



The Imperial College of Science, Technology and Medicine

Department of Physics

Sideband Cooling of Ion Coulomb Crystals in a Penning Trap

Vincent Pierre Yves Jarlaud

Submitted in part fulfilment of the requirements for the degree of
Doctor of Philosophy in Physics of Imperial College London.

May 2018

Abstract

This thesis reports on the progress of resolved sideband cooling on the $S_{1/2} \longleftrightarrow D_{5/2}$ transition of $^{40}\text{Ca}^+$ ions in a Penning trap. We demonstrate cooling of the axial motion of a single ion to the ground state over a wide range of trapping frequencies (67 kHz to 420 kHz) with a mean motional state ranging from $\bar{n}_z = 0.07(1)$ to $\bar{n}_z = 0.015(18)$. We are also able to cool the radial motion of the ion close to the ground state with mean motional states below one for both the modified cyclotron and magnetron modes. In order to carry out sideband cooling of the radial mode, the ion is initially cooled with Doppler cooling in the presence of an axialisation field. Efficient sideband cooling outside the Lamb-Dicke regime is performed using complex cooling sequences featuring laser pulses at different frequencies.

The sideband cooling technique is extended to one- and two- dimensional ion Coulomb crystals. For a two-ion chain, aligned with the magnetic field, we measure for the centre of mass mode a final mean motional state $\bar{n}_c = 0.25(6)$ and for the breathing mode $\bar{n}_b = 0.07(4)$ with respective heating rates of $11(2)\text{ s}^{-1}$ and $1(1)\text{ s}^{-1}$. Near ground state cooling of the transverse (axial) modes of two-dimensional planar crystals made of up to 10 ions is achieved. A coherent drive of the cold ions is demonstrated by the observation of Rabi oscillations.

Ramsey experiments are performed on a single ion to study the coherence of the system. We find a $1/e$ coherence time of the optical transition $S_{1/2} \longleftrightarrow D_{5/2}$ equal to $1.76(7)$ ms. This was increased to $13.2(6)$ ms using Uhrig dynamical decoupling. By creating superpositions of the motional states of the ion, we measure the coherence time of the axial motion. A maximum of $565(21)$ ms is found at 420 kHz.

Keywords: axialization, calcium, coherence, Doppler cooling, ion Coulomb crystals, Penning trap, Ramsey experiment, sideband cooling.

Declaration of Originality

This thesis is my own work. When material and contributions from others are used, the sources are acknowledged and referenced.

Copyright Declaration

The copyright of this thesis rests with the author and is made available under a Creative Commons Attribution Non-Commercial No Derivatives licence. Researchers are free to copy, distribute or transmit the thesis on the condition that they attribute it, that they do not use it for commercial purposes and that they do not alter, transform or build upon it. For any reuse or redistribution, researchers must make clear to others the licence terms of this work.

Acknowledgements

I would like to thank my supervisors Richard Thompson and Danny Segal for welcoming me into their group and for their guidance and support during my PhD. It has been a pleasure for me to work at the Ion Trapping Group over the past few years and I owe it largely to them for creating a friendly working environment. Unfortunately, Danny got ill shortly after I joined the group and passed away about a year after. Nevertheless he continued to be involved in our group for as long as he could. He was a truly nice person and was passionate about his work. I am very grateful to Richard for being available throughout my PhD despite a busy schedule and for the abundance of comments on my thesis.

Thank you to my labmates and fellow PhD students: Ollie Corfield, Joe Goodwin, Pavel Hrmo, Manoj Joshi and Graham Stutter. They were all very nice to work with and I learned a lot from them. I must thank in particular Pavel and Manoj who joined our group at the same time as I and with whom I spent a significant amount of time in – and outside – the lab. Our discussions – sometimes agitated – were generally fruitful and I am glad we were able to work together in a good atmosphere. More generally I am grateful to the PhD students and postdocs of the Quantum Optics and Laser Science group for making my stay at Imperial College enjoyable. I should also thank the members of the Centre for Cold Matter for useful advice about experimental techniques and for beneficial trades of equipment.

Thanks to Brian Willey in the workshop who brought his expertise in the development of many of our home-built equipments and for ensuring the safe operation of the laboratory.

I extend my thanks to the administrative staff of the QOLS group: Judith, Marcia and Sanja who helped me navigate the troubled waters of the administration and to Doug Plato for coordinating the FQT programme.

My PhD was part of the doctoral programme Frontiers in Quantum Technology funded by the European Commission under the Marie Skłodowska-Curie Actions. In this framework, I had the opportunity to spend a few months at the GSI Helmholtz-

trum für Schwerionenforschung in Darmstadt as a visiting student. Thank you to Wolfgang Quint, Manuel Vogel and the other members of the Atomic Physics group for receiving me there.

Last, I would like to thank my parents for their continuous support throughout my studies.

Contents

I	Introduction	10
II	Mechanical Properties of Ions in a Penning Trap	15
II.1	Classical Motion of a Single Ion	17
II.2	Quantum Description	20
II.3	Ion Coulomb Crystals	24
	Ion chains	25
	Planar crystals	27
	Quantum description	36
III	Interaction of Light with a Two-Level System	37
III.1	Free Atom	37
III.2	Trapped Ion	40
	Axial direction	41
	Radial direction	46
	Coulomb crystals	49
IV	Theory of Laser Cooling in a Penning Trap	51
IV.1	The Energy Structure of Singly Ionised Calcium-40	51
	Zeeman effect	52
	j -mixing	54
IV.2	Doppler Cooling	55
	Doppler cooling of $^{40}\text{Ca}^+$	60
IV.3	Axialisation	61
IV.4	Optical Resolved Sideband Cooling	67
	Principle	67
	Sideband cooling with a three-level ion	67
	Cooling limit in the Lamb-Dicke regime for $^{40}\text{Ca}^+$	69

