \int_0^\infty d\omega J(\omega) \left[ \coth \left( \frac{\hbar \omega}{2} \right) \cos(\omega t) - i \sin(\omega t) \right].
\] (4.159)

For the Hamiltonian in Eq. (4.154) the spectral density takes the form
\[
J(\omega) = \pi \sum_n \lambda_n^2 \delta(\omega - \omega_n).
\] (4.160)

For environments made up of a macroscopic number of constituents the spectral density is taken to be a smooth function of frequency.

For most spectral densities one cannot solve the dynamics of the spin-boson model exactly and therefore has to resort to either approximate or numerical treatments. Numerical treatments are usually limited by the memory time of the environment. Hence, a physical system which simulates the dynamics of spin-boson models has the advantage that it will not suffer from this limitation. Of course, in the end there will be experimental limitations that define the range of applicability of such a device, but a physical quantum simulator could potentially explore parameter regimes which are inaccessible with nowadays numerical treatments.

The challenge to build such a device with trapped ions is to engineer the environment. As we pointed out earlier, one could consider a large crystal with 50-100 ions that features many motional modes and these could serve as the environment. This idea has been studied previously in [34]. One drawback of this approach is that it is difficult to engineer the shape of the spectral density of the environment in addition to the fact that it is hard to control such large crystals.

Here, we want to take a different approach inspired by the result of [129] that an oscillator damped by a bath with Ohmic spectral density yields the effective spectral density
\[
J_{\text{eff}}(\omega) = \lambda^2 \left[ \frac{\kappa}{\kappa^2 + (\omega - \omega_m)^2} \right. - \left. \frac{\kappa}{\kappa^2 + (\omega + \omega_m)^2} \right].
\] (4.161)

Here, \(\lambda\) is the coupling of the oscillator to the spin as in Eq. (4.154) above, \(\omega_m\) its reduced oscillation frequency that relates to the free oscillation frequency \(\Omega\) and the damping rate \(\kappa\) through \(\omega_m = \sqrt{\Omega^2 - \kappa^2}\). We remark that we consider the underdamped regime \(\kappa < \Omega\).
The above result was obtained within a path integral approach. Now, if we use that the combined effect of several independent environments is given by the product of their individual influence functionals, Eq. (4.48), we obtain a reservoir correlation function

\[ L(t) = \frac{1}{\pi} \int_0^\infty d\omega \sum_n J_{\text{eff},n}(\omega) \left[ \coth \left( \frac{\hbar \beta_n \omega}{2} \right) \cos(\omega t) - i \sin(\omega t) \right], \tag{4.162} \]

where \( J_{\text{eff},n}(\omega) \) is the spectral density generated by oscillator \( n \) given by Eq. (4.161) with the corresponding \( \lambda_n, \kappa_n, \omega_n \). Thus, we can understand the combined effect of several damped harmonic oscillators as that of one large environment if all oscillators are coupled to baths of the same temperature. In practice, it probably suffices to have \( \beta_n \approx \beta \) for all \( n \). Hence, if we have several independent damped oscillators in thermal equilibrium with baths at the same temperature, we may construct arbitrary spectral densities

\[ J(\omega) = \sum_n J_{\text{eff},n}(\omega). \tag{4.163} \]

The idea of decomposing spectral densities in terms of Lorentzians of the form in Eq. (4.161) has already been explored in [149] to derive a set of equations similar to the hierarchical equations of motion for the time evolution of non-Markovian quantum systems [149]. In order to approximate a certain target spectral density \( J_T(\omega) \) as in Eq. (4.163) the authors of [149] find the values of the \( \lambda_n, \kappa_n \) and \( \omega_n \) by minimizing the functional

\[ E[\{\lambda_n, \kappa_n, \omega_n\}] = \int_0^\infty d\omega |J_T(\omega) - J(\omega)|^2. \tag{4.164} \]

Remarkably, an Ohmic spectral density can be approximated very well by only three Lorentzians [149]. This procedure theoretically works for all spectral densities if the number of available oscillators is large enough. Note that performing the above minimization is a considerable numerical task already for five to ten modes since it involves three parameters for each mode. Hence, finding a decomposition of a spectral density as in Eq. (4.163) is already a formidable task which is interesting in its own right.

Due to the results of the previous section, we can adapt this procedure to trapped-ion experiments. After laser cooling, the motion of ions in a Coulomb crystal is described by a set of independent normal modes in each spatial direction under normal trapping conditions. Each of the modes is a harmonic oscillator and cooling of the modes is usually described by a Lindblad equation (see Section 2.6). In the previous section we have seen that we can associate the spectral density in Eq. (4.161) to the damped oscillator in Lindblad description if \( \kappa \ll \omega_m \) and \( \hbar \beta \kappa \ll 1 \). Note that these restrictions make the decomposition in Eq. (4.164) more difficult and one should expect that more damped oscillators are needed than if the condition \( \kappa \ll \omega_m \) is relaxed. Let us acknowledge once again that the use of damped harmonic oscillators to model (non-Markovian) environments has been considered previously for oscillator systems [150, 151] and in pseudomode theory [152, 153].

Now that we have presented the general idea, we will present a detailed case study for the simulation of a spin-boson model with trapped ions. We will investigate the simplest case where a spin is coupled to a single damped oscillator which corresponds to a spin-boson model with Lorentzian spectral density as in Eq. (4.161). This system may seem very simple but actually exhibits an interesting phenomenology and has been studied with a variety of numerical and analytical approaches, see e.g. [155–158].

For the spin-boson model with Lorentzian spectral density we only need one spin and one mode. Since laser cooling involves dissipative processes on the internal degrees of freedom of the ions, we cannot use the same ion to encode the spin and to cool the crystal. Therefore, we
need at least $N = 2$ ions. In order to avoid that the cooling lasers interfere with the internal degrees of freedom of the ion encoding the spin, it seems advantageous to work with mixed species crystals. In this case we can use one ion to implement laser cooling and the other to encode the spin. Alternatively we could rely on single site addressing where the lasers are tightly focused such that they only interact with a single ion at a time.

For concreteness we consider a $^{24}\text{Mg}^+ - ^{25}\text{Mg}^+$ crystal. $^{25}\text{Mg}^+$ has a nuclear spin and accordingly features a hyperfine structure. There are two electronic hyperfine ground states manifolds with total angular momentum $F = 2, 3$ whose degeneracy can be lifted by applying an external magnetic field. The hyperfine splitting between the $F = 2$ and $F = 3$ manifolds is about $\omega_0/2\pi \approx 1.8\text{GHz}$. A possible choice for a qubit are the states $|F = 3, m_F = 3 \rangle \equiv |g \rangle$ and $|F = 2, m_F = 2 \rangle \equiv |e \rangle$. Note that these states are susceptible to magnetic field fluctuations and thus less resilient to fluctuating ambient magnetic fields. This is the price we have to pay if we want to use spin-motion coupling in $\sigma^z$. This kind of coupling cannot be implemented on clock states because there will be no differential ac-Stark shift for typical parameters of the applied fields [159]. At a magnetic field of a few Gauss the other hyperfine states are well-separated from the qubit states due to the Zeeman interaction and we can describe the internal levels of $^{25}\text{Mg}^+$ by the two-state Hamiltonian [19, 55]

$$H_s = \hbar \frac{\omega_0}{2} \sigma^z.$$  \hfill (4.165)

We assume that the trapping potentials are harmonic and chosen such that the ions form a string along $z$. Furthermore, we assume that the ions are cold enough that their motion is described by a set of normal modes in all spatial directions, cf. Eq. (2.57),

$$H_m = \sum_{\alpha=x,y,z} \sum_{n=1}^{2} \hbar \omega_{\alpha n} a^\dagger_{\alpha n} a_{\alpha n} ,$$  \hfill (4.166)

where $\omega_{\alpha n}$ is the frequency of mode $n$ in direction $\alpha$ and $a_{\alpha n}$ and $a^\dagger_{\alpha n}$ are the associated annihilation and creation operators. Gathering the motional and spin Hamiltonians, we define

$$H_0 = \hbar \frac{\omega_0}{2} \sigma^z + \sum_{\alpha,n} \hbar \omega_{\alpha n} a^\dagger_{\alpha n} a_{\alpha n} .$$  \hfill (4.167)

The internal states of $^{24}\text{Mg}^+$ do not appear in the above Hamiltonian because we consider it is used to sympathetically cool the crystal. As we have seen in Section 2.6, the internal levels are adiabatically eliminated in the description of laser cooling. Accordingly, we omit the internal levels of $^{24}\text{Mg}^+$ in $H_0$.

The spin transition lies in the microwave regime and accordingly can be driven either by a microwave field or by two laser fields in a two-photon stimulated-Raman configuration which we discussed in Section 2.5. Irrespective of the physical realization we will call the field driving the spin transition the “microwave” in order to avoid confusion with the lasers used to induce the spin-motion coupling.

Let us now consider that the spin is driven by a microwave with frequency $\omega_d$ and Rabi frequency $\Omega_d$ while simultaneously a spin-dependent optical dipole force is applied. The spin dependent forces discussed in Section 2.5 can be applied to $^{25}\text{Mg}^+$, see e.g. [19], by off-resonantly coupling the spin states to the levels of the $P$ manifold with two laser beams near 280nm whose beat note is tuned close to one of the motional mode frequencies. In a rotating wave approximation the interaction of the $^{25}\text{Mg}^+$ ion with the applied fields according to Eqs. (2.124) and (2.288) is described by the Hamiltonian

$$H_{int} = \hbar \frac{\Omega_d}{2} \sigma^+ e^{-i \omega_d t} + \hbar \frac{\Omega_{\text{dof}}}{2} e^{i (k_0 r_2 + \phi_0)} e^{-i \omega_{\text{dof}} t} \sigma^z + \text{H.c.} ,$$  \hfill (4.168)
where $\omega_{\lambda}$, $k_L$, and $\phi_L$ are the effective laser frequency, wave vector and phase, respectively, and $r_2$ is the position operator of the $^{25}$Mg$^+$ ion. In the above equation, we also set the microwave phase to zero and $\Omega_{\text{odf}}$ denotes the effective laser Rabi frequency where we assumed that the Rabi frequency of the running wave contribution to the optical dipole force vanishes $\Omega_{\text{rw}} = 0$. Finally, we used that $k_d \cdot r_2 \approx 0$, where $k_d$ is the microwave wave vector. Note that this assumption also holds if the spin is driven in a two-photon stimulated Raman configuration with copropagating laser beams.

The full Hamiltonian of the system then reads

$$H = H_0 + H_{\text{int}}.$$  \hspace{1cm} (4.169)

We now assume that the effective laser wave vector $k_L = k e_z$ points along the trap axis such that the laser only couples to the motion along $z$. The axial coordinates of the ions read $r_{jz} = z_j^0 + q_{jz}$, where the equilibrium positions $z_j^0$ are determined from Eqs. (2.22) and (2.23) and the displacements from equilibrium can be written in terms of the quantized normal modes

$$q_{jz} = \sum_n \bar{B}_{jn} \sqrt{\frac{\hbar}{2m_j \omega_n}} (a_n + a_n^\dagger),$$  \hspace{1cm} (4.170)

where $m_j$ is the mass of ion $j$ while $\omega_n \equiv \omega_{n,z}$ and $a_n \equiv a_{n,z}$. Note that we are considering a mixed-species crystal with inhomogeneous masses. Thus, $\bar{B}_{jn}$ defined in Eq. (2.69) is the matrix that diagonalizes the harmonic approximation of the full potential $V^z$ from Eq. (2.68) in mass-weighted coordinates. Accordingly, the $\bar{B}_{jn}$ are the normalized amplitudes of normal mode $n$ at ion $j$ in mass-weighted coordinates.

The next step is to transform the Hamiltonian to an interaction with respect to

$$\tilde{H}_0 = \hbar \frac{\Omega_{\text{odf}}}{2} \sigma^z + \hbar \sum_{\alpha,n} \omega_{n,\alpha} a_n^\dagger a_{n,\alpha}.$$  \hspace{1cm} (4.171)

This yields the Hamiltonian

$$\tilde{H}_{\text{int}} = e^{i\tilde{H}_0 (H - \tilde{H}_0)} e^{-i\tilde{H}_0}$$
$$= \hbar \frac{\delta}{2} \sigma^z + \hbar \frac{\Omega_{\text{odf}}}{2} \sigma^z + \left( \hbar \frac{\Omega_{\text{odf}}}{2} \right) \sum_{\alpha,n} \eta_{jn}(a_n e^{-i\omega_{n,\alpha} t} + a_n^\dagger e^{i\omega_{n,\alpha} t}) e^{-i\omega_{n,\alpha} t} \sigma^z + \text{H.c.},$$  \hspace{1cm} (4.172)

where $\delta = \omega_0 - \omega_d$ is the detuning of the microwave from the spin transition and $\tilde{\Omega}_{\text{odf}} = \Omega_{\text{odf}} e^{i(k_d^2 r_2 + \phi_L)}$. In the above equation, we also assumed that the Rabi frequency $\Omega_d$ is real and introduced the Lamb-Dicke factors

$$\eta_{jn} = \bar{B}_{jn} k \sqrt{\frac{\hbar}{2m_j \omega_n}}.$$  \hspace{1cm} (4.173)

Since the masses of the two ions are almost equal, we expect that the mode structure of the crystal is basically that of a crystal with ions of equal masses. We thus expect an in- and an out-of-phase mode of motion that are well separated in frequency similar to the center-of-mass and stretch modes. We will see below that these assumptions hold.