	Sideband cooling outside the Lamb-Dicke regime	74
V	Apparatus and Experimental Techniques	76
V.1	The Penning Trap at Imperial College	76
V.2	Doppler Cooling Lasers	77
V.3	Spectroscopy Laser and Associated Devices	81
	Narrow linewidth spectroscopy laser	81
	Intensity stabilisation (noise eater)	82
	Tunable AOM driver (direct digital synthesiser)	83
V.4	Other Optical Systems	87
V.5	Control Systems	89
V.6	Resolved Sideband Spectroscopy	91
	Windowing	92
	Interleaved spectra	93
	State determination	93
	Spectroscopy on Coulomb crystals	96
V.7	General Maintenance	97
VI	Sideband Cooling of the Axial and Radial Motional Modes of a Single Ion	98
VI.1	Axial Cooling	98
	Doppler cooling	98
	Continuous and pulsed sideband cooling	101
	Single-stage SBC	104
	Multiple-stage SBC	106
	Adiabatic cooling	110
	Rabi oscillations	115
VI.2	Radial Cooling	116
	Doppler cooling	116
	Radial SBC	120
VII	Sideband Cooling of Ion Coulomb Crystals	123
VII.1	Sideband Cooling of a Two-Ion Chain	123
VII.2	Planar Crystals	132
	Two-ion planar crystal	133
	Larger crystals	140
VIII	Coherence Studies on a Single Ion	149
VIII.1	Optical Coherence	150
	Ramsey experiments	150
	Dynamical decoupling	155

VIII.2	Motional Coherence	159
	Superpositions near the ground state	159
	Phonon echo	164
	Superposition of three states	167
VIII.3	Coherent Manipulations in High Phonon States	170
	Sideband heating	170
	Superpositions of high phonon states	174
VIII.4	Coherent States	179
IX	Conclusion	185
	Appendices	197
A	Mathematics	198
	A.1 Classical Motion of a Single Ion	198
	A.2 Quantum Description	200
	A.3 Ion Coulomb Crystals	204
	A.4 Motional Superposition	206
B	Planar Coulomb Crystals	209
	B.1 Equilibrium Positions of Planar ICC	209
	B.2 Axial Modes of Planar ICC	211
C	Electronic Circuits	216

I

Introduction

Whereas in the macroscopic world we are used to, we only interact with large ensembles of atoms and molecules, ion traps offer the rare opportunity to observe and manipulate individual particles. Consequently, they have played an important part in experimental quantum physics since their initial development in the late 1950's. Ion traps use electric and magnetic fields to confine charged particles in all dimensions of space, usually in an ultra high vacuum. Two types of ion traps can be distinguished: Paul traps which use an oscillating electric field to confine charged particles and Penning traps which use static electric and magnetic fields. The Paul trap, also referred to as radio-frequency trap was named after its inventor Wolfgang Paul. The name Penning trap was coined by Hans Georg Dehmelt, who built the first trap of this type, in reference to the Dutch physicist Frans Michel Penning for his work on electric discharges. Both Paul and Dehmelt were awarded the Nobel prize in physics in 1989¹ “for the development of the ion trap technique” [1].

As versatile platforms, ion traps have notably found applications in the fields of quantum computing [2], quantum simulation [3, 4], metrology [5, 6] and fundamental physics [7, 8, 9]. A great variety of particles are used in ion trapping experiments: electrons [10], atomic and molecular ions [11] as well as more exotic highly charged ions [12, 13] and antiparticles [14]. Trapped charged particles, depending on their nature, may be cooled through different techniques down to very low temperatures. With laser cooling, certain species of atomic ions can be cooled to temperatures equal to a small fraction of a kelvin above the absolute zero. Cold trapped ions are an excellent system for experiments that require a high degree of control and precision as well as a strong isolation from the environment.

At the Ion Trapping Group of Imperial College London, we focus on developing laser cooling techniques in Penning traps and performing simple coherent manipulations

¹Paul and Dehmelt were awarded one half of the prize. The other half was awarded to the American physicist Norman F. Ramsey “for the invention of the separated oscillatory fields method and its use in the hydrogen maser and other atomic clocks”.

of the trapped ions' electronic and motional states via laser excitations. This will be the object of this thesis.

When the electric potential energy of the ions greatly exceeds their thermal energy, they form a crystalline structure called an ion Coulomb crystal (ICC). This condition is only met at very low temperatures, exclusively attained with laser cooling [15]. The formation of such crystals is the result of an equilibrium between attractive forces from the ion trap and repulsive forces due to the Coulomb interactions between the ions. In an ICC, like in a crystalline solid, the equilibrium positions of the ions are fixed relatively to each other; however the spacings between the ions are much larger, typically of the order of $10\text{ }\mu\text{m}$ against a few ångströms (10^{-10} m) in solids. Depending on the ion trap and the number of ions present, ICCs can take different spatial arrangements (configurations) in one-, two- or three-dimensional structures corresponding to an energy minimum. The ions' motion in Coulomb crystals is described by collective motional modes akin to those of a solid crystal. In analogy with condensed matter physics, a quantum of excitation of these motional modes is called a 'phonon'. We define the motional ground state as the lowest energy level (zero phonon) of a given mode.

In the context of ion trapping experiments, laser cooling can be broadly divided into two categories: Doppler cooling and sub-Doppler cooling which includes in particular resolved sideband cooling (the technique used at Imperial College) and electromagnetically induced transparency (EIT) cooling. Doppler cooling is the most commonly used technique and is applicable to a variety of trapped ions as well as neutral atoms and molecules. Details will be given in the following chapters. Briefly, Doppler cooling consists in reducing the velocity of the ions (and therefore their temperature) through the absorption of photons from the laser. The laser is tuned to a frequency slightly lower than that of an electric dipole transition such that absorption occurs preferentially when the ions experience a blue Doppler shift, that is when the ions and the photons are propagating in opposite directions and therefore leads to a decrease in the ions' momenta. The minimum temperature reached with Doppler cooling is limited by the recoil experienced by the ions when re-emitting a photon. Only sub-Doppler cooling techniques allow reaching the motional ground state.

Over the past two decades, laser cooled trapped ions have been at the forefront of advancements in quantum information processing (QIP) starting in 1995 with the proposal by Juan Ignacio Cirac and Peter Zoller for a trapped-ion based quantum gate [16]. As of 2018, several research groups throughout the world are working on the development of technologies for trapped-ion quantum computing [17, 18, 19, 20] while spin-off companies have been established with the aim to build a commercial quantum computer [21, 22].

In the original scheme of Cirac and Zoller, as well as in later ones such as the one devised by Mølmer and Sørensen [23], the common motional modes of laser cooled ICCs are used to transmit information between the ions which each carry one quantum bit (qubit) in its internal (electronic) levels. Performing a quantum gate thus necessitates to interact with the electronic and motional states of the ions. This is usually done with laser or microwave radiations. The first experimental realisation of a quantum gate was performed in 1995 at the Ion Storage Group of NIST² using an adaptation of the Cirac-Zoller scheme for a single laser cooled beryllium ion confined in a Paul trap [25]. A few years later, high-fidelity quantum gates with two trapped ions were demonstrated [26, 27]. The majority of QIP experiments have been carried out in radio-frequency traps, in particular in linear Paul traps where ions are aligned in a chain along a zero-field axis. This was for instance the type of trap involved in the proposal of Cirac and Zoller [16]. Linear Paul traps have noteworthy achievements to their credit. In quantum computing, they were employed for the realisation of quantum algorithms [28, 29] and entanglement of up to 20 ions has been shown [30]. They have also been used for quantum simulations, in particular of magnetism and quantum phase transitions [31, 32, 33, 34, 35]. There is however interest in going beyond the one-dimensional ion chains found in linear Paul traps and using multiple-dimension crystals for experiments in quantum computing and simulations, in particular with structures where ions are arranged in two-dimensional lattices [36, 37, 38, 39]. Penning traps are particularly well suited in this case [40] as two-dimensional crystals naturally occur under certain conditions (which we will discuss in this thesis) without the need for a specialised trap with a complex electrode structure. One of their appealing aspects is their ability to trap large and stable crystals with hundreds or thousands of ions. Besides, ions in Penning traps are not affected by the micromotion due to the radio-frequency field used in Paul traps. Micromotion has several detrimental effects including unwanted Doppler shifts and heating of the ions sitting outside of the RF null [41]. Possible applications of Penning traps include simulations of quantum magnetism [3], quantum memories [42, 43] and quantum error correction protocols [44]. However, there have not been many experimental realisations with Penning traps in QIP with the notable exception of John Bollinger’s group at NIST [3, 45]. This can be explained in part by the somewhat higher complexity and cost of Penning trap systems over RF traps, not least in terms of laser cooling. Consequently, laser cooling and coherent control techniques in Penning traps have not benefited from the same development as they did in Paul traps. Cooling to the motional ground state of a single ion was only achieved recently, at Imperial College [46] whereas it was demonstrated more

²National Institute of Standards and Technology in Boulder, Colorado, USA. David J. Wineland of NIST was awarded the 2012 Nobel prize in Physics, jointly with Serge Haroche, “for groundbreaking experimental methods that enable measuring and manipulation of individual quantum systems” [24].

than twenty years ago in Paul traps [47]. Difficulties in performing laser cooling have been regarded as a significant obstacle to the development of quantum information in Penning traps [36, 43].

Aside from quantum information, progress in laser cooling techniques should benefit other experiments where Penning traps have been employed. For instance, sympathetic laser cooling has been suggested to cool trapped protons and antiprotons to temperatures in the millikelvin regime for the baryon antibaryon symmetry experiment (BASE) at CERN [48]. This experiment which aims to measure the g -factor of protons and antiprotons would be improved by laser cooling as a reduced temperature would signify a higher measurement precision [49]. Sympathetic laser cooling is a process where charged particles that cannot be directly laser cooled – due to a lack of suitable electronic transition – are trapped together with ions that can, typically alkaline earth metals (e.g. $^9\text{Be}^+$, $^{24}\text{Mg}^+$, $^{40}\text{Ca}^+$). Owing to the Coulomb interactions between the ions and the other charged particles, the latter will thermalise with the cooled ions. Experiments with highly charged ions (HCI) can also take advantage of this sympathetic laser cooling. HCI have applications in atomic clocks [50] and fundamental physics [51], for instance to perform tests of quantum electrodynamics [12]. These experiments rely on high precision spectroscopy and thus benefit greatly from laser cooling as it reduces dramatically the line broadening due to the Doppler effect. Sympathetic cooling of HCI has been achieved in a linear Paul trap [13] and is under development for Penning traps [52, 53].

This thesis consists of three parts. First, chapters II, III and IV introduce the theoretical background necessary to understand the experiments conducted at Imperial College. In chapter II, the motion of ions in Penning traps is studied both with a classical and a quantum treatment. Chapters III and IV explain the interaction of an atomic ion with laser light and the theory of Doppler cooling and resolved sideband cooling in the Penning trap, applied in particular to calcium ions. Then, for a second part, the experimental set-up of the Ion Trapping Group is presented in chapter V. Modifications and improvements made since September 2014, the time I joined the group, will be stressed. Most of the set-up had been built before but an overview of the whole system will be given nevertheless. This chapter also describes the laser spectroscopy technique used in many of our experiments. In the last part comprising chapters VI, VII and VIII, the experimental results are discussed. Chapter VI concerns laser cooling of a single ion; the methods employed to obtain near ground-state cooling of an ion's motional modes held in a Penning trap are explained. Chapter VII presents the extension of the laser cooling techniques to multiple ions ordered in one- and two-dimensional Coulomb crystals. In chapter VIII, we describe experiments of coherent manipulation performed to study the optical and motional coherences of a single trapped ion. It should be noted that

the results presented in this thesis are the product of a common effort with other members of the group. In particular, the majority of the experiments presented here were carried out together by Pavel Hrmo, Manoj Joshi and myself.

I personally took part in experiments of laser cooling of a single ion, both for the axial and radial modes (see chapter VI). I led the effort in performing sideband cooling of the larger (4 ions and more) two-dimensional planar ion Coulomb crystals and contributed to the conception and realisation of coherent control experiments. All the experimental data presented here were analysed by me, often in collaboration with my colleagues, unless said otherwise. Concerning the experimental set-up, my most significant contribution is the development of a new radio-frequency source discussed in chapter V. I also brought more minor changes which I refer to in this thesis, including the installation of an additional acousto-optic modulator and the design and building of an electronic circuit used for adiabatic cooling (see chapter VI). Our research led to a recent publication in a peer-reviewed journal which I co-authored [54].

The research leading to these results has received funding from the People Programme (Marie Curie Actions) of the European Union's Seventh Framework Programme (FP7/2007-2013) under REA grant agreement number 31723.

This thesis includes corrections made after the viva that took place on 24 July 2018, following the comments of the examiners.