We now use that the $\eta_{jn} \ll 1$ because $k_d$ is an optical wave vector and expand the exponentials to first order in the $\eta_{2jn}$ to obtain a coupling Hamiltonian of the type in Eq. (2.293). Choosing the laser frequency close to the out-of-phase mode frequency $\omega_0 \approx \omega_2$ and assuming $\Omega_{\text{odf}} \ll 2\omega_0$, as well as $\eta_0 \Omega_{\text{odf}} \ll |\omega_1 - \omega_0|$, we can neglect all terms except the one coupling the spin to
the out-of-phase mode in a rotating wave approximation. The Hamiltonian in Eq. (4.172) then becomes
\[
\hat{H}_{\text{int}} = \hbar \frac{\delta}{2} \sigma^z + \hbar \frac{\Omega_d}{2} \sigma^x + \left( \hbar i \eta \frac{\tilde{\Omega}_{\text{odf}}}{2} a_2^\dagger \sigma^z e^{i\delta_n^\prime} + \text{H.c.} \right),
\]
(4.174)
where we have introduced $\delta_m = \omega_2 - \omega_\kappa$ which satisfies $\delta_m \ll \omega_2$. Finally, by moving to yet another interaction picture with respect to $\tilde{\delta} \equiv \eta \frac{\tilde{\Omega}_{\text{odf}}}{2} a_2^\dagger \sigma^z$, we can transform $\hat{H}_{\text{int}}$ to the time-independent Hamiltonian
\[
\hat{H}_{\text{int}} = \hbar \frac{\delta}{2} \sigma^z + \hbar \frac{\Omega_d}{2} \sigma^x + \left( \hbar i \eta \frac{\tilde{\Omega}_{\text{odf}}}{2} a_2^\dagger \sigma^z + \text{H.c.} \right) + \hbar \delta_m a_2^\dagger a_2.
\]
(4.176)
If we now identify
\[
\delta = \epsilon / \hbar,
\]
(4.177)
\[
\Delta = -\Omega_d,
\]
(4.178)
\[
\lambda = -i \eta \frac{\tilde{\Omega}_{\text{odf}}}{2} = -i \eta \frac{\tilde{\Omega}_{\text{odf}}}{2} e^{i(k_0 + \phi_0)},
\]
(4.179)
\[
\omega_m = \delta_m = \omega_2 - \omega_\kappa,
\]
(4.180)
$\hat{H}_{\text{int}}$ can be written as
\[
\hat{H}_{\text{sb,1}} = \frac{\epsilon}{2} \sigma^z - \frac{\hbar \Delta}{2} \sigma^x - \frac{\hbar \lambda}{2} \sigma^z (a + a^\dagger) + \hbar \omega_m a^\dagger a,
\]
(4.181)
where we have omitted the index of the creation and annihilation operators and have taken $\lambda$ to be real. We note in passing that in this setting $\lambda$ may always be taken to be real. This is the spin-boson Hamiltonian in Eq. (4.154) for a single mode.

Let us remark that the mode frequency in our simulation $\omega_m = \delta_m$ is given by the detuning of the spin dependent force from the driven mode $\omega_m = \omega_2 - \omega_\kappa$. For the simulation to be physically meaningful we need $\delta_m > 0$. Hence, the spin-dependent force needs to be red-detuned.

The last step is to add the damping on the mode due to the cooling of the $^{24}\text{Mg}^+$ ion. Cooling is described by the dissipator in Eq. (2.366). The full system then evolves according to
\[
\rho = -\frac{i}{\hbar} [\hat{H}_{\text{sb,1}}, \rho] + \mathcal{D}_{\kappa, \eta} \rho,
\]
(4.182)
with
\[
\mathcal{D}_{\kappa, \eta} \rho = \kappa (\eta + 1) [\sigma a^\dagger - a^\dagger \sigma \rho] + \kappa \eta [\sigma a^\dagger \rho a - a a^\dagger \rho] + \text{H.c.}
\]
(4.183)
Note that the cooling rate on the mode is reduced by a factor $\tilde{B}_{12}^2$ due to Eq. (2.391).

According to our considerations of the previous paragraph the system evolving according to Eq. (4.182) should produce the same spin dynamics as a spin coupled to a macroscopic oscillator bath with spectral density $J_{\text{eff}}(\omega)$, Eq. (4.161), if the conditions $\kappa \ll \omega_m$ and $\hbar \beta \kappa \ll 1$ are met. Note that in the ion-trap implementation the effective mode frequency is the detuning $\delta_m$ of the state dependent force and is therefore much smaller than the physical mode frequency. One has to make sure that the above conditions are met for the effective frequency so that the correspondence to the effective macroscopic harmonic environment holds for the simulation. For the values of $\omega_m$, $\kappa$ and $\eta (\omega_m)$ or equivalently $\hbar \beta$ reported in Tab. 4.1, we obtained excellent agreement between the reservoir correlation functions of the oscillator damped by an Ohmic bath and that of the Lindblad oscillator. Hence, we also consider these parameters when we benchmark the effective model in Eq. (4.182) with a full many body simulation of the spin-boson model.
Table 4.1.: Simulation parameters. The table lists the parameters used in the numerical simulations of the spin-boson model comparing the effective theory in Eq. (4.182) and a full many body simulation using the TEDOPA algorithm. The same parameters are also used for the simulation of the proposed ion trap experiments. The values of $\Delta$ and $\lambda$ are not reported because they were varied in the simulations.

<table>
<thead>
<tr>
<th>$\omega_m/2\pi$</th>
<th>$\kappa/2\pi$</th>
<th>$\bar{n}(\omega_m)$</th>
<th>$\bar{\hbar}\beta$</th>
<th>$\epsilon/2\pi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>100kHz</td>
<td>1.25kHz</td>
<td>0.025</td>
<td>5.91 $\cdot$ 10^{-6}s</td>
<td>0kHz</td>
</tr>
</tbody>
</table>

In order to underpin the considerations of the previous section, we simulated the dynamics of $\langle \sigma^z(t) \rangle$ and $\langle \sigma^x(t) \rangle$ for the full spin-boson Hamiltonian of Eq. (4.154) with spectral density $J_{\text{eff}}(\omega)$, Eq. (4.161), using the numerically exact TEDOPA algorithm [133, 134] and compared them with the dynamics generated by Eq. (4.182) for two sets of parameters. A brief account of the idea behind the TEDOPA algorithm [133, 134] is given in App. C. Let us remark that the results were produced together with Dario Tamascelli who conducted the TEDOPA simulations.

For the simulations we considered an initial product state $|e\rangle \langle e| \otimes \rho_0$ with the environment in a thermal state at inverse temperature $\bar{\hbar}\beta = 5.91 \cdot 10^{-6}$ s. We considered an unbiased system, i.e. $\epsilon = 0$, and chose a spin-motion coupling $\lambda/2\pi = 100$ kHz. The remaining simulation parameters are summarized in Tab. 4.1. Note that we have not specified the tunnel element $\Delta$ so far because it was varied in the simulations.

We simulated the time evolution for spin energies $\Delta/2\pi = -50$ kHz and $-100$ kHz. Smaller values of $\Delta$ would have led to excessive computation times for the TEDOPA simulations. One simulation for $\Delta/2\pi = -50$ kHz took 15 days using 16 cores on a computing cluster, which once more indicates the value of a trapped-ion simulator of spin-boson models.

The results are shown in Fig. 4.6. For both values of $\Delta$ we obtain very good agreement between the TEDOPA results and Eq. (4.182), which confirms our considerations that the Lindblad oscillator can be identified with a macroscopic environment with spectral density $J_{\text{eff}}(\omega)$ of Eq. (4.161) for appropriate parameters. In particular, the results show that this correspondence holds in a non-perturbative regime and that it also holds when we probe the spectral density away from the resonance.

With the results shown in Fig. 4.6 we can be confident that the proposed simulation protocol works. Moreover, we can also explore parameter regimes which are hard to tackle numerically with the TEDOPA algorithm. In the remainder of this section we present simulations of the dynamics of ion trap experiments using Eq. (4.182) for realistic parameters. Among these there will be simulations entering this parameter regime.

We considered a trap with an axial potential that yields a center-of-mass frequency $\omega_{\text{com}}/2\pi = 2.54$ MHz for a single $^{24}\text{Mg}^+$ ion. For the $^{24}\text{Mg}^+ - ^{25}\text{Mg}^+$ crystal the normal modes amplitudes in mass-weighted coordinates $\tilde{B}$ read

$$\tilde{B} = \begin{pmatrix} 0.6925 & -0.7214 \\ 0.7214 & 0.6925 \end{pmatrix}$$  (4.184)

yielding the real space amplitudes

$$B = \begin{pmatrix} 0.6999 & -0.7284 \\ 0.7143 & 0.6851 \end{pmatrix}.$$  (4.185)

As expected the amplitudes do not differ too much from those of homogeneous crystal, see Eqs. (2.24), (2.46) and (2.47). For the considered trap potential the in- and out-of-phase mode frequencies become

$$\omega_1/2\pi = 2.51 \text{ MHz} \quad \text{and} \quad \omega_2/2\pi = 4.36 \text{ MHz},$$  (4.186)
4. Simulations of the dynamics of spin-boson models with trapped ions

Figure 4.6.: Benchmark of the spin-boson simulation protocol. Part a) shows the time evolution of $\langle \sigma_z(t) \rangle$ in natural time units $|\Delta| \cdot t$. The solid lines show the time evolution under the full spin-boson Hamiltonian in Eq. (4.154) with spectral density $J_{\text{eff}}(\omega)$ from Eq. (4.161). We consider an initial product state $|e\rangle\langle e| \otimes \rho_\beta$ with the environment in a thermal state at inverse temperature $\bar{\hbar} \beta = 5.91 \cdot 10^{-6}$ s. The spin-motion coupling was $\lambda/2\pi = 100$ kHz and the spin energies were $\Delta/2\pi = -50$ kHz and $\Delta/2\pi = -100$ kHz. The remaining simulation parameters are summarized Tab. 4.1.

b) Same as a) for $\langle \sigma_x(t) \rangle$. which confirms our considerations before Eq. (4.174). The normal mode frequencies together with Eq. (4.184) yield the Lamb-Dicke parameters

$$\eta = \begin{pmatrix} 0.2012 & -0.1592 \\ 0.2053 & 0.1497 \end{pmatrix},$$

(4.187)

where we assumed that the lasers producing the state dependent force are at right angles and the effective wave vector is directed along the trap axis.

At the beginning of this section we presented a derivation of the Hamiltonian in Eq. (4.181) for the $^{25}$Mg$^+$ ion which encodes the spin. We assumed that the $^{25}$Mg$^+$ ion is sympathetically cooled by EIT cooling on the $^{24}$Mg$^+$ ion leading to the evolution in Eq. (4.182) of the whole system. $^{24}$Mg$^+$ has already been used for sympathetic cooling of mixed-species ion crystals [91] and the cooling rate and asymptotic population of the cooled mode given in Tab. Eq. (4.1) seem realistic in light of the results of [91]. We chose the microwave to be resonant such that $\varepsilon = 0$ for the Hamiltonian in Eq. (4.181) and considered an initial product state of spin and mode

$$\rho_0 = |e\rangle\langle e| \otimes \rho_\beta,$$

(4.188)

where the thermal state $\rho_\beta$ of the environment has a mean occupation number $\bar{n}(\omega_m) = 0.025$ as in Tab. 4.1. In all simulations of Eq. (4.182) that are presented here, the motional Hilbert space was truncated at $n_{\text{max}} = 15$ excitations to make truncation errors negligible.

The results of the simulations for different spin energies and spin-motion couplings $\lambda$ are depicted in Fig. 4.7. In part a) the figure shows the dynamics of $\langle \sigma_z(t) \rangle$ for a spin energy $\Delta/2\pi = 3$ kHz and varying spin-motion coupling $\lambda/2\pi = 10 - 200$ kHz. In this case, the spin samples the low frequency part of the spectral density. In the limit $\omega \ll \omega_m$ we can approximate $J_{\text{eff}}$ from Eq. (4.161) as

$$J_{\text{eff}}(\omega) \approx \frac{4\lambda^2 \kappa \omega_m}{(\kappa^2 + \omega_m^2)^2} \omega.$$

(4.189)
4.6. Experimental simulations of the spin-boson model using trapped ions

\[ \langle \sigma_z(t) \rangle \]

\[ \lambda/2\pi \text{[kHz]} = 10, 100, 200 \]

\[ \Delta \cdot t \]

\[ \langle \sigma_z(t) \rangle \]

Figure 4.7.: The figure depicts the dynamics of \( \langle \sigma_z(t) \rangle \) under Eq. (4.182) for spin energies \( \Delta/2\pi = 3\text{kHz} [\text{a}] \) and \( \Delta/2\pi = 100\text{kHz} [\text{b}] \) for varying spin-motion coupling. The time axis is given in natural time units \( \Delta \cdot t \). The remaining parameters can be found in the text. In part \( \text{a} \) \( \Delta/2\pi = 3\text{kHz} \) is much smaller than the mode frequency \( \omega_m/2\pi = 100\text{kHz} \) so that the environment is approximately Ohmic. In part \( \text{b} \) the spin is resonant with the mode frequency.

Thus, for small frequencies the spectral density is approximately Ohmic and we expect that the dynamics of \( \langle \sigma_z(t) \rangle \) are similar to that of a spin coupled to an environment with Ohmic spectral density. We observe a transition from underdamped to overdamped oscillations in \( \langle \sigma_z(t) \rangle \) when the spin-motion coupling is increased. This is indeed expected for a spin coupled to an Ohmic bath at finite temperatures \([127, 128]\). We should note, however, that \( J_{\text{eff}}(\omega) \) yields a different reservoir correlation function than the Ohmic environment and accordingly, we can only expect qualitatively similar dynamics of the spin in the low frequency regime where the spectral density is approximately Ohmic \([155, 156]\). Note that we could not enter this regime of the dynamics with the TEDOPA simulations because the time scales of the considered time evolution, which are determined by \( \Delta/2\pi = 3\text{kHz} \) in this case, would have led to excessive simulation times, see also App. C.

In panel \( \text{b} \) we show \( \langle \sigma_z(t) \rangle \) for \( \Delta/2\pi = 100\text{kHz} \) and the remaining parameters as in \( \text{a} \). In this case the spin is resonant with the mode and we observe a very rich behavior in the dynamics of \( \langle \sigma_z(t) \rangle \). While for the weakest spin-motion coupling \( \langle \sigma_z(t) \rangle \) shows damped oscillations, we can observe a partial revival of the spin-excitation in the case \( \lambda/2\pi = 50\text{kHz} \). An excitation leaving into the environment and coming back to the system is actually a paradigmatic example for non-Markovian behavior of an environment. Motivated by this observation we actually quantified the degree of non-Markovianity of the dynamics and found that the dynamics is non-Markovian for both considered values of \( \Delta \) for all finite values of \( \lambda \). The results are presented in detail in Section 4.8.

In summary, in this section we have shown that a damped oscillator in Lindblad description can indeed be identified with a macroscopic environment with Lorentzian spectral density. To this end we compared the dynamics of a spin coupled to a damped harmonic oscillator in Lindblad description with a numerically exact simulation of the full many body Hamiltonian and obtained excellent agreement. We have then further shown how more complex spectral densities can be tailored with combinations of Lindblad damped oscillators. Finally, we have illustrated that these ideas can be implemented with trapped ions deriving the relevant Hamiltonian and simulating an experiment with parameters realistic for a state-of-the-art experiment.
4.7. Impact of experimental sources of noise

Any experiment is inevitably subject to external influences which make the experimental system an open quantum system. Many times the external influences are weak and the dynamics is approximately still that of a closed quantum system. Eventually though, the environment will effect that the quantum system loses its quantum properties.

If we want to simulate the dynamics of a quantum system, we need to ensure that the simulator reproduces the dynamics of the system to simulate, also when subject to external sources of noise. We note that there are ideas such as quantum error correction that try to minimize or cancel the effects of the environment but they come at the cost of more experimental overhead [1]. In the present section we will investigate the impact of typical sources of noise in ion trap experiments on the quality of our simulation of spin-boson model dynamics. We will focus on two sources of noise here: The first is dephasing noise which can have different causes, such as fluctuating ambient magnetic fields or laser phase fluctuations [61] and is ubiquitous in ion trap experiments. The second source of noise is in a way specific to the implementation of the spin-boson model we propose. It is the decoherence that is caused implementing the spin-motion coupling in the $\sigma^z$-basis.

4.7.1. Dephasing noise

Encoding the qubit in the $|g\rangle \equiv |F = 3, m_F = 3\rangle$ and $|e\rangle \equiv |F = 2, m_F = 2\rangle$ states of $^{25}\text{Mg}^+$ permits to make use of the $\sigma^z$ spin-motion coupling but it has the disadvantage that the $|F = 3, m_F = 3\rangle \leftrightarrow |F = 2, m_F = 2\rangle$ transition is magnetic-field sensitive. This means that it is prone to dephasing due to fluctuations of the ambient magnetic field, see Section 2.3. We do not assume a particular noise model for the magnetic field fluctuations but rather take a phenomenological approach. We assume that the qubit levels are subject to a generic dephasing noise which also includes other possible sources of dephasing.

In order to capture the effects of the noise on the spin, we add a dissipator on the spin in Eq. (4.182) such that the state $\rho$ of spin and mode obeys the equation of motion

$$\dot{\rho} = -\frac{i}{\hbar}[H_{sb1}, \rho] + (\mathcal{D}_{K, \hbar} + \mathcal{D}_s)\rho, \quad (4.190)$$

where $H_{sb1}$ and $\mathcal{D}_{K, \hbar}$ have been introduced in Eqs. (4.181) and (4.183) above. The additional dissipator $\mathcal{D}_s$ accounting for dissipative effects on the spin for dephasing noise reads [160]

$$\mathcal{D}_s \rho = \frac{\Gamma}{2} (\sigma^z \rho \sigma^z - \rho). \quad (4.191)$$

This dissipator leads to a decay $e^{-\Gamma t}$ of the spin coherences where the decay constant $\Gamma$ is related to the qubit coherence time $T_{2}^*$ by

$$\Gamma = 1/T_{2}^*. \quad (4.192)$$

In order to investigate the effects of dephasing noise, we computed the dynamics of Eq. (4.190) using the parameters from Tab. 4.1 and the initial condition of Eq. (4.188) for different coherence times $T_{2}^*$ while the spin-motion coupling was varied in the range $\lambda/2\pi = 10 - 200\text{kHz}$. In Fig. 4.8 we present the resulting dynamics of $\langle \sigma^z(t) \rangle$ together with those given by Eq. (4.182) for the same parameters but without noise. The noise free dynamics are indicated by solid lines while symbols depict the noisy time evolutions. In panels a) and b) of Fig. 4.8 we show $\langle \sigma^z(t) \rangle$ for a spin energy $\Delta/2\pi = 3\text{kHz}$, where the environment appears approximately Ohmic, for qubit coherence times $T_{2}^* = 1\text{ms}$ and $T_{2}^* = 10\text{ms}$. We observe that for $T_{2}^* = 10\text{ms}$ there is already no visible effect of the dephasing on the dynamics. Let us note that the coherence times we...
4.7. Impact of experimental sources of noise

Considered here are a rather pessimistic estimate and have already been surpassed even with magnetic-field sensitive trapped-ion qubits, e.g. in [161] a $1/\sqrt{e}$ decay time of 300 ms has been observed on a magnetic-field sensitive transition in $^{40}\text{Ca}^+$. In part c) of Fig. 4.8 we depict the dynamics for the resonant case $\Delta/2\pi = 100$ kHz and a coherence time $T_2^* = 1$ ms. It can be appreciated in the figure that there is no visible effect on the dynamics already for this smaller qubit coherence time. Of course, this is because of the much shorter time scale of the time evolution as compared to the dephasing time in this case.

In the light of the above results it seems fair to neglect dephasing noise.

4.7.2. Decoherence due to $\sigma^z$ spin-motion coupling

The second noise mechanism that we consider is the decoherence introduced by off-resonant excitations out of the qubit manifold due to the spin-motion coupling. Here, we consider a simplified level scheme for $^{25}\text{Mg}^+$. We will consider a three-level $\Lambda$ system as we considered in Section 2.5. In practice, there are actually different $P$ levels contributing to the optical dipole forces in $^{25}\text{Mg}^+$. However, our simplified treatment still captures the essential effects.

In Section 2.5 we have seen that the off-resonant coupling of the qubit levels to a decaying excited state leads to dissipative effects. For the considered model these are given in Eqs. (2.301) and (2.302) in a rotating wave approximation. The cause of the dissipative effects is residual off-resonant excitation of the upper level, due to the finite detuning $\Delta_R$ of the laser beams, followed by an emission event. The dissipator in Eq. (2.301) describes Rayleigh scattering where the spin state is not changed upon a scattering event but the state can acquire an unknown phase leading to dephasing. In the Raman scattering processes described by Eq. (2.302) the spin states is changed upon photon scattering. Both processes lead to decoherence and are therefore harmful to the simulation.

Due to the results of the previous paragraph, we do not consider additional dephasing noise here. Including the noise effects, the spin and mode evolve according to Eq. (4.190). The spin dissipator now reads

$$D_s \rho = \sum_{j,k=e.g} \left( L_{jk} \rho L^\dagger_{jk} - \frac{1}{2} \{L^\dagger_{jk} L_{jk}, \rho \} \right),$$  

4.7. Impact of experimental sources of noise

Figure 4.8.: Impact of dephasing on the spin-boson model simulations with trapped ions. The figure shows the dynamics of $\langle \sigma^z(t) \rangle$. Solid lines depict the noise free dynamics generated by Eq. (4.182) and symbols those given by Eq. (4.191) that incorporates dephasing noise. The simulation parameters, except for the spin-motion coupling $\lambda$, are given in Tab. 4.1. The values of $\lambda/2\pi$ in kHz are given in the legends. In parts a) and b) we consider the quasi Ohmic case with spin energy $\Delta/2\pi = 3$ kHz and coherence times $T_2^* = 1$ ms and $T_2^* = 10$ ms, respectively. Part c) shows the dynamics for the resonant case $\Delta/2\pi = 100$ kHz and $T_2^* = 1$ ms.
4. Simulations of the dynamics of spin-boson models with trapped ions

The numerical results of this section show that our simulation protocol is robust to the most common sources of noise in ion traps. The quality of the simulations is largely unaffected by typical dephasing rates while decoherence due to the $\sigma^z$ spin-motion coupling can be suppressed by choosing large enough Raman detunings. Note that this source of decoherence can also be overcome by working in a rotated spin-basis and using a Mølmer-Sørensen type spin-motion coupling [124].
4.8. Quantification of the degree of non-Markovianity of the spin-boson dynamics

Upon inspecting the dynamics in panel b) of Fig. 4.7 we conjectured that the spin dynamics is non-Markovian when the spin is on resonance with the damped mode constituting the environment. Although the concept of non-Markovianity is not new [162], the definition of quantitative measures of non-Markovianity in the quantum realm is quite a recent topic and there is no unique quantitative measure of non-Markovianity. Several proposals for measures of non-Markovianity exist, see e.g. [77, 163], and we will quantify the non-Markovian character of the time evolution in the spin-boson model employing two of these measures.

We start this section by reviewing the definitions of the two relevant measures of non-Markovianity, \( \mathcal{N}_{\text{RHP}} \) and \( \mathcal{N}_{\text{BLP}} \). Our treatment is based on those in [77, 163]. Thereafter, we present our results on the non-Markovian character of the time evolution of the spin-boson model dynamics presented in Section 4.6 as quantified by these two measures.

4.8.1. The RHP measure of non-Markovianity

To begin our considerations we briefly sketch why there is no straightforward generalization of classical Markov processes to quantum Markov processes. A classical stochastic process \( X(t), t > 0 \), can be understood as a random variable whose distribution depends on time parametrically. The process has to fulfill certain mathematical properties which we will not discuss here, see e.g. [69]. We only note that a stochastic process that may assume discrete values of some set \( \{x_i\} \) gives rise to a hierarchy of joint probability distributions \( P_n(x_n, t_n; \ldots; x_1, t_1), n \in \mathbb{N} \) [69]. This expression gives the probability that \( X(t) \) assumes the value \( x_1 \) at time \( t_1 \), \( x_2 \) at time \( t_2 \) and so on. For a Markovian process, the conditional probability

\[
P(x_{n+1}, t_{n+1} | x_n, t_n; \ldots; x_1, t_1) = \frac{P(x_{n+1}, t_{n+1}; x_n, t_n; \ldots; x_1, t_1)}{P(x_n, t_n; \ldots; x_1, t_1)}
\] (4.194)

satisfies the Markov condition

\[
P(x_{n+1}, t_{n+1} | x_n, t_n; \ldots; x_1, t_1) = P(x_{n+1}, t_{n+1} | x_n, t_n).
\] (4.195)

Thus, for a Markov process the probability that the process assumes the value \( x_{n+1} \) at time \( t_{n+1} \), under the condition that it assumed the values \( x_i \) at times \( t_i \), in fact only depends on the probability that the process assumed values \( x_n \) at time \( t_n \). In this sense, Markov processes are said to be memoryless since the probability to assume the value \( x_{n+1} \) at time \( t_{n+1} \) only depends on the probability to assume the values \( x_n \) at time \( t_n \) and all previous times do not play a role.

Let us now consider a quantum system that evolves unitarily such that we may write the time evolution as \( \rho(t_2) = G(t_2, t_1)\rho(t_1) = U(t_2, t_1)\rho(t_1)U^T(t_2, t_1), \) where \( U(t_2, t_1) \) is the unitary time evolution operator and \( G(t_2, t_1) \) a shorthand notation for the propagator. Let us consider a non-degenerate observable \( O = \sum_x \langle \psi_x | \psi_x \rangle \) on the Hilbert space of the open system that is projectively measured at times \( t_i \). With the projector to the state corresponding to measurement outcome \( x \), \( P_x \rho = |\psi_x\rangle \langle \psi_x| \rho |\psi_x\rangle \langle \psi_x| \), we may write the joint probability to measure \( x_n \) at time \( t_n \), \( x_{n-1} \) at time \( t_{n-1} \) and so on as

\[
P_n(x_n, t_n; \ldots; x_1, t_1) = \text{Tr}[P_n G(t_n, t_{n-1}) \ldots P_1 G(t_1, t_0) \rho(t_0)].
\] (4.196)

Obviously, in general the joint probability above does not satisfy the Markov condition in Eq. (4.195). The projective measurements alter the state of the quantum system and thus influence its future evolution and measurement outcomes. Also, a different measurement process...
can lead to a different result. Therefore, non-Markovianity should be formulated in terms of
the dynamical map describing the time evolution of a system rather than joint probabilities of
measurement outcomes.

The first measure of non-Markovianity we want to consider is $\mathcal{N}_{\text{RHP}}$ where “RHP” stands
for “Rivas-Huelga-Plenio”, who introduced the measure. We present the definition that can be
found in [163]. Let us consider an open system of finite dimension $d$ that is initially in a state
$\rho_0(t_0)$ and whose time evolution to a later time $t \geq t_0$ is given by the dynamical map $\mathcal{E}(t,t_0)$

$$\rho_s(t) = \mathcal{E}(t,t_0)\rho_s(t_0).$$

(4.197)

For a physical time evolution we demand that for every $t \geq t_0$ $\mathcal{E}(t,t_0)$ is a completely positive
and trace preserving map (see Section 2.4.1). Let us now consider that the time evolution is split
up in two parts: an evolution from $t_0$ to an intermediate time $t_1$ and from $t_1$ to the final time $t$
such that the full time evolution reads [163]

$$\mathcal{E}(t,t_0) = \mathcal{E}(t,t_1)\mathcal{E}(t_1,t_0).$$

(4.198)

Assuming that the inverse of $\mathcal{E}(t_1,t_0)$ exists, we obtain the time evolution from $t_1$ to $t$

$$\mathcal{E}(t,t_1) = \mathcal{E}(t,t_0)\mathcal{E}^{-1}(t_1,t_0).$$

(4.199)

Now Ref. [163] defines non-Markovianity in the following way: the family of dynamical maps
$\{\mathcal{E}(t,t_0)|t \geq t_0\}$ describes a Markovian time evolution if and only if the map $\mathcal{E}(t,t_1)$ exists and
is completely positive for all $t \geq t_1 \geq t_0$. If a map satisfies Eq. (4.199) for all times and $\mathcal{E}(t,t_1)$
is completely positive, the map is also said to be CP divisible. Hence, Markovianity is identified
with CP divisibility of the dynamical map. The degree of non-Markovianity $\mathcal{N}_{\text{RHP}}$ of a given
time evolution over a time interval $I = [t_0,t_2]$ is quantified by measuring the departure of the
maps $\mathcal{E}(t_2,t_1)$, $t_2 \geq t_1 \geq t_0$, from complete positivity over that interval.