II

Mechanical Properties of Ions in a Penning Trap

Due to their charge, ions are subject to the Lorentz force when in the presence of an electromagnetic field and it is therefore relatively easy to control their motion and trap them. Confinement requires the ions to be at a minimum of potential energy such that any displacement from this minimum results in a restoring force that drives the ions back towards the centre of the trap, i.e. at the point of the minimum potential. It is however impossible to create such a potential in all dimensions of space solely with a static electric field. Indeed, assuming the zero point of the potential ϕ (we can always chose the minimum potential to be zero since potentials are defined relatively) is at the Cartesian coordinates $(x,y,z) = (0,0,0)$ the components of the restoring force, for a singly charged cation, are:

$$\begin{aligned} F_x &= -e \frac{\partial \phi}{\partial x} = -e\alpha x \\ F_y &= -e \frac{\partial \phi}{\partial y} = -e\beta y \\ F_z &= -e \frac{\partial \phi}{\partial z} = -e\gamma z \end{aligned} \tag{1}$$

where α , β and γ are positive constants and e the elementary charge. This leads to an electric potential:

$$\phi = \frac{1}{2}(\alpha x^2 + \beta y^2 + \gamma z^2). \tag{2}$$

However, Gauss' law requires $\Delta\phi = 0$ in the absence of charge so $\alpha + \beta + \gamma = 0$ which contradicts that all these constants are positive. This proves that a three dimensional confining electrostatic potential cannot exist. Nevertheless, it is possible to have a trapping force in one or two directions by having at least one of the constants negative. From now on, we shall call the $(x,y,0)$ plane the radial plane and the z axis, the axial direction. Assuming a cylindrical symmetry around the z axis, the

potential now has the form:

$$\phi = A(2z^2 - x^2 - y^2). \quad (3)$$

We see from this equation that, for a positive A , there is an attractive force in the axial direction and a repulsive one in the radial plane. From here, there are two approaches to achieve a confinement in all dimensions: the first one consists in changing the sign of A regularly so that the trapping force is alternatively in the axial direction and in the radial plane. Using a radio-frequency signal to generate the electric potential, it is possible to create a situation where all the components of the force on the ions are on average attractive. Paul traps also known as radio-frequency traps use this approach. The second option is to maintain a static electric field (with A positive for positive ions) and add a magnetic field in the axial direction forcing the ions to orbit around it effectively trapping them in the radial plane. This is utilised in Penning traps which will be described in more detail below.

To create a potential of the form of equation 3, the ideal Penning trap has three electrodes: two end caps and a ring electrode shaped as hyperboloids of revolution of two and one sheet respectively. A static voltage is applied between the end caps and the ring electrode creating a quadrupolar electric field within the trap to ensure trapping in the axial direction while a strong magnetic field along the axis joining the centres of the end caps (the z axis) maintains radial confinement. The resulting electric potential is:

$$\phi = \frac{U}{R^2}(2z^2 - x^2 - y^2) \quad (4)$$

where U is the voltage applied between the electrodes and R a geometrical factor given by $R^2 = 2z_0^2 + r_0^2$, z_0 and r_0 being the distances between the centre of the trap and the end caps and the ring electrode respectively as depicted in figure II.1. The corresponding electric field is:

$$\vec{E} = \frac{2U}{R^2} \begin{pmatrix} x \\ y \\ -2z \end{pmatrix} \quad (5)$$

and the magnetic field:

$$\vec{B} = B \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}. \quad (6)$$

For practical reasons such as machining of the electrodes and optical access, real Penning traps rarely look like the one shown in figure II.1. Various geometries for ion

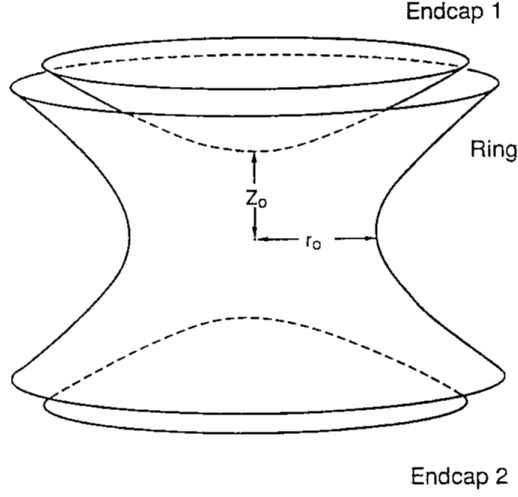


Figure II.1: Electrodes of an ideal Penning Trap. Reproduced from ref. [55].

traps exist: linear, cylindrical, planar [56, 57, 58] and although these geometries will result in a potential different from equation 4, a good harmonicity can be maintained at the vicinity of the trap centre by carefully choosing the dimensions of the trap. The trap used in Imperial College is a hollow cylinder made of a stack of ring electrodes; a description is given in section V.1.

II.1 Classical Motion of a Single Ion

Let us consider the motion of a single ion of charge e and mass m in a Penning trap. We assume that the electric potential has the form given by equation 4 and that the magnetic field is uniform. Derivations are made in the laboratory frame in Cartesian coordinates unless specified otherwise.

The Lorentz force experienced by the ion is:

$$\vec{F} = e(\vec{E} + \vec{v} \times \vec{B}) \quad (7)$$

and Newton's second law gives:

$$\begin{pmatrix} \ddot{x} \\ \ddot{y} \\ \ddot{z} \end{pmatrix} = \frac{2Ue}{mR^2} \begin{pmatrix} x \\ y \\ -2z \end{pmatrix} + \frac{eB}{m} \begin{pmatrix} \dot{x} \\ \dot{y} \\ \dot{z} \end{pmatrix} \times \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \quad (8)$$

which yields:

$$\begin{aligned}\ddot{x} &= \frac{2eU}{mR^2}x + \frac{eB}{m}\dot{y} \\ \ddot{y} &= \frac{2eU}{mR^2}y - \frac{eB}{m}\dot{x} \\ \ddot{z} &= -\frac{4eU}{mR^2}z\end{aligned}\tag{9}$$

For the z axis, solving the differential equation is straightforward as it corresponds to a simple harmonic motion of angular frequency¹:

$$\omega_z = \sqrt{\frac{4eU}{mR^2}}.\tag{10}$$

In the radial plane, the system of coupled differential equations yields two motional modes: the modified cyclotron and the magnetron motions with respective frequencies:

$$\omega_+ = \frac{1}{2}(\omega_c + 2\omega_1)\tag{11}$$

$$\omega_- = \frac{1}{2}(\omega_c - 2\omega_1)\tag{12}$$

where we have defined the true cyclotron frequency:

$$\omega_c = \frac{eB}{m}\tag{13}$$

and introduced $\omega_1 = \frac{1}{2}\sqrt{\omega_c^2 - 2\omega_z^2}$. In the literature, the modified cyclotron and the magnetron frequencies are also denoted ω'_c and ω_m respectively. The derivation of these frequencies can be found in the appendix (section A.1). These frequencies satisfy the following relations:

$$\omega_c = \omega_+ + \omega_-\tag{14}$$

$$\omega_c^2 = \omega_z^2 + \omega_+^2 + \omega_-^2.\tag{15}$$

The above equations are valid only if $\omega_c^2 > 2\omega_z^2$. This is a necessary condition for the stability of the trap.