In order to quantify this departure, [163] considers the so-called Choi matrix of the dynamical
map $\mathcal{E}(t + \varepsilon, t)$

$$[\mathcal{E}(t + \varepsilon, t) \otimes \mathbb{1}_d]|\psi\rangle \langle \psi|,$$

(4.200)

where $\varepsilon > 0$. Here, $|\psi\rangle = \frac{1}{\sqrt{d}} \sum_{n=1}^{d} |n,n\rangle$ is a maximally entangled state of the open system
with an ancillary system of the same dimension. The Choi matrix is positive semidefinite if and only
if $\mathcal{E}(t + \varepsilon, t)$ is completely positive [164]. Moreover, $\mathcal{E}(t + \varepsilon, t)$ is trace preserving since it is a
composition of trace preserving maps, see Eq. (4.199). Therefore, the trace norm of the Choi matrix

$$||(\mathcal{E}(t + \varepsilon, t) \otimes \mathbb{1}_d)|\psi\rangle \langle \psi||_1$$

(4.201)

can be used to quantify deviations from complete positivity. For a Markovian process it will be
one, since $\mathcal{E}(t + \varepsilon, t)$ is completely positive and trace preserving for all $t$ and $|\psi\rangle \langle \psi|$ has a unit
trace. If one the other hand $\mathcal{E}(t + \varepsilon, t)$ is not completely positive, the map $\mathcal{E}(t + \varepsilon, t) \otimes \mathbb{1}_d$ is still
trace preserving but the Choi matrix has negative eigenvalues leading to a trace norm larger than
one.

The degree of non-Markovianity over an interval $I$ is then defined as [163]

$$\mathcal{N}_{\text{RHP}} = \frac{\int_{I, g > 0} \bar{g}(t) \, dt}{\int_{I, g > 0} \chi[\bar{g}(t)] \, dt},$$

(4.202)

where the integral extends over those intervals in $I$ where $\bar{g}(t) > 0$ and

$$\chi[x] = \begin{cases} 1 & \text{for } x > 0 \\ 0 & \text{else.} \end{cases}$$

(4.203)
4.8. Quantification of the degree of non-Markovianity of the spin-boson dynamics

By definition “0/0” = 0 in Eq. (4.202). The function \( g(t) \) is defined as
\[
g(t) = \lim_{\epsilon \to 0^+} \frac{\| [\mathcal{E}(t + \epsilon) \otimes \mathbb{I}_d] \psi \|_1 - 1}{\epsilon}.
\]
(4.204)

Note that, by construction, \( g(t) \) vanishes if and only if the dynamics is Markovian and is normalized to unity [163].

We evaluated \( \mathcal{N}_{\text{RHP}} \) numerically for the dynamics of the spin-boson dynamics presented in Fig. 4.7. The numerical evaluation is not completely straightforward and the details are given in App. D. The results for \( \mathcal{N}_{\text{RHP}} \) are shown in Fig. 4.10a. We considered the dynamics given by Eq. (4.182) with the parameters that were used in Fig. 4.7. These are reported in Tab. 4.1 while the initial condition is given in Eq. (4.188). We considered \( \Delta/2\pi = 3 \text{kHz} \) and \( \Delta/2\pi = 100 \text{kHz} \) and a spin-motion coupling in the range \( \lambda/2\pi = 0 - 200 \text{kHz} \). For \( \Delta/2\pi = 3 \text{kHz} \) we considered the final time \( t = 0.01/\Delta \) and \( N = 10^4 \) steps while for \( \Delta/2\pi = 100 \text{kHz} \) the final time was \( t = 0.1/\Delta \) also with \( N = 10^4 \) steps. One might be tempted to use a smaller time steps but this eventually leads to discontinuous behavior in \( \mathcal{N}_{\text{RHP}} \). The figure shows that the dynamics of the simulated spin-boson model is non-Markovian according to \( \mathcal{N}_{\text{RHP}} \) for all \( \lambda > 0 \).

4.8.2. The BLP measure of non-Markovianity

The second measure we consider is \( \mathcal{N}_{\text{BLP}} \) where “BLP” stands for “Breuer-Laine-Piilo”, who proposed the measure originally [165]. Here, we use the definition of \( \mathcal{N}_{\text{BLP}} \) presented in [77]. While \( \mathcal{N}_{\text{RHP}} \) has a well-motivated mathematical definition, it suffers from the fact that one would need to perform map tomography to obtain the measure experimentally. This is, of course, feasible but very time consuming. It is therefore desirable to quantify non-Markovianity by a more time efficient procedure. The measure \( \mathcal{N}_{\text{BLP}} \) involves a maximization over all possible initial states of the open quantum system and is therefore theoretically as hard to evaluate as the map tomography for \( \mathcal{N}_{\text{RHP}} \). However, with wisely chosen initial states one can provide a useful lower bound on \( \mathcal{N}_{\text{BLP}} \) that only demands state tomography of the time evolution of two states, which produces less experimental overhead than map tomography. Let us comment, though, that also state tomography becomes inefficient very quickly.

For the definition and interpretation of the measure of non-Markovianity \( \mathcal{N}_{\text{RHP}} \), we need two properties of the trace distance. The trace distance of two states \( \rho_1 \) and \( \rho_2 \) is defined as [1]
\[
D(\rho_1, \rho_2) = \frac{1}{2} \| \rho_1 - \rho_2 \|_1.
\]
(4.205)

The trace distance assumes values \( 0 \leq D(\rho_1, \rho_2) \leq 1 \). It is 1 if and only if two states are orthogonal while it is 0 if and only if two states are equal [77].

The first property of the trace distance that is employed for the interpretation of \( \mathcal{N}_{\text{RHP}} \) is that the trace distance can be interpreted as a measure of how distinguishable two states are. Consider that we obtain a system that was prepared in one of two states \( \rho_1 \) or \( \rho_2 \) with probability 1/2 each. Let us further consider that we should find out which of the two states the system is actually in with a single measurement. It turns out that applying the optimal measurement procedure the maximal probability of success to identify the right state is [77]
\[
P_{\text{max}} = \frac{1}{2} [1 + D(\rho_1, \rho_2)].
\]
(4.206)

Thus, the trace distance gives a bias in favor of the right decision and can be taken as a measure of how distinguishable the two states are. In fact, for orthogonal states \( \rho_1 \) and \( \rho_2 \) the trace distance \( D(\rho_1, \rho_2) = 1 \). Then, \( P_{\text{max}} = 1 \) and we can can distinguish the states with certainty, which is a well known fact for orthogonal states.
The second property of the trace distance important for us is that completely positive and trace preserving maps are contractions for the trace distance. For a completely positive and trace preserving map \( E(t, t_0) \) and states \( \rho_1 \) and \( \rho_2 \), we have [1]

\[
D(E(t, t_0)\rho_1, E(t, t_0)\rho_2) \leq D(\rho_1, \rho_2). \tag{4.207}
\]

The combination of properties (4.206) and (4.207) forms the basis for the definition of the measure of non-Markovianity \( \mathcal{M}_{\text{BLP}} \). With the interpretation of the trace distance as a measure of our ability to distinguish two states, Eq. (4.207) tells us that two initial states \( \rho_1(t_0) \) and \( \rho_2(t_0) \) of an open quantum system can only become less distinguishable under a completely positive and trace preserving dynamical map \( E(t, t_0) \)

\[
D(\rho_1(t), \rho_2(t)) \leq D(\rho_1(t_0), \rho_2(t_0)), \tag{4.208}
\]

where \( t \geq t_0 \) and \( \rho_i(t) = E(t, t_0)\rho_i(t_0) \). This can be interpreted as a loss of information about the state of the system into the environment [77]. On the other hand, an increase in the trace distance is interpreted as a flow of information from the environment back into the system. This leads to the definition of non-Markovianity \( \mathcal{M}_{\text{BLP}} \). A family of dynamical maps \( \{E(t, t_0)\} \) is called Markovian if \( D(\rho_1(t), \rho_2(t)) \) decreases monotonically for all times and all pairs of initial states. A non-Markovian dynamics is then defined as a family of dynamical maps \( \{E(t, t_0)\} \), where \( D(\rho_1(t), \rho_2(t)) \) is non-monotonic for some pair of initial states \( \rho_1(t_0) \) and \( \rho_2(t_0) \).

The definition of a measure of non-Markovianity over an interval \( I \) according to these considerations leads to [77]

\[
\mathcal{M}_{\text{BLP}} = \max_{\rho_1(t)/\rho_2(t)} \int_{I, \sigma > 0} \sigma(t) \, dt, \tag{4.209}
\]

where \( \sigma(t) = \frac{d}{dt} D(E(t, t_0)\rho_1, E(t, t_0)\rho_2) \) and the integral extends over those intervals in \( I \) where \( \sigma(t) > 0 \). It was shown in [166] that optimal initial state pairs \( \rho_1, \rho_2 \) that saturate the maximum in Eq. (4.209) are orthogonal and lie on the boundary of state space.

Let us note that the above definition can be generalized. If we have an initial pair of states \( \rho_1 \) and \( \rho_2 \) that are prepared with probabilities \( p_1 \) and \( p_2 \) that sum up to unity, the maximal probability to identify the prepared state in a single measurement is given by [77]

\[
P_{\text{max}} = \frac{1}{2} [1 + ||\Delta_H||_1], \tag{4.210}
\]

where \( \Delta_H = p_1\rho_1 - p_2\rho_2 \) is the so-called Helstrom matrix. Thus, for an equal preparation probability for the two states, the Helstrom matrix gives a bias in favor of distinguishing the two states. For \( p_1 = p_2 = 1/2 \) we recover Eq. (4.206). One can now define a generalized measure \( \mathcal{M}_{\text{BLP}} \), where \( \sigma(t) = \frac{d}{dt} ||\sigma(t)\rho_H||_1 \) in Eq. (4.209) and the maximization is also over the weights \( p_{1/2} \). This generalized definition of non-Markovianity corresponds to \( P \) divisibility of the dynamical map \( \sigma(t, t_0) \), i.e. \( \sigma(t, t_1) \) from Eq. (4.199) exists and is positive [77]. Let us note that the interpretation of non-Markovianity representing a flow of information from the environment to the system does not always hold strictly [163].

After this little detour we remark that our purpose is only to witness non-Markovian dynamics. Accordingly, we do not need to perform the maximization in Eq. (4.209). It suffices for us to find a pair of initial states that shows a non-monotonic behavior of the trace distance and we can take it as a lower bound for \( \mathcal{M}_{\text{BLP}} \). Due to the knowledge that optimal state pairs are orthogonal and lie on the boundary of state space, we can provide a useful lower bound on \( \mathcal{M}_{\text{BLP}} \) by computing the measure taking the states \( |\psi\rangle, |g\rangle, |\pm\rangle_x \) and \( |\pm\rangle_y \) as initial states. Here, \( |\pm\rangle_x \) and \( |\pm\rangle_y \) are the eigenstates of \( \sigma^x \) and \( \sigma^y \) corresponding to eigenvalues \( \pm 1 \).

For the numerical computation of \( \mathcal{M}_{\text{BLP}} \) we considered the whole interval \([0, 20/\Delta]\) of the dynamics shown in Fig 4.7 and took the same parameters as we used there. The parameters

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150
are summarized in Tab. 4.1. In addition to these parameters we considered \( \Delta/2\pi = 3\) kHz and 100 kHz and varied the spin-motion coupling \( \lambda/2\pi = 0 - 200\) kHz. We considered \( N = 10^4 \) equally spaced points \( t_i \) in the interval and computed the time evolution for the spin starting in each of the eigenstates of the Pauli matrices. From the time evolved states we computed a discrete version of \( N_{\text{RHP}} \) according to

\[
N_{\text{BLP}} = \sum_{i, D(t_{i+1}) - D(t_i) > 0} [D(t_{i+1}) - D(t_i)].
\]  

Here, the sum runs over those \( i \) where \( D(t_{i+1}) - D(t_i) > 0 \) and \( D(t_i) = D(\mathcal{E}(t_i, t_0) \rho_1, \mathcal{E}(t_i, t_0) \rho_2) \).

Note that we evaluated the measure only taking the eigenstates of the same Pauli matrix as an initial pair. We also note that Eq. (4.211) only yields an approximation to the true value of \( N_{\text{RHP}} \) due to the finite number of “measurements” [167].

The results of this computation are shown in part b) of Fig. 4.10. The displayed values for the “Ohmic” case \( \Delta/2\pi = 3\) kHz are obtained for the initial spin states \( \rho_s(0) = |\pm\rangle\langle|\pm|_x \) and in the resonant case \( \Delta/2\pi = 100\) kHz for the initial spin states \( \rho_s(0) = |\epsilon\rangle\langle\epsilon|, |g\rangle\langle g| \).

![Figure 4.10.: Degree of non-Markovianity of the spin-boson dynamics presented in Fig 4.7:
In panel a) we show the measure of non-Markovianity \( N_{\text{RHP}} \), Eq. (4.202), as a function of the spin-motion coupling \( \lambda \) in the intervals \([0.01/\Delta]\) and \([0.1/\Delta]\) for the “Ohmic” (\( \Delta/2\pi = 3\) kHz) and resonant (\( \Delta/2\pi = 100\) kHz) cases, respectively. The remaining simulation parameters are given in Tab. 4.1 and the initial condition in Eq. (4.188). Panel b) depicts a lower bound for the measure of non-Markovianity \( N_{\text{BLP}} \) from Eq. (4.209) for the same cases as in a). For \( N_{\text{BLP}} \) we considered the whole interval \([0, 20/\Delta]\).](image)

Fig. 4.10 shows that in both cases both measures \( N_{\text{RHP}} \) and \( N_{\text{BLP}} \) are non-zero for all couplings \( \lambda/2\pi > 0 \). Although \( N_{\text{RHP}} \) witnesses non-Markovianity for a smaller class of processes than \( N_{\text{BLP}} \) [77], both measures witness non-Markovianity in the same regions. The somewhat discontinuous behavior of \( N_{\text{BLP}} \) for the resonant case is due to the fact that \( N_{\text{BLP}} \) is not normalized and because we are only sampling a finite time interval.

### 4.9. Summary and Outlook

In this chapter, we have developed a scheme for a trapped-ion quantum simulator of the dynamics of spin-boson models and have illustrated the feasibility of our protocol by simulating a proof-of-principle experiment.

The theoretical basis for our protocol was provided by showing in a path integral approach that a damped harmonic oscillator in Lindblad description yields the same effective environment as an oscillator damped by a bath with Ohmic spectral density for appropriate parameters. This investigation was inspired by previous results that were proposing the use of damped oscillators...
4. Simulations of the dynamics of spin-boson models with trapped ions

in Lindblad description for the simulation of open quantum systems [150–153]. The resulting spectral density is a Lorentzian whose width is given by the cooling rate and its center by the frequency of the damped harmonic oscillator. The height is determined by the coupling of the spin to the damped mode.

We benchmarked this result by comparing the dynamics of a spin coupled to a damped harmonic oscillator in Lindblad description with a full many-body simulation of a spin coupled to a bath of harmonic oscillators with Lorentzian spectral density. We obtained excellent agreement between the dynamics of the two models both for a non-perturbative coupling between the spin and the mode and when probing the spectral density away from the resonance, which corroborates our analytical result. Furthermore, the set of parameters used for the simulations is realistic for a current ion trap experiment. In order to underpin the idea that the simulation of the dynamics of spin-boson models can be carried out in a real experiment, we took the most common experimental errors into account and showed that the dynamics and thus the simulation of spin-boson dynamics is robust against state-of-the art error rates.

The largest effects on the simulations were those of the decoherence due to the proposed spin-motion coupling in $\sigma^z$. If one cannot choose a detuning large enough to suppress photon scattering by the Raman lasers sufficiently, one could rotate the spin basis and provide spin-motion coupling in a different basis, e.g. $\sigma^x$, by a Mølmer-Sørensen interaction [124]. Then, one could also use clock states to encode the spin.

There are several ways to extend the simulations of the proposed experiment. The most straightforward of these is to include a finite bias $\varepsilon$ by introducing a detuning to the field driving the spin transition.

While the proposed proof-of-principle experiment easily enters regimes that are almost intractable with the full many-body simulations, for the effective model this is not so. The mapping of the spin-boson model with Lorentzian spectral density to a spin coupled to a damped oscillator in Lindblad description allows us to simulate the dynamics much more efficiently. Hence, if we use the additional knowledge that we have about the problem, we do not enter regimes that are hard to tackle on a classical computer. It is important to note, though, that the numerical simulation of the dynamics of a spin coupled to several Lindblad oscillators quickly becomes inefficient on a classical computer. Thus, the regime where the spectral density can be decomposed into, say, 5-10 Lorentzians would be very interesting, because here the numerical simulation of the full dynamics becomes difficult also for the effective model, especially when we are dealing with finite temperature. In this regime, one could obtain a quantum simulator of spin-boson dynamics that truly enters parameter regimes which are out of reach for classical simulation methods.

Due to the constraint that the cooling rate has to be considerably smaller than the effective mode frequency, it seems that the proposed method is better suited to model structured environments which have pronounced resonances. This may seem to restrict the applicability of a potential simulator, but structured environments are of interest because of two reasons. Firstly, the strong resonances are usually caused by long-lived vibrational modes [140] and these lead to long environmental memory times. As we stated previously, long environmental memory times usually limit numerical methods and therefore a quantum simulator could be helpful here. Second and more importantly, structured environments play a crucial role in the dynamics of photosynthetic pigment protein complexes [125]. Understanding the dynamics of these complexes might help to unravel the physical processes taking place in photosynthesis. Apart from more complex spectral densities than the one we considered here, models of photosynthetic pigment protein complexes typically involve several interacting spins. This makes the simulation of these models more demanding than the simulation of the simple model we considered here, but simulations of such models should in principle be possible.
In order to tailor more complex spectral densities than the one studied here, one needs to combine several damped modes each of which is described by three parameters. For several modes the optimization in Eq. (4.164), which is used to find the right coefficients to model a certain spectral density, eventually becomes quite a formidable task. This is not only because of the large number of parameters, but also because the three parameters of each mode are not completely independent. The cooling rate needs to be considerably smaller than the mode frequency for each mode. Finally, also the structure of the mode frequencies of the crystal has to be taken into account.

An interesting route to follow to solve the optimization is probabilistic programming that performs Bayesian inference using Markov chain Monte Carlo methods \([168]\). In Fig. 4.11 we show fits of ten Lorentzians to an Ohmic spectral density \(J_\text{O}(\omega) = \omega e^{-\omega/\omega_c}\) with a cutoff \(\omega_c/2\pi = 50\text{kHz}\). The fits were obtained with the Python library PyMC3. The Lorentzians are subject to the condition that their width is at maximum one tenth \([a])\) or one fifth \([c])\) of their central frequency. In part \(b)\) we show the Lorentzians constituting the effective spectral density of part \(a)\). Although the effective spectral density in part \(a)\) is a bit ragged, it captures the structure of the target spectral density. In part \(c)\) we can appreciate that to simulate unstructured environments, it is desirable to have broader Lorentzians available. While the fit to the spectral density is poor for small frequencies in both cases, it is reasonable above 50kHz. In part \(c)\) the agreement is actually very good. This example shows that simulating unstructured environments is not completely hopeless if several modes are available. And, secondly, it shows that the method can be useful if one has the a priori knowledge that the dynamics of an open system is determined only by a certain part of the spectral density. Then, the method could be used to reproduce this specific part of a spectral density.

Let us now turn to a few practical aspects. In order to simulate more complex spectral densities, the spin needs to be coupled to more modes of motion. This can be realized by either coupling the spin to the modes of different spatial directions or embedding more ions into the crystal and coupling the spin to several modes of the same direction. Note that it is advantageous to add cooling and not spin ions if we only consider one spin, because then cooling becomes more efficient. Depending on the structure of the spectral density to be simulated, it might be more advantageous to work with either radial or axial modes of motion.

The radial modes are more closely spaced in frequency and thus coupling to as well as cooling several modes at the same time is easier. However, addressing several modes with the same laser, due to the condition that the cooling rates need to be much smaller than the mode frequencies one will obtain gaps in the spectral density using only one phonon branch. Let us recall that this restriction arises because if the cooling rates are larger than the spacing between normal modes,
the cooling can introduce correlations between the modes. Then, the modes cannot be considered independent anymore, which is an essential assumption for our proposal. Furthermore, very strong cooling on one ion can lead to a decoupling of the ion from the rest of the crystal. Then, cooling has to be described locally and the description of cooling in terms of independent normal modes is not valid anymore. Coming back to the problem of tailoring a spectral density with radial modes: If the gaps in the effective spectral density are undesired, one needs the phonons of a second direction to fill them. Hence, in order to form a continuous spectral density involving overlapping Lorentzians, one would need to use the normal modes of at least two spatial directions.

Another option is to use the axial modes which have a larger bandwidth. Here, one could address several modes individually using different laser frequencies such that the effective mode frequencies are independent. This approach requires that the off-resonant coupling to the other modes is negligible, which puts an upper bound on the coupling strength to the individual modes. This idea is theoretically also possible for radial modes but harder to implement due to the smaller bandwidth of the radial phonon frequencies. Finally, which axis is to be preferred, depends on the distribution of the central frequencies, cooling rates and couplings that need to be realized.

Before we close, let us come back to the simulation of the dynamics of photosynthetic pigment protein complexes, which is one of the most exciting perspectives for the simulation of spin-boson models with trapped ions. First steps into this direction have already been taken [169]. In order to implement models of photosynthetic pigment protein complexes, we need to incorporate several interacting spin ions into the crystal. Interactions between the internal levels of different ions are usually realized by coupling them through the phonons of one spatial direction. Hence, for the simulation of such models the modes of one direction are reserved for the spin-spin interactions. The normal modes of the two remaining spatial directions can then be used to engineer the reservoir(s) in the model. Local reservoirs, as they are typically assumed in these models, could be realized by coupling each spin ion only to one or a few modes.

In this context, we can also connect the results of this chapter to those of Chapter 3. One could use trapped ions to simulate higher order spectral responses of the photosynthetic pigment protein complexes, as for example 2D electronic spectroscopy [101]. This is interesting because 2D spectroscopy is one of the standard interrogation methods for the dynamics in these complexes and the spectra are extremely hard to compute numerically, even for a few electronic sites coupled to an environment with structured spectral density [126]. While the simulation of the dynamics or 2D spectra of photosynthetic pigment protein complexes would generate formidable experimental effort, it is a very exciting future perspective. Trapped ions could then contribute to the understanding of the physical mechanisms participating in photosynthesis.

In summary, in this chapter we have presented a method for the simulation of spin-boson models with trapped ions and illustrated the procedure by simulating a proof-of-principle experiment. The model can be extended to almost arbitrary spectral densities and can, in principle, be extended to simulate the dynamics of interacting multi-spin-boson models. More generally, the simulation method offers exciting perspectives for the simulation of so far unexplored parameter regimes of open quantum systems with trapped ions.
5. Summary and outlook

The increasing complexity of trapped ion experiments demands the design of new protocols for the interrogation of ion Coulomb crystals so that relevant information can be extracted efficiently from the ever more complex experiments. The first part of this thesis addressed this issue. We developed a protocol for the investigation of nonlinear dynamics in crystals of trapped ions. Nonlinear dynamics play a role in different realms of trapped ion experiments. On one hand, it has been proposed to add nonlinear potentials to ion traps for the purpose of simulating models of friction [14–16] or many-body models, such as the Bose-Hubbard model [33]. On the other hand, nonlinear dynamics are intrinsic to ion Coulomb crystals because the Coulomb interaction between ions is itself nonlinear. Usually, the intrinsic nonlinearities are negligible. However, they can become important for the dynamics of ion crystals, for example in the vicinity of the linear-to-zigzag transition [32]. If not taken care of, they can even act as a source of noise in quantum information experiments [113]. In this work, we have focused on the intrinsic nonlinearities of ion Coulomb crystals.

We have adapted two-dimensional (2D) spectroscopy, which has been very successfully applied in other fields of physics such as nuclear magnetic resonance (NMR) [30] or the investigation of electronic dynamics in molecules [31], to trapped ions. We devised a two-dimensional spectroscopy protocol for the investigation of nonlinear dynamics of crystals of trapped ions. We showed that the protocol can be applied in state-of-the-art trapped ion setups by simulating two experiments with realistic parameters. In the simulated experiments, we considered regimes where resonant contributions from the third- and fourth-order expansion of the Coulomb potential contribute to the dynamics and then showed how these resonant effects can be detected with 2D spectroscopy.

In the first simulated experiment, we showed how 2D spectroscopy can be used to detect resonant interactions due to fourth-order contributions of the Coulomb potential that emerge in the vicinity of the linear-to-zigzag transition. In the second simulated experiment, we showed how 2D spectroscopy can detect energy transfer between normal modes due to a resonance in the third-order expansion of the Coulomb potential.

The protocol we have developed has several assets. We showed that it can reveal information which is not accessible in a conventional 1D experiment. Furthermore, for suitably chosen parameters there are no harmonic contributions to the spectrum, which are usually much stronger than the nonlinear contributions, and hence nonlinear effects are easier to identify. Finally, we showed that the protocol is robust with respect to the most common sources of experimental noise and that we can identify different noise mechanisms in the 2D spectra.

The future perspectives of two-dimensional spectroscopy with trapped ions are promising. The technique could be used to investigate whether nonlinear interactions play a role in the dynamics of large 2D or 3D crystals. Two-dimensional spectroscopy could also be adapted to investigate spin-spin interactions in larger ion crystals, an application very similar to NMR where the technique was developed.

In the second part of the thesis, we devised a protocol for the quantum simulation of the dynamics of open quantum systems. As a consequence of the increased level of control and the larger amount of tools available in trapped ion experiments, the goals of experiments have be-
come more and more ambitious. Recent quantum simulation experiments aim to access parameter regimes and particle numbers which cannot be simulated on classical computers. Especially the simulation of spin models has received a lot of attention. Experiments in linear traps probing spin models have increased the number of spins in the experiments from just two [19] to the order of ten ions [20, 21] and recently experiments with up to 53 ions have been reported [23]. The simulation of the dynamics of open quantum systems, however, has received much less attention.

This is rather surprising since the simulation of open quantum systems is of great practical relevance and offers the potential to reach regimes which are intractable on a classical computer. Especially when the environments are strongly coupled to the principal system or when they have long memory times, classical simulations are limited.

In this work, we devised a protocol for the simulation of spin-boson models with trapped ions. Spin-boson models are archetypical models of an open quantum systems. In these models a single spin is coupled to an infinite oscillator environment and one is interested in finding the reduced dynamics of the spin. Although the model is seemingly simple, in general one cannot find a closed solution for the dynamics of the spin. Since the model finds numerous applications, for example in condensed matter physics [127], chemical reactions [129] or the modeling of photosynthetic pigment protein complexes [125], it has been investigated by a plethora of analytical and numerical methods. Still, many regions of parameter space remain unexplored, and hence spin-boson models lend themselves as a candidate for quantum simulation.