¹In this thesis, I shall use the Greek letter ν to designate a frequency (expressed in Hz) and the letter ω for an angular frequency (in rad s^{-1}). I will sometimes employ the word *frequency* to refer to the quantity measured in rad s^{-1} without specifying *angular* in the interest of style. Unfortunately, the shorter and non-ambiguous term *pulsatance* is seldom used and seems outmoded.

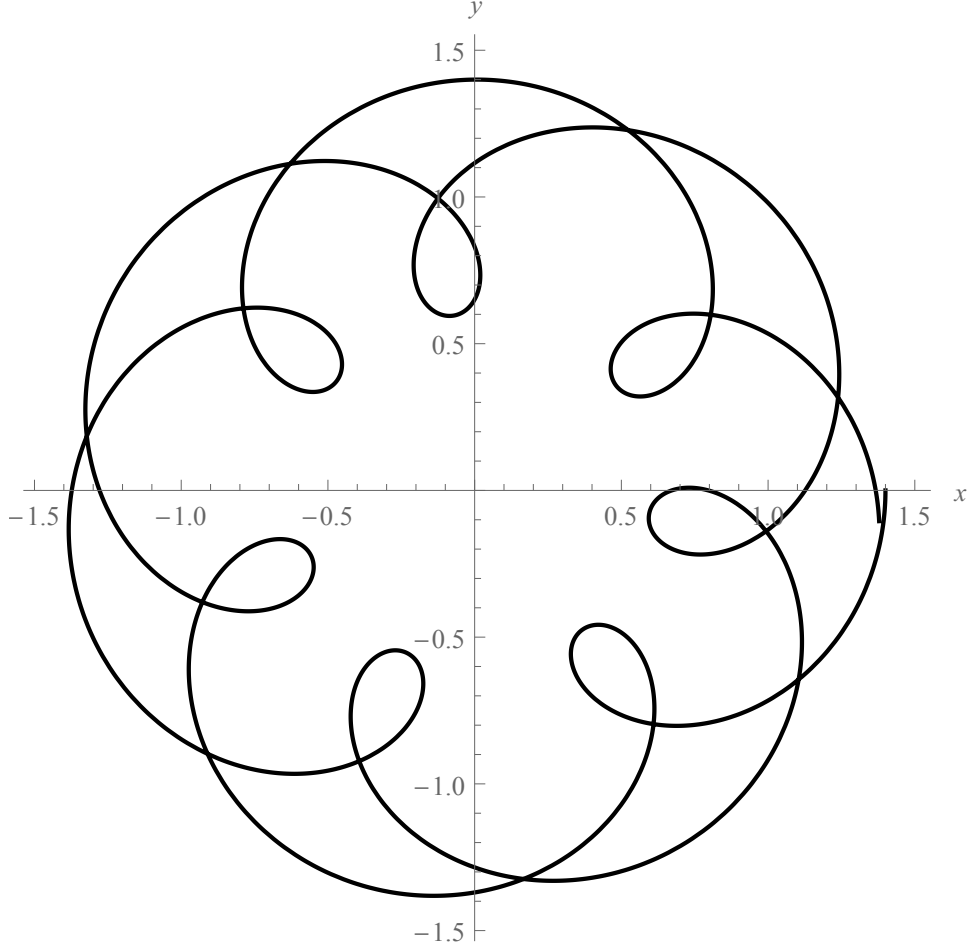


Figure II.2: Radial trajectory of an ion in the radial (x, y) plane. $R_+/R_- = 0.4$, $\nu_z = 400$ kHz, $\nu_c = 729.4$ kHz. These frequencies are typical of the trap used at Imperial College.

Solving equation 9 gives:

$$\begin{aligned}
 x &= R_+ \cos(\omega_+ t + \phi_+) + R_- \cos(\omega_- t + \phi_-) \\
 y &= -R_+ \sin(\omega_+ t + \phi_+) - R_- \sin(\omega_- t + \phi_-) \\
 z &= Z \cos(\omega_z t + \phi_z)
 \end{aligned} \tag{16}$$

where R_+ , R_- and Z are the amplitude of the modified cyclotron, magnetron and axial motions respectively. ϕ_+ , ϕ_- and ϕ_z are phases determined by the initial conditions. The trajectory of the ion in the radial plane is shown in figure II.2.

The Hamiltonian of the system, which represents the total energy, can be calculated by adding the kinetic energy and the potential energy (see appendix section A.1):

$$H = T + V = \frac{1}{2}mv^2 + e\phi \tag{17}$$

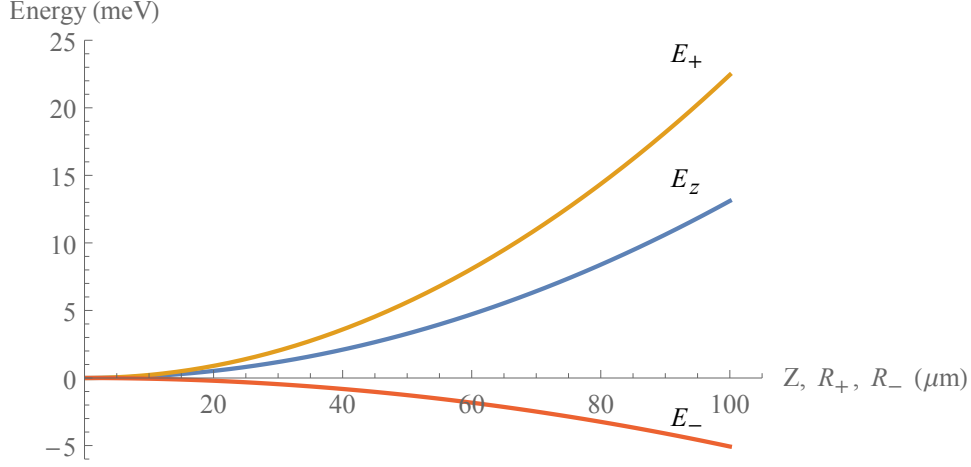


Figure II.3: Energies associated with the axial (E_z), modified cyclotron (E_+) and magnetron (E_-) motions as a function of the motions' amplitude. Calculated for a calcium ion with a cyclotron frequency of 729.4 kHz and an axial frequency of 400 kHz.

which may be split into an axial and a radial component:

$$H_z = \frac{1}{2}m(\dot{z}^2 + \omega_z^2 z^2) \quad (18a)$$

$$H_\rho = \frac{1}{2}m(\dot{\rho}^2 - \frac{1}{2}\omega_z^2 \rho^2) \quad (18b)$$

where $\rho^2 = x^2 + y^2$. Substituting the equations 16 and their time derivatives into the Hamiltonian yields:

$$H = \frac{1}{2}m(\omega_z^2 Z^2 + 2\omega_1(\omega_+ R_+^2 - \omega_- R_-^2)). \quad (19)$$

We note that the contribution of the magnetron motion to the total energy is negative which makes it unstable (see figure II.3). However, the damping time is usually very long [59] so it can be considered as metastable².