The protocol for the simulation of spin-boson models that we devised makes use of the knowledge that the influence of thermal oscillator environments on the spin is characterized by a single scalar function, the spectral density. For macroscopic environments, the spectral density is usually taken to be a continuous function of frequency. Thus, the question arises how one can simulate macroscopic environments incorporating infinitely many harmonic oscillators using only a finite number of ions.

It is known that a harmonic oscillator which itself is damped by an oscillator bath with Ohmic spectral density yields an effective environment with Lorentzian spectral density [129]. Since this effective spectral density is a continuous function of frequency, the damped oscillator can be identified with a macroscopic reservoir. The idea underlying our simulation protocol is that Lorentzians can serve as a basis to decompose any spectral density. Thus, arbitrary spectral densities can be generated by combining several damped modes. Since spins are easily encoded with trapped ions and spin-motion coupling and laser cooling are standard techniques, it seems that such a simulation is readily implemented.

However, the damping of trapped ions is described by a Lindblad equation and not the damping model above. Therefore, we showed in a path integral approach that a damped oscillator in Lindblad description can be identified with the oscillator damped by an Ohmic bath for appropriate parameters. Hence, for appropriate parameters also the damped oscillator in Lindblad description corresponds to a bath with Lorentzian spectral density. We corroborated these analytical considerations by comparing numerical simulations of a spin coupled to a damped oscillator in Lindblad description with simulations of the full many-body dynamics of a spin-boson model with Lorentzian spectral density. We obtained excellent agreement between the simulations of the effective and the full many-body models. Based on these results, we concluded that the idea for the simulation of spin-boson models can be realized using trapped ions.

We then showed that our protocol can be implemented with current ion trap technology by simulating an experiment with parameters where the above correspondence holds and which are realistic for current experiments. The simulated experiment corresponds to the model of a spin coupled to a bath with Lorentzian spectral density. Interestingly, the tunability of the experimental parameters allows us to enter parameter regimes which are not accessible with
the full many-body simulation. We should note, though, that the effective model can still be simulated efficiently on a conventional computer. Finally, we also showed that the proposed simulation protocol is robust with respect to the most common experimental errors.

Extending the simulations beyond such a proof-of-principle experiment, our protocol could be used to simulate spin-boson models in parameter regimes which are not accessible with currently available classical simulations. The model in the simulated experiment is easily extended to spin-boson models with more complex spectral densities by coupling the spin to more than one mode. Since the identification of the damped oscillator with the macroscopic bath works best when the cooling of the mode is considerably smaller than the mode frequencies, our simulation protocol is well-suited for the simulation of structured environments with pronounced resonances.

In these environments the resonances correspond to long-lived vibrational modes \cite{140}, which can lead to long memory times of the environment. As we remarked earlier, environments with long memory times are hard to simulate with classical computers. Here, a trapped-ion simulator could be beneficial. One instance where structured spectral densities are used is the modeling of photosynthetic pigment protein complexes. The models of these complexes typically involve several coupled two-level systems which are subject to damping \cite{125}. Interactions between spins are nowadays routinely implemented in ion trap experiments by coupling the spins through one or several modes of motion \cite{19–21}. The remaining modes could then serve to model the environment employing our protocol. Hence, our protocol could be used to simulate the dynamics of photosynthetic pigment protein complexes.

We can now also make a connection to the results of the first part of the thesis. A standard tool for the investigation of photosynthetic pigment protein complexes is 2D spectroscopy. The 2D spectra can yield insight into the mechanisms of photosynthesis and are exceedingly hard to simulate on classical computers \cite{126}. Hence, trapped ions could serve as a quantum simulator for two-dimensional spectra of pigment protein complexes or, if one is more ambitious, as a simulator of the dynamics of these complexes. In this way, trapped ions could help to understand the physical mechanisms participating in photosynthesis.
A. Derivation of EIT cooling master equation

This appendix is devoted to the derivation of the master equation for EIT cooling given in Eq. (2.354) starting from Eq. (2.353). The second line of Eq. (2.353) reads

\[ \frac{\partial}{\partial t} P_0 \rho(t) = \left[ P_0 L_2 P_0 + \int_0^\infty d\tau P_0 L_1 Q_0 e^{i\tau} Q_0 L_1 P_0 \right] \rho(t). \]  

(A.1)

In order to evaluate the right-hand side of Eq. (A.1) we recall that

\[ L_1 \rho = -i \hbar [V_1 q_z, \rho] \]  

\[ L_2 \rho = -i \hbar [V_2 q_z^2, \rho] + D_2 \rho = -i \hbar [V_2 q_z^2, \rho] - \frac{i}{\hbar} [V_2, \rho] q_z^2 + D_2 \rho. \]  

(A.2)

We start by evaluating the first term on the right-hand side of Eq. (A.1)

\[ P_0 L_2 P_0 \rho(t) = P_0 \left[ \frac{i}{\hbar} V_2 q_z \otimes \mu(t) - \frac{i}{\hbar} [V_2, \rho] \otimes \mu(t) q_z^2 + D_2 \rho \otimes \mu(t) \right] \rho(t) \]

\[ = \rho_0 \otimes \left[ -\frac{i}{\hbar} \text{Tr}_I (V_2 \rho_0) \rho_0 \otimes [q_z^2, \mu(t)] - \frac{i}{\hbar} \text{Tr}_I ([V_2, \rho_0]) \rho_0 \otimes [q_z^2, \mu(t)] \right] \]

\[ = -\frac{i}{\hbar} \text{Tr}_I (V_2 \rho_0) \rho_0 \otimes \rho_0 \otimes [q_z^2, \mu(t)] \]

\[ = 0. \]

From the second to the third line we used that \( D_2 \rho_0 \otimes \mu(t) = 0 \) since the dark state does not have any populations in \( |e\rangle \langle e| \), while from the third to the fourth line we used the cyclic property of the trace. The remaining expression vanishes because it only takes the populations in \( \rho_0 \) to off-diagonal elements which do not contribute to the trace.

Before we move to the second term of the right-hand side of Eq. (A.1), we split \( L_0 \) from Eq. (2.344) into its parts acting on the internal and external degrees of freedom

\[ L_0 = L_{0,1} + L_{0,E} \]  

(A.4)

\[ L_{0,1} \rho = -\frac{i}{\hbar} [H_{\text{EIT},0}, \rho] + D_0 \rho \]  

(A.5)

\[ L_{0,E} \rho = -\frac{i}{\hbar} [H_m, \rho] = -i [\omega a^\dagger a, \rho]. \]  

(A.6)

Using these new Liouvillians, the second term on the right-hand side of Eq. (A.1) can be written.
as
\[
\int_0^\infty d\tau P_0 L_1 Q_0 e^{z_0 \tau} Q_0 L_1 P_0 \rho(t) = \int_0^\infty d\tau P_0 L_1 Q_0 e^{z_0 \tau} Q_0 L_1 \rho_\Box \otimes \mu(t)
\]
\[
= -\frac{1}{\hbar^2} \int_0^\infty d\tau P_0 \left\{ V_1 e^{z_0 \tau} V_1 \rho_\Box \otimes [q_z, e^{z_0 \tau}[q_z, \mu(t)]] + [V_1, e^{z_0 \tau} V_1 \rho_\Box] \otimes \left( e^{z_0 \tau}[q_z, \mu(t)] \right) q_z + V_1 e^{z_0 \tau} V_1 \rho_\Box \otimes [q_z, e^{z_0 \tau} \mu(t) q_z] + [V_1, e^{z_0 \tau} V_1 \rho_\Box] \otimes \left( e^{z_0 \tau} \mu(t) q_z \right) q_z \right\}
\]
\[
= -\frac{1}{\hbar^2} \int_0^\infty d\tau \rho_\Box \otimes \left\{ T_1 \left( V_1 e^{z_0 \tau} V_1 \rho_\Box \right) P_E[q_z, e^{z_0 \tau}[q_z, \mu(t)]] + T_1 \left( V_1 e^{z_0 \tau} V_1 \rho_\Box \right) P_E[q_z, e^{z_0 \tau} \mu(t) q_z] \right\}.
\]

For the last equality we have used that the second and fourth terms in the penultimate expression vanish, as can easily be seen using the cycling property of the trace. Using that the evolution of the external degrees of freedom is unitary and that \(\mu(t)\) is diagonal, we may write
\[
e^{z_0 \tau}[q_z, \mu(t)] = [q_z(\tau), \mu(t)] \quad (A.7)
\]
where \(q_z(\tau) = e^{-\frac{i}{\hbar}z_0 \tau} q_z e^{\frac{i}{\hbar}z_0 \tau}\). After performing the trace over the internal degrees of freedom and using the notation above, we can write Eq. (A.1) as
\[
\mu(t) = -\frac{1}{\hbar^2} \int_0^\infty d\tau \left\{ T_1 \left( V_1 e^{z_0 \tau} V_1 \rho_\Box \right) P_E[q_z, \mu(t)] + T_1 \left( V_1 e^{z_0 \tau} V_1 \rho_\Box \right) P_E[q_z(\tau), \mu(t)] q_z(\tau) \right\}. \quad (A.8)
\]
In the above equation we can identify the two-time correlation functions of the internal operator \(V_1\) in the stationary state
\[
\langle V_1(\tau)V_1(0)\rangle_{\text{ss}} = T_1[V_1 e^{z_0 \tau} V_1 \rho_\Box], \quad (A.9)
\]
\[
\langle V_1(0)V_1(0)\rangle_{\text{ss}} = T_1[V_1 e^{z_0 \tau} \rho_\Box V_1], \quad (A.10)
\]
where the subscript “ss” stands for stationary state. Introducing the two-time correlation functions in Eq. (A.8) and performing the projector \(P_E\) over the external degrees of freedom, we arrive at
\[
\bar{\mu}(t) = -\frac{z_0^2}{\hbar^2} \int_0^\infty d\tau \left\{ \langle V_1(\tau)V_1(0)\rangle_{\text{ss}} \left[ a^\dagger a(\tau) \mu(t) + a a^\dagger(\tau) \mu(t) - a(\tau) \mu(t) a^\dagger - a^\dagger(\tau) \mu(t) a \right] + \langle V_1(0)V_1(0)\rangle_{\text{ss}} \left[ -a(\tau) a^\dagger(\tau) - a^\dagger(\tau) \mu(t) a + \mu(t) a(\tau) a + \mu(t) a^\dagger(\tau) a^\dagger \right] \right\} \quad (A.11)
\]
where we have used the decomposition of \(z\) in terms of creation and annihilation operators \(z = z_0(a + a^\dagger)\) with \(z_0 = \sqrt{\hbar/(2m\omega_0)}\). Note that here, in contrast to the usual interaction picture operators, we have \(a(\tau) = e^{i\omega_0 \tau} a\) and \(a^\dagger(\tau) = e^{-i\omega_0 \tau} a^\dagger\). With the help of the identity \(\langle V_1(\tau)V_1(0)\rangle_{\text{ss}} = \langle V_1(0)V_1(\tau)\rangle_{\text{ss}}\) we reach the final result
\[
\bar{\mu}(t) = \Lambda_\pm [a(\mu(t) a^\dagger - a^\dagger a(\mu(t))] + \Lambda_\pm [a^\dagger(\mu(t) a - a(\mu(t)) a^\dagger] + \text{H.c.}, \quad (A.12)
\]
where we have introduced the rates
\[
\Lambda_\pm = z_0^2 S(\pm \omega_0) = \frac{z_0^2}{\hbar^2} \int_0^\infty d\tau \langle V_1(\tau)V_1(0)\rangle_{\text{ss}} e^{\pm i\omega_0 \tau} \quad (A.13)
\]
that are determined by the fluctuation spectra of $V_1$
\[ S(\pm \omega_e) = \frac{1}{\hbar} \int_0^\infty d\tau \langle V_1(\tau)V_1(0) \rangle_{ss} e^{\pm i\omega_e \tau}. \] (A.14)

Finally, we briefly sketch the evaluation of the fluctuation spectra of $V_1$ in Eq. (2.355). Actually, by defining $\langle V_1(\tau)V_1(0) \rangle_{ss}$ and $\langle V_1(0)V_1(\tau) \rangle_{ss}$ in Eqs. (A.9) and (A.10) we have invoked the quantum regression theorem, see section 2.4.3. According to the regression theorem the correlations obey the same equations of motion as the mean values. Therefore, we start by considering the equations of motion for the internal levels, i.e. the optical Bloch equations for the three-level system in Fig. 2.7 a) that evolves according to Eq. (2.336). Let us define the vector $\boldsymbol{\sigma}$ containing the nine basis states
\[ \boldsymbol{\sigma} = (|g_1\rangle \langle g_1|, |g_1\rangle \langle g_2|, |g_1\rangle \langle e|, |g_2\rangle \langle g_1|, |g_2\rangle \langle g_2|, |g_2\rangle \langle e|, |e\rangle \langle g_1|, |e\rangle \langle g_2|, |e\rangle \langle e|). \] (A.15)

The optical Bloch equations resulting from Eq. (2.336) are given by
\[ \langle \dot{\boldsymbol{\sigma}} \rangle = M \langle \boldsymbol{\sigma} \rangle, \] (A.16)
where the coefficient matrix $M$ is given by
\[
M = \begin{pmatrix}
0 & 0 & -i \frac{\Omega_1}{2} & 0 & 0 & 0 & i \frac{\Omega_1}{2} & 0 & \Gamma_1 \\
0 & 0 & -i \frac{\Omega_2}{2} & 0 & 0 & 0 & i \frac{\Omega_2}{2} & 0 & 0 \\
-i \frac{\Omega_1}{2} & -i \frac{\Omega_2}{2} & -i \Delta_{\text{EIT}} + \frac{\Gamma}{2} & 0 & 0 & 0 & i \frac{\Omega_1}{2} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & -i \frac{\Omega_2}{2} & 0 & i \frac{\Omega_2}{2} & 0 \\
i \frac{\Omega_1}{2} & 0 & 0 & -i \frac{\Omega_2}{2} & -i \Delta_{\text{EIT}} + \frac{\Gamma}{2} & 0 & 0 & i \frac{\Omega_2}{2} & 0 \\
i \frac{\Omega_2}{2} & 0 & 0 & i \frac{\Omega_1}{2} & 0 & 0 & i \Delta_{\text{EIT}} - \frac{\Gamma}{2} & 0 & -i \frac{\Omega_1}{2} \\
i \frac{\Omega_2}{2} & 0 & 0 & i \frac{\Omega_1}{2} & 0 & 0 & i \Delta_{\text{EIT}} - \frac{\Gamma}{2} & 0 & -i \frac{\Omega_1}{2} \\
0 & i \frac{\Omega_1}{2} & 0 & 0 & i \frac{\Omega_2}{2} & 0 & 0 & i \frac{\Omega_2}{2} & -i \frac{\Omega_1}{2} \\
0 & 0 & i \frac{\Omega_1}{2} & 0 & 0 & i \frac{\Omega_2}{2} & -i \frac{\Omega_1}{2} & -i \frac{\Omega_2}{2} & -\Gamma
\end{pmatrix}. \] (A.17)

According to the regression theorem the correlations obey the equations of motion
\[ \frac{d}{d\tau} \langle \boldsymbol{\sigma}(\tau + t)\sigma_i(\tau) \rangle = M \langle \boldsymbol{\sigma}(\tau + t)\sigma_i(\tau) \rangle, \] (A.18)
where $\sigma_i$ is the $i$th entry of $\boldsymbol{\sigma}$. Formally, we consider the limit $\tau \to \infty$ because we actually consider the fluctuation spectra in the stationary (asymptotic) state of the dynamics. Hence, we write Eq. (A.18) as
\[ \frac{d}{dt} \langle \sigma(t)\sigma_i(0) \rangle_{ss} = M \langle \sigma(t)\sigma_i(0) \rangle_{ss}. \] (A.19)

The above equation can be easily solved for its Laplace transform. Applying a Laplace transform to both sides of Eq. (A.19) yields
\[ \int_0^\infty e^{-st} \frac{d}{dt} \langle \sigma(t)\sigma_i(0) \rangle_{ss} dt = M \int_0^\infty e^{-st} \langle \sigma(t)\sigma_i(0) \rangle_{ss} dt, \] (A.20)
where $s = \pm i\omega + \varepsilon$ with $\omega \in \mathbb{R}$ and $\varepsilon > 0$. Performing an integration by parts on the left-hand side of the above equation we obtain
\[ \int_0^\infty e^{-st} \langle \sigma(t)\sigma_i(0) \rangle_{ss} dt = (sI - M)^{-1} \langle \sigma(0)\sigma_i(0) \rangle_{ss}. \] (A.21)

Comparing with Eq. (A.14), we note that the quantity we are looking for is the left hand side of the above equation with $s = \pm i\omega$. Yet, for the derivation of the above equality we have assumed
that $s = \pm i\omega + \varepsilon$ with $\varepsilon > 0$. The idea is that we consider $\varepsilon$ very small such that it ensures convergence and let it tend to zero in the end of the calculation. Hence, we can solve for the fluctuation spectra by just computing the matrix inverse $(s\mathbb{I} - M)^{-1}$, once we know the initial conditions.

It is easy to see that $\langle \sigma(0)\sigma(0) \rangle_{ss}$ only contains elements of $\langle \sigma \rangle_{ss}$. Thus, in order to solve Eq. (A.21) and thereafter Eq. (A.14), we need to find the steady state $\langle \sigma \rangle_{ss}$. One way to find $\langle \sigma \rangle_{ss}$ is to bring the optical Bloch equations in Eq. (A.16) to an inhomogeneous form

$$\frac{d}{dt} \langle \tilde{\sigma}(t) \rangle = \tilde{M} \langle \tilde{\sigma}(t) \rangle + a,$$

(A.22)

where $\tilde{\sigma}$ is $\sigma$ from Eq. (A.15) without the last entry, $|e\rangle\langle e|$. One can arrive at Eq. (A.22) from Eq. (A.16) using $|e\rangle\langle e| = 1 - |g_1\rangle\langle g_1| - |g_2\rangle\langle g_2|$. $\langle \sigma \rangle_{ss}$ is readily obtained from Eq. (A.22) as

$$\langle \tilde{\sigma} \rangle_{ss} = -\tilde{M}^{-1}a.$$

(A.23)

Using $|e\rangle\langle e| = 1 - |g_1\rangle\langle g_1| - |g_2\rangle\langle g_2|$ again, we obtain $\langle \tilde{\sigma} \rangle_{ss}$ from the above expression. Inserting the result into Eq. (A.21) we finally obtain the spectra $S(\pm \omega\varepsilon)$ from Eq. (A.14), which allow us to compute the rates $A\pm$ in Eq. (2.362) and the shift $\Delta \omega\varepsilon$ in Eq. (2.365).
B. Derivation of the absorption spectrum of the \( \Lambda \) system in the EIT cooling configuration

In this appendix we show one way to obtain the absorption spectrum displayed in Fig. 2.7 b). It is the absorption spectrum of a \( \Lambda \)-system irradiated by the laser configuration shown in part a) of the same figure. In an interaction picture with respect to \( H_0 = - \sum s h \omega_s \langle g_s | g_s \rangle \), the interaction of the \( \Lambda \)-system with the applied lasers is described by the Hamiltonian of Eq. (2.329)

\[
H_{\text{EIT},0} = - \hbar \Delta_{\text{EIT}} \sum_{s=1,2} |g_s \rangle \langle g_s | + \frac{\hbar \Omega}{2} |e \rangle \langle \psi_c | + \text{H.c.},
\]

where we have introduced the state

\[
|\psi_c \rangle = \frac{1}{\Omega} (\Omega_1 |g_1 \rangle + \Omega_2 |g_2 \rangle).
\]

Spontaneous emission from the upper level is accounted for by the dissipator of Eq. (2.331)

\[
\mathcal{D}_0 \rho = \sum_{s=1,2} \Gamma_s |g_s \rangle \langle e | \rho | g_s \rangle \langle e | - \frac{\Gamma}{2} \{ |e \rangle \langle e |, \rho \}.
\]

Let us now consider that additionally a weak probe field is applied on the transition \( |g_1 \rangle \leftrightarrow |e \rangle \). The interaction of the \( \Lambda \)-system with the probe field is described by

\[
H_p = \frac{\hbar \Omega_p}{2} \left( e^{-i \Delta_p t} |e \rangle \langle g_1 | + \text{H.c.} \right),
\]

where \( \Delta_p = \Delta_{\text{EIT}} - \Delta_p \) and \( \Delta_p = \omega_1 - \omega_p \) is the detuning of the probe field of frequency \( \omega_p \). \( \Omega_p \) is taken to be of order \( \varepsilon \) times a typical entry of \( H_{\text{EIT},0} \), where \( \varepsilon \ll 1 \). We now want to find the excited state population due to the probe field. Since the probe is assumed to be weak, we work in first order perturbation theory. Defining the Liouvillians

\[
\mathcal{L}_0 \rho = - \frac{i}{\hbar} [H_{\text{EIT},0}, \rho] + \mathcal{D}_0 \rho,
\]

\[
\mathcal{L}_1 \rho = - \frac{i}{\hbar} [H_p, \rho],
\]

we may write the time evolution of the system as

\[
\dot{\rho} = (\mathcal{L}_0 + \mathcal{L}_1) \rho.
\]

We now proceed in the usual way and expand the density operator in powers of \( \varepsilon \): \( \rho = \rho^{(0)} + \varepsilon \rho^{(1)} + \varepsilon^2 \rho^{(2)} + \ldots \). Equating terms of equal powers in \( \varepsilon \) yields the zeroth and first order contributions

\[
\dot{\rho}^{(0)} = \mathcal{L}_0 \rho^{(0)},
\]

\[
\dot{\rho}^{(1)} = \mathcal{L}_0 \rho^{(1)} + \mathcal{L}_1 \rho^{(0)}.
\]
B. Derivation of the absorption spectrum of the $\Lambda$ system in the EIT cooling configuration

We assume that the initial state is the dark state $|\psi_d\rangle$ and $\rho(0) = \rho^{(0)}(0) = |\psi_d\rangle\langle\psi_d|$. We then find from Eq. (B.8) that $\dot{\rho}^{(0)}(0) = 0$ and hence $\rho^{(0)}(t) = |\psi_d\rangle\langle\psi_d|$. Inserting this result into the equation for $\rho^{(1)}$ yields

$$\dot{\rho}^{(1)} = \mathcal{L}_0 \rho^{(1)} + \mathcal{L}_1 |\psi_d\rangle\langle\psi_d|. \quad (B.10)$$

Since $\rho^{(1)}(0) = 0$ and the homogeneous solution $\rho^{(1)}_{\text{hom}}(t) = e^{\mathcal{L}_0 t} \rho^{(1)}_{\text{hom}}(0)$, we can disregard the homogeneous solution. A particular solution of Eq. (B.10) is given by

$$\rho^{(1)}_p(t) = \int_0^t dt' e^{\mathcal{L}_0 (t-t')} \mathcal{L}_1 (t') |\psi_d\rangle\langle\psi_d|, \quad (B.11)$$

where we have written $\mathcal{L}_1$ with a time argument to make clear that it is time dependent. Applying $\mathcal{L}_1$ to the dark state, we obtain

$$\rho^{(1)}_p(t) = \frac{\Omega_p^2 \Omega_2}{2\Omega} e^{\mathcal{L}_0 (t-t')} \int_0^t dt' e^{\mathcal{L}_0 t'} \left[ e^{i\Delta_p t'} |\psi_d\rangle\langle\psi_d| - \text{H.c.} \right]. \quad (B.12)$$

In order to solve the above equation, it would be desirable to be able to expand $|\psi_d\rangle\langle\psi|_d$ and its Hermitian conjugate in terms of eigenstates of $\mathcal{L}_0$. We know that $|\psi_d\rangle\langle\psi_d|$ is an eigenstate of $\mathcal{L}_0$ with eigenvalue zero and with the ansatz $\mathcal{L}_0 A = \lambda A$, where $A = |\psi_d\rangle\langle\psi_d| + b|\psi_e\rangle\langle\psi_e| + c|\psi_c\rangle\langle\psi_c|$, we find the eigenstates and eigenvalues

$$A_\pm = |\psi_d\rangle \left( e^\pm i\Omega \frac{\lambda_\pm}{\lambda_\pm} \langle\psi_c| \right), \quad (B.13)$$

$$\lambda_\pm = \frac{i\Delta_{\text{EIT}} - \Gamma/2 \pm \sqrt{(i\Delta_{\text{EIT}} - \Gamma/2)^2 - \Omega^2}}{2}. \quad (B.14)$$

Now, $|\psi_d\rangle\langle\psi|$ can be written in terms of the above eigenvectors and -values as

$$|\psi_d\rangle\langle\psi| = \frac{\lambda_+ A_+ - \lambda_- A_-}{\lambda_+ - \lambda_-}. \quad (B.15)$$

Inserting the above expansion of $|\psi_d\rangle\langle\psi|$ into Eq. (B.12) and solving the integral, we obtain

$$\rho^{(1)}_p(t) = \frac{\Omega_p^2 \Omega_2}{2\Omega} \left[ \frac{\lambda_+}{\lambda_+ - \lambda_-} A_+ e^{i\Delta_p t} - e^{\lambda_+ t} \frac{\lambda_-}{\lambda_+ - \lambda_-} A_- e^{i\Delta_p t} - \text{H.c.} \right]. \quad (B.16)$$

In the considered regime the eigenvalues have a negative real part and we can write the above equation for times $t$ where $-\text{Re}(\lambda_\pm) t \gg 1$ as

$$\rho^{(1)}_p(t) = \frac{\Omega_p^2 \Omega_2}{2\Omega} \left[ \frac{\lambda_+}{\lambda_+ - \lambda_-} A_+ e^{i\Delta_p t} - e^{\lambda_+ t} \frac{\lambda_-}{\lambda_+ - \lambda_-} A_- e^{i\Delta_p t} - \text{H.c.} \right]. \quad (B.17)$$

Let us now consider the optical Bloch equations for the time evolution in Eq. (B.7). For the excited state population we obtain

$$\rho_{ee} = \text{Im}(\Omega_1 |\rho_{1, e}\rangle) + \text{Im}(\Omega_2 |\rho_{2, e}\rangle) - \Gamma \rho_{ee} + \text{Im}(\Omega_p e^{-i\Delta_p} |\rho_{1, e}\rangle), \quad (B.18)$$

where $|\rho_{ij}\rangle = \langle i |\rho| j\rangle$. The first two terms of the above equation represent absorption from the EIT beams and the third the decay of the excited state. The fourth term describes absorption from the probe beam. Thus, we have to consider the term

$$\gamma_{abs} = \text{Im}(\Omega_p e^{-i\Delta_p} |\rho_{1, e}\rangle) \quad (B.19)$$

to obtain the absorption spectrum of the probe beam. Inserting the element $|\rho_{1, e}\rangle$ from Eq. (B.17) into the above expression we obtain

$$\gamma_{abs} = \frac{\Omega_p^2 \Omega_2^2}{2\Omega^2} \text{Re} \left[ \frac{\lambda_+}{\lambda_+ - \lambda_-} \frac{1}{\lambda_+ - \lambda_-} - \frac{\lambda_-}{\lambda_+ - \lambda_-} \frac{1}{\lambda_+ - \lambda_-} \right]. \quad (B.20)$$

This is the expression we were looking for.
C. Linear chain mappings and the spin-boson model

This appendix gives a brief presentation of the main results that allow us to map the spin-boson model Hamiltonian in Eq. (4.5) to a semi-infinite linear chain with only nearest neighbor couplings. The details of this derivation may be found in [134]. The motivation to map the spin-boson model to the chain configuration is that the chain as a one-dimensional quantum system is amenable to numerical treatment with techniques based on the time-dependent density matrix renormalization group (t-DMRG). We shall not be concerned with the details of the t-DMRG simulations here. A recent review on density renormalization group methods is [170] and details regarding the simulations presented in the main text may be found in the supplemental material of [171]. Let us only comment that the employed time evolving density matrix with orthogonal polynomials algorithm (TEDOPA) was introduced in [133, 134] and is a certifiable and numerically exact method to treat open quantum system dynamics [172].