II.2 Quantum Description

To understand the motion of an ion at the very low temperatures it reaches after laser cooling, a quantum treatment is necessary. The following is largely based on the work of Brown and Gabrielse [60].

We start from the quantum Hamiltonian:

$$\hat{H} = \frac{(\hat{P} - e\hat{A})^2}{2m} + e\hat{\phi} \quad (20)$$

²In the Penning trap at Imperial College, the lifetime of an ion typically ranges from a few days to a couple of weeks

where $\hat{P}$ is the canonical momentum defined as:

$$\hat{P} = m\hat{v} + e\hat{A}. \quad (21)$$

We use the symmetric gauge for $\hat{A}$ such that:

$$\hat{A} = \frac{B}{2} \begin{pmatrix} -\hat{y} \\ \hat{x} \\ 0 \end{pmatrix}. \quad (22)$$

As discussed above, the motion of the ion in an ideal Penning trap is composed of an axial and a radial motion which are uncoupled. The Hamiltonian can thus be split into two independent components. For the axial motion, this is:

$$H_z = \frac{P_z^2}{2m} + \frac{m\omega_z^2}{2}z^2 \quad (23)$$

The hats on the operators have been dropped for simplicity. We note that for the axial mode, the canonical momentum P_z is equal to the kinetic momentum p_z and so the equation 23 is that of a quantum harmonic oscillator and we can employ the standard ladder operators method. We thus define:

$$a_z = \sqrt{\frac{m\omega_z}{2\hbar}} \left(z + i \frac{p_z}{m\omega_z} \right) \quad (24a)$$

$$a_z^\dagger = \sqrt{\frac{m\omega_z}{2\hbar}} \left(z - i \frac{p_z}{m\omega_z} \right) \quad (24b)$$

The operators a_z and $a_z^\dagger$ are respectively called annihilation and creation operators and the position and momentum operators are rewritten as follows:

$$z = \sqrt{\frac{\hbar}{2m\omega_z}} (a_z^\dagger + a_z) \quad (25a)$$

$$p_z = i\sqrt{\frac{\hbar m\omega_z}{2}} (a_z^\dagger - a_z) \quad (25b)$$

Substituting 25a and 25b into 23 gives:

$$H_z = \hbar\omega_z \left(a_z^\dagger a_z + \frac{1}{2} \right). \quad (26)$$

The associated eigenenergies are:

$$E_z = \hbar\omega_z \left(n_z + \frac{1}{2} \right) \quad , \quad n_z \in \mathbb{N}. \quad (27)$$

For the radial motion, we define the vectors:

$$\vec{V}_+ = \vec{\rho} + \omega_- \hat{z} \times \vec{\rho} \quad (28a)$$

$$\vec{V}_- = \vec{\rho} + \omega_+ \hat{z} \times \vec{\rho} \quad (28b)$$

$\vec{\rho}$ is the coordinate vector in the plane. We can rewrite the radial Hamiltonian (equation 18b) as a function of these vectors (derivation in appendix A.2):

$$H_\rho = \frac{1}{4}m \frac{\omega_+ V_+^2 - \omega_- V_-^2}{\omega_1}. \quad (29)$$

Using equation 21 and the canonical commutation relations between the position and momentum operators, we find (derivation in appendix A.2):

$$[\rho_k, \dot{\rho}_l] = \frac{i\hbar}{m} \delta_{kl} \quad , \quad k, l \in \{x, y\} \quad (30)$$

and:

$$\begin{aligned} [\dot{\rho}_x, \dot{\rho}_x] &= 0 \\ [\dot{\rho}_y, \dot{\rho}_y] &= 0 \\ [\dot{\rho}_x, \dot{\rho}_y] &= i \frac{\hbar \omega_c}{m} \\ [\dot{\rho}_y, \dot{\rho}_x] &= -i \frac{\hbar \omega_c}{m} \end{aligned} \quad (31)$$

which leads to:

$$[V_{+,k}, V_{-,l}] = 0 \quad , \quad k, l \in \{x, y\}. \quad (32)$$

This last commutator shows that the two operators V_+ and V_- are independent. We also have:

$$\begin{aligned} [V_{+,x}, V_{+,y}] &= i \frac{2\hbar \omega_1}{m} \\ [V_{-,x}, V_{-,y}] &= -i \frac{2\hbar \omega_1}{m} \end{aligned} \quad (33)$$

meaning that, to a normalisation factor, $V_{+,x}$ and $V_{+,y}$ as well as $V_{-,x}$ and $V_{-,y}$ are canonically conjugate operators that can be used to build ladder operators. We

therefore construct:

$$a_+ = \sqrt{\frac{m}{4\hbar\omega_1}}(V_{+,x} + iV_{+,y}) \quad (34a)$$

$$a_+^\dagger = \sqrt{\frac{m}{4\hbar\omega_1}}(V_{+,x} - iV_{+,y}) \quad (34b)$$

$$a_- = \sqrt{\frac{m}{4\hbar\omega_1}}(V_{-,x} - iV_{-,y}) \quad (34c)$$

$$a_-^\dagger = \sqrt{\frac{m}{4\hbar\omega_1}}(V_{-,x} + iV_{-,y}) \quad (34d)$$

It can also be useful to express the position operators as a function of the ladder operators:

$$x = -i\sqrt{\frac{\hbar}{4m\omega_1}}(a_+^\dagger - a_+ + a_-^\dagger - a_-) \quad (35a)$$

$$y = \sqrt{\frac{\hbar}{4m\omega_1}}(a_+^\dagger + a_+ - a_-^\dagger - a_-) \quad (35b)$$

With these definitions and the commutator $[a_\pm, a_\pm^\dagger] = 1$, we find:

$$\begin{aligned} V_+^2 &= \frac{2\hbar\omega_1}{m}(2a_+^\dagger a_+ + 1) \\ V_-^2 &= \frac{2\hbar\omega_1}{m}(2a_-^\dagger a_- + 1) \end{aligned} \quad (36)$$

and the radial Hamiltonian (equation 29) becomes:

$$H_\rho = \hbar\omega_+ \left(a_+^\dagger a_+ + \frac{1}{2} \right) - \hbar\omega_- \left(a_-^\dagger a_- + \frac{1}{2} \right) \quad (37)$$

which corresponds to two harmonic oscillators with eigenenergies:

$$\begin{aligned} E_\rho &= E_+ + E_- \\ E_\rho &= \hbar\omega_+ \left(n_+ + \frac{1}{2} \right) - \hbar\omega_- \left(n_- + \frac{1}{2} \right) \quad n_+, n_- \in \mathbb{N}. \end{aligned} \quad (38)$$

The negative contribution of the magnetron motion to the total energy appears here as well and is consistent with the results found in the classical treatment.