For an environment with a macroscopic number of oscillators with a continuous distribution of frequencies, the spin-boson Hamiltonian may be written as [134]

\[ H = H_S + H_E + H_{\text{int}}, \]

\[ H_S = \frac{\varepsilon}{2} \sigma^z - \frac{\hbar \Delta}{2} \sigma^x, \]

\[ H_E = \hbar \int_0^{\omega_{\text{max}}} d\omega \ g(\omega) a_\omega^\dagger a_\omega, \]

\[ H_{\text{int}} = -\sigma^z \frac{\hbar}{2} \int_0^{\omega_{\text{max}}} d\omega \ h(\omega)(a_\omega + a_\omega^\dagger). \]

Here we assumed a hard cutoff \( \Theta(\omega - \omega_{\text{max}}) \) for the frequencies of the oscillators in the environment, \( g(\omega) \) is the dispersion relation of the oscillators in the environment and \( h(\omega) \) the coupling of the spin to the environmental mode of frequency \( \omega \). Note that \( h(\omega) \) is a real quantity. The annihilation and creation operators \( a_{\omega} \) and \( a_{\omega}^\dagger \) of the environmental modes obey the commutation relation \( [a_\omega, a_\omega^\dagger] = \delta(\omega - \omega') \).

The influence of the environment is determined by the spectral density \( J(\omega) \). The spectral density can be written in terms of the functions \( g(\omega) \) and \( h(\omega) \) as [132]

\[ J(\omega) = \pi \hbar^2 [g^{-1}(\omega)] \frac{dg^{-1}(\omega)}{d\omega}, \]

where \( \frac{dg^{-1}(\omega)}{d\omega} \) may be understood as a density of states [134]. Interestingly, if an environment is characterized by a certain spectral density, the functions \( g(\omega) \) and \( h(\omega) \) are not uniquely defined which allows us the choice \( g(\omega) = \omega \).

For this choice of the dispersion relation, we obtain

\[ J(\omega) = \pi \hbar^2 (\omega). \]

We now define new creation and annihilation operators \( b_\omega^\dagger \) and \( b_\omega \) according to

\[ b_\omega^\dagger = \int_0^{\omega_{\text{max}}} d\omega \ U_\omega(\omega) a_\omega^\dagger, \]

\[ b_\omega = \int_0^{\omega_{\text{max}}} d\omega \ U_\omega(\omega) a_\omega, \]

\[ U_\omega(\omega) = \frac{\hbar}{\sqrt{\pi}} \frac{1}{\omega} \exp\left(-\frac{\omega^2}{\omega^2_{\text{max}}} \right) \]
where the $U_n(\omega)$ are unitary transformations given by

$$U_n(\omega) = h(\omega)p_n(\omega). \quad (C.8)$$

The $p_n(\omega)$, $n = 0, 1, \ldots$ are orthogonal polynomials with respect to the measure $d\mu(\omega) = h^2(\omega)d\omega$ induced by the coupling [134]. Writing the Hamiltonian in Eq. (C.1) in terms of the new operators $b_n, b_n^\dagger$ from Eq. (C.7) and using the choice $g(\omega) = \omega$, we obtain [134]

$$H = H_S - \hbar \frac{t_0}{2} \sigma^z (b_0 + b_0^\dagger) + \sum_{n=0}^{\infty} \hbar \omega_n b_n^\dagger b_n + \sum_{n=0}^{\infty} \hbar t_n (b_n b_{n+1}^\dagger + b_{n+1} b_n^\dagger), \quad (C.9)$$

where the coupling $t_0$ is given by

$$t_0 = \sqrt{\frac{1}{\pi} \int_0^\infty d\omega J(\omega)}, \quad (C.10)$$

while the frequencies $\omega_n$ and couplings $t_n$ are determined by the coefficients of the recursion relation of the $p_n$. Note that while in some cases it is possible to find these coefficients analytically in the majority of cases one will have to resort to numerical methods to find the coefficients [134]. Now that the transformation of the Hamiltonian of the spin-boson model to a linear chain is complete the system can be treated with t-DMRG.

Let us finally comment on the time scales of the simulations. The frequencies and couplings of the chain become homogeneous very quickly such that excitations traveling away from the spin (or in general the open system) will not be scattered and reflected once they have entered this region. For the simulations, the chain of course has to be truncated at some point. At this “hard end” excitations will be reflected and one has to take care that these excitations do not come back to the spin during the simulation time. Furthermore, the local Hilbert spaces of the oscillators along the chain have to be truncated. Obviously, for lower temperatures of the reservoir, and thus the chain, smaller local dimensions suffice while finite temperatures demand a higher local dimension. Consequently, at lower temperatures the chains, and thus the simulation times, can be chosen longer. For the simulations in Chapter 4 the relevant time scale was $|20/\Delta|$ and we considered finite temperatures. For the simulation with $\Delta/2\pi = -50\text{kHz}$ the chain had $n = 15$ sites with a local dimension of the oscillators $\sim 5$. This chain length was sufficient to avoid excitations coming back to the spin during the considered time interval. However, these simulations took 15 days on the BWfor cluster JUSTUS using 16 cores. The time scale of the simulated evolution for the “Ohmic” case, where $\Delta/2\pi = 3\text{kHz}$, would be about a factor of 15 larger demanding a chain longer by roughly the same factor. Accordingly, such simulation with the current method is not available.
D. Numerical evaluation of the measure of non-Markovianity $\mathcal{N}_{\text{RHP}}$

This appendix describes the details of the numerical evaluation of the measure of non-Markovianity $\mathcal{N}_{\text{RHP}}$. In order to evaluate $\mathcal{N}_{\text{RHP}}$ for a given internal $I = [0, t]$ we divide the time interval in equally spaced discrete times $t_i = i\Delta t$, $i = 0, \ldots, N$, where $\Delta t = t/N$. We then compute the time evolution of the basis states $|k\rangle\langle j|$, $k, j \in \{e, g\}$ for all $t_i$. We denote the time-evolved states by $|k\rangle\langle j|(t_i) = \rho_{kj}(t_i)$. Note that while the $|k\rangle\langle j|$ are single elements of $2 \times 2$ matrices the $\rho_{kj}(t_i)$ are in general full complex $2 \times 2$ matrices. We now write the $\rho_{kj}(t_i)$ as vectors

$$v_{kj}(t_i) = \begin{pmatrix} \rho_{kj,ee}(t_i) \\ \rho_{kj,eg}(t_i) \\ \rho_{kj,ge}(t_i) \\ \rho_{kj,gg}(t_i) \end{pmatrix}, \quad (D.1)$$

where $\rho_{kj,ee}(t_i) = \langle e|\rho_{kj}(t_i)|e\rangle$ and so on. Note that this is not the ordering that is usually taken for the operation of “vectorization”. With the vectors $v_{kj}(t_i)$ we can write down a matrix representation of the dynamical map $\mathcal{E}(t_i, t_0)$

$$E(t_i, t_0) = [v_{ee}(t_i), v_{eg}(t_i), v_{ge}(t_i), v_{gg}(t_i)]. \quad (D.2)$$

The matrix representation of the dynamical map $\mathcal{E}(t_2, t_1)$, $t_2 \geq t_1 \geq t_0$ is then computed according to Eq. (4.199) as

$$E(t_2, t_1) = E(t_2, t_0)E^{-1}(t_1, t_0), \quad (D.3)$$

where $E^{-1}(t_1, t_0)$ is the normal matrix inverse of $E(t_1, t_0)$. Now, we need to construct the Choi matrix $[\mathcal{E}(t_2, t_1) \otimes \mathbb{I}_d]|\psi\rangle\langle\psi|$ from $E(t_2, t_1)$. It turns out that the Choi matrix is proportional to the reshuffled matrix $E^R(t_2, t_1)$ of the matrix $E(t_2, t_1)$ [173]

$$[\mathcal{E}(t_2, t_1) \otimes \mathbb{I}_d]|\psi\rangle\langle\psi| = \frac{1}{d}E^R(t_2, t_1), \quad (D.4)$$

where $d$ is the dimension of the considered open quantum system. For the case of a spin $E^R(t_2, t_1)$ reads

$$E^R(t_2, t_1) = \begin{pmatrix} E_{11} & E_{12} & E_{21} & E_{22} \\ E_{13} & E_{14} & E_{23} & E_{24} \\ E_{31} & E_{32} & E_{41} & E_{42} \\ E_{33} & E_{34} & E_{43} & E_{44} \end{pmatrix}, \quad (D.5)$$

where $E_{mn}$ is the entry of row $m$ and column $n$ of the $4 \times 4$ matrix $E(t_2, t_1)$. From the Choi matrices we obtain a discrete version of $g(t)$ at times $t_i$

$$g(t_i) = \frac{||[\mathcal{E}(t_{i+1}, t_i) \otimes \mathbb{I}_d]|\psi\rangle\langle\psi||| - 1}{t_{i+1} - t_i} = \frac{\frac{1}{d}E^R(t_{i+1}, t_i)|| - 1}{\Delta t}, \quad (D.6)$$

which allows us to evaluate $\mathcal{N}_{\text{RHP}}$ on the grid given by the $t_i$. The discrete version of Eq. (4.202) then becomes

$$\mathcal{N}_{\text{RHP}} = \sum_{i=1}^{N} g(t_i) > 0 \tanh[g(t_i)] \quad (D.7)$$

\text{167}
where $N_{g(t_i)>0}$ is the number of events where $g(t_i) > 0$.

Note that the actual difficulty in evaluating $\mathcal{M}_{RHP}$ is the numerical evaluation of $g(t_i)$. The problem is to define a threshold that defines which values of $g(t_i)$ count as zero and which are counted as finite. For example, a value of $10^{-10}$ for $g(t_i)$ might seem negligible but becomes appreciable if it is divided by a time step of the same order of magnitude. In order to define a threshold, we tried to estimate the numerical precision of the calculations. To this end, we compared the density matrices obtained by using different routines to perform the time evolution. The numerical calculations were performed in Python using the Numpy and Scipy libraries. The time evolution was performed by vectorizing the Lindblad equation (4.182) and applying the matrix exponential of the Liouvillian on the vectorized initial state. Usually we used the scipy.sparse.linalg.expm_multiply routine for this. For a number of parameters we performed the time evolution by solving the matrix exponential first using the scipy.sparse.linalg.expm routine and then multiplying the matrix obtained in the first step with the vectorized initial state. We compared the resulting density matrices by computing the trace distance between them. For all initial states $|k\rangle\langle j|$, $k, j \in \{e, g\}$ the time evolved states typically showed trace distances of a few times $10^{-16}$ and the sum of the largest errors of all the basis states yielded a few times $10^{-15}$. We took this value as a rough estimate of the numerical precision and set $g(t_i) = 0$ for $g(t_i) < 10^{-14}$. 


Bibliography


Bibliography


177


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Publications

Two-Dimensional Spectroscopy for the Study of Ion Coulomb Crystals,

Simulating spin-boson models with trapped ions,

[L3] A. Lemmer, A. Bermudez, and M. B. Plenio,
Noise Studies of Driven Geometric Phase Gates with Trapped Ions,
in Ion Traps for Tomorrows Applications – Proceedings of the International School of

[L4] A. Lemmer, A. Bermudez, and M. B. Plenio,
Driven geometric phase gates with trapped ions,

Conference contributions

Aug. 2016  Simulating spin-boson models with trapped ions
Poster, European Conference on Trapped Ions 2016, 29/08/16-02/09/16, Arosa, Switzerland.

Mar. 2016  Simulating spin-boson models with trapped ions
Talk, Frühjahrstagung der Deutschen Physikalischen Gesellschaft 29/02/16-04/03/16, Hannover, Germany, 02/03/2016.

Dec. 2014  Two-dimensional spectroscopy for the study of ion Coulomb crystals
01/12/14-05/12/14, Bad Honnef, Germany.

Sep. 2014  Two-dimensional spectroscopy for the study of ion Coulomb crystals
Poster, European Conference on Trapped Ions 2014, 15/09/14-19/09/14, Mainz, Germany.

Apr. 2014  Observing Non-Lineairties in Ion Traps Using Two-Dimensional Spectroscopy
Talk, Qion ’14, 31/03/14-04/04/14, Cartagena, Spain, 03/04/2014.

Dec. 2013  Driven Geometric Phase Gates with Trapped Ions
Poster, iQSim Workshop on Quantum Simulations with Trapped Ions, 16/12/13-19/12/13,
Brighton, UK.
June 2013 *Driven Geometric Phase Gates with Trapped Ions*
Poster, International Conference on Squeezed States and Uncertainty Relations (ICSSUR), 24/06/13-28/06/13, Nürnberg, Germany.

Mar. 2013 *Driven single-sideband geometric phase gates with trapped ions*
Talk, Frühjahrstagung der Deutschen Physikalischen Gesellschaft, 18/03/13-22/03/13, Hannover, Germany, 18/03/2013.

Nov. 2012 *Fast and robust trapped-ion quantum logic gates by a single nearly-resonant sideband excitation*

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**Seminars**

July 2016 *Simulating spin-boson models with trapped ions*
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Erklärung wissenschaftlicher Redlichkeit

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Ulm, den