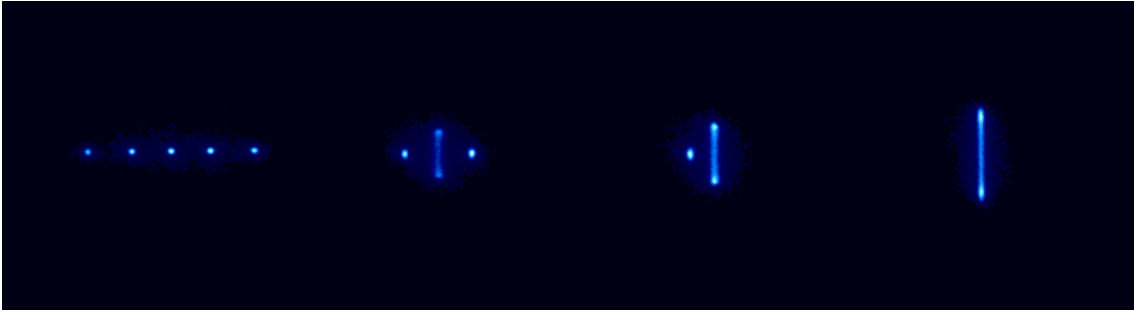


Figure II.4: Pictures of five-ion Coulomb crystals with different configurations. The axial trapping strength (and so the frequency) increases from left to right. Due to the rotation of the crystal, the ions outside the z axis (horizontal) appear as a blurred line. The pictures, taken with an EMCCD camera, are accumulations of thirty individual pictures with a one second exposure.

II.3 Ion Coulomb Crystals

When several trapped ions are at a sufficiently low temperature, they arrange themselves into Coulomb crystals formed by the competitive attractive force of the trap and the repulsion from electrostatic interactions between the ions. In such crystals the ions experience a small motion around a fixed equilibrium position with modes common to all the ions. Depending on various trapping parameters (trapping strength, laser cooling, additional radio-frequency fields, ...) the ions can take different configurations. Of particular interest are the two extreme cases that are the ion chain where all the ions are aligned along the magnetic field axis and the planar crystal where the ions lie in a single plane perpendicular to the magnetic field axis. Between these two, there usually exist a range of three-dimensional configurations that can be taken by the ions (see figure II.4).

In this section we will only describe the chain and the planar crystal and focus in particular on the latter as the chain configuration has already been extensively studied in the context of linear Paul traps [61]. We will seek to determine the equilibrium positions of the ions as well as the normal axial modes.

Let us consider N identical singly charged ions of mass m . The Lagrangian in the laboratory frame is [62]:

$$\begin{aligned}\mathcal{L} &= \mathcal{T} - \mathcal{V} \\ \mathcal{L} &= \sum_{j=1}^N \frac{1}{2} m |\dot{\vec{r}}_j|^2 - e\phi_j + e\vec{A}_j \cdot \vec{r}_j\end{aligned}\tag{39}$$

where $\vec{A}_j$ is the vector potential for the ion j still defined with the symmetric gauge, $\vec{r}_j = (x_j, y_j, r_j)$ the vector position and $e\phi_j$ the total electrostatic potential for the ion j . This potential comprises a term coming from the trapping potential and one

from the Coulomb interactions between ions:

$$e\phi_j = \frac{1}{4}m\omega_z^2(2z_j^2 - \rho_j^2) + \frac{e^2}{8\pi\epsilon_0} \sum_{k=1, k \neq j}^N \frac{1}{r_{jk}} \quad (40)$$

with $r_{jk} = |\vec{r}_k - \vec{r}_j|$.

Ion chains

Ion chains are a platform of choice for quantum information experiments [16, 27, 26] and are usually found in linear Paul traps. They can however be formed in Penning traps as well if the axial trapping strength is low enough. We therefore consider a chain of N ions and assume that the ions always remain aligned with the magnetic field such that their radial motion is not affected by the Coulomb interaction they exert on each other. Given these assumptions, the x and y coordinates are zero for all ions in the Cartesian system defined in the previous sections. This situation is analogous to that of a linear Paul trap and the following is based on the work of Daniel F. V. James [61]. Here, the Lagrangian (equation 39) simplifies to:

$$\mathcal{L} = \sum_{j=1}^N \frac{1}{2}m\dot{z}_j^2 - \frac{1}{2}m\omega_z^2 z_j^2 + \frac{e^2}{8\pi\epsilon_0} \sum_{k=1, k \neq j}^N \frac{1}{|z_k - z_j|}. \quad (41)$$

Let us first consider the simplest case of a two-ion chain. The total potential energy is:

$$\mathcal{V}_2 = \frac{1}{2}m\omega_z^2(z_1^2 + z_2^2) + \frac{e^2}{4\pi\epsilon_0} \frac{1}{|z_1 - z_2|}. \quad (42)$$

The equilibrium positions (z_j^0) are found by equating the partial derivatives of the potential energy with respect to the ions positions to zero:

$$\left[\frac{\partial \mathcal{V}_2}{\partial z_j} \right]_{z_j=z_j^0} = 0 \quad , \quad j \in \{1, 2\}. \quad (43)$$

Differentiating with respect to z_1 gives:

$$(z_1^0)^3 - 2(z_1^0)^2 z_2^0 + z_1^0 (z_2^0)^2 = \frac{e^2}{4\pi\epsilon_0 m\omega_z^2}. \quad (44)$$

By symmetry, $z_1^0 = -z_2^0$, so

$$4(z_1^0)^3 = \frac{e^2}{4\pi\epsilon_0 m\omega_z^2} \quad (45)$$

and finally:

$$z_j^0 = \pm \left(\frac{1}{2} \right)^{2/3} \left(\frac{e^2}{4\pi\epsilon_0 m \omega_z^2} \right)^{1/3}. \quad (46)$$

We introduce the length scale ℓ_z defined by

$$\ell_z = \left(\frac{e^2}{4\pi\epsilon_0 m \omega_z^2} \right)^{1/3} \quad (47)$$

and the dimensionless parameter $w_j = z_j/\ell$. In the general case for N ions, we can rewrite the equilibrium condition:

$$\begin{aligned} \left[\frac{\partial \mathcal{V}}{\partial z_j} \right]_{z_j=z_j^0} &= 0 \quad , \quad \forall j \\ \Leftrightarrow \quad w_j - \sum_{k=1, k \neq j}^N \frac{1}{(w_k - w_j)^2} &= 0 \quad , \quad \forall j \end{aligned} \quad (48)$$

This system of N equations can be solved numerically to determine the equilibrium positions.

Assuming we have found the equilibrium positions, we can now calculate the frequencies of the normal modes. We consider that the amplitude of the motion of the ions is small around their equilibrium point so that we can write $z_j(t) = z_j^0 + q_j(t)$ and approximate the Lagrangian 41 by taking the Taylor expansion of the potential up to the second order:

$$\mathcal{L} = \sum_{j=1}^N \frac{1}{2} m \dot{q}_j^2 - \frac{1}{2} \sum_{j=1}^N \sum_{k=1}^N \left(\frac{\partial^2 \mathcal{V}}{\partial z_j \partial z_k} \right)_0 q_j q_k. \quad (49)$$

Because the expansion is around the equilibrium positions, the first order term is null. Calculating the partial derivative gives:

$$\begin{aligned} \frac{\partial^2 \mathcal{V}}{\partial z_j^2} &= m \omega_z^2 \left(1 + 2 \sum_{k=1, k \neq j}^N \frac{1}{|w_k - w_j|^3} \right) \\ \frac{\partial^2 \mathcal{V}}{\partial z_j \partial z_k} &= -m \omega_z^2 \frac{2}{|w_k - w_j|^3} \quad , \quad j \neq k \end{aligned} \quad (50)$$

We introduce the matrix K^z with elements K_{jk}^z defined by:

$$K_{jk}^z = \begin{cases} \omega_z^2 \left(1 + 2 \sum_{n=1, n \neq j}^N \frac{1}{|w_n - w_j|^3} \right) & \text{if } j = k \\ -\frac{2\omega_z^2}{|w_k - w_j|^3} & \text{if } j \neq k \end{cases} \quad (51)$$

These elements have the dimension of a squared angular frequency. We can rewrite the Lagrangian as follows:

$$\mathcal{L} = \frac{m}{2} \left[\sum_{j=1}^N \dot{q}_j^2 - \sum_{j=1}^N \sum_{k=1}^N K_{jk}^z q_j q_k \right] \quad (52)$$

and finding the frequencies of the normal modes consists in finding the eigenvalues of the matrix K^z [61]. Let μ_n be the N eigenvalues of K^z , the normal frequencies are:

$$\omega_n = \sqrt{\mu_n} \quad , \quad n \in \{1, \dots, N\}. \quad (53)$$

Note that the normal frequencies are all proportional to and larger or equal to ω_z . For two ions, we get ω_z and $\sqrt{3}\omega_z$ with associated eigenvectors $(1, 1)$ and $(1, -1)$ respectively (see appendix A.3). The first mode is the centre of mass (COM) mode where the ions oscillate in phase with a constant spacing between them while in the second mode, the ions oscillate in opposition of phase. This second mode is commonly referred to as the *breathing* or *stretch* mode.

Planar crystals

Due to the rotation of the crystal around the magnetic field axis, the equilibrium positions in a planar crystal are always changing in the laboratory frame. We therefore change the frame of reference to a rotating frame where the equilibrium positions are constant. Let ω_r be the rotation frequency of the crystal assumed constant in the laboratory frame. In the rotating frame, the Lagrangian is transformed to [62]:

$$\mathcal{L}^R = \sum_{j=1}^N \left[\frac{1}{2} m |\dot{r}_j^R|^2 - \frac{eB_{eff}(\omega_r)}{2} (\dot{x}_j^R y_j^R - \dot{y}_j^R x_j^R) - e\phi_j^R \right]. \quad (54)$$

Following the notation of ref. [62], the superscript R denote the rotating frame and the effective magnetic field is defined by $B_{eff}(\omega_r) = B - 2m\omega_r/e$. The potential energy in the rotating frame transforms into:

$$e\phi_j^R = \frac{1}{2} m \left[\omega_z^2 (z_j^R)^2 + \left(\omega_c \omega_r - \omega_r^2 - \frac{\omega_z^2}{2} \right) (\rho_j^R)^2 \right] + \frac{e^2}{8\pi\epsilon_0} \sum_{k=1, k \neq j}^N \frac{1}{r_{jk}^R} \quad (55)$$

and contains two additional terms compared to equation 40 corresponding to the centrifugal and Lorentz forces. We introduce an effective radial frequency

$$\omega_{eff} = \sqrt{\omega_r(\omega_c - \omega_r) - \frac{\omega_z^2}{2}}. \quad (56)$$

This quantity is defined only if the rotation frequency is strictly between ω_- and ω_+ . Consequently, the rotation frequency of a planar crystal is necessarily within these boundaries. We can distinguish three terms in the expression of the potential: an axial trapping term with a characteristic frequency ω_z which is, as one could expect, unchanged from the laboratory frame, a radial trapping term with a characteristic frequency ω_{eff} and a term due to the Coulomb repulsion. Comparing the two first terms gives a measure of the relative axial and radial confinement strengths. For the ions to remain in the radial plane, the axial confinement should be much stronger than the radial so we should have [62]:

$$\beta = \frac{\omega_z^2}{\omega_{eff}^2} \gg 1. \quad (57)$$

This ratio increases with the axial frequency (see figure II.5) and decreases for larger rotation frequencies up to $\omega_c/2$. This means that planar crystals will be stable for large trapping voltages (and so axial frequencies) which is beneficial for laser cooling as we will see in chapter IV. Assuming this condition is satisfied, we look for the equilibrium positions and axial normal modes of a radial crystal in a similar way as done for the ion chain. The two-ion case is here again solvable analytically and the equilibrium coordinates in the rotating frame are³:

$$\begin{aligned} x_1^{R,0} &= x_2^{R,0} = 0 \\ y_1^{R,0} &= -y_2^{R,0} = \left(\frac{1}{2}\right)^{2/3} \left(\frac{e^2}{4\pi\epsilon_0 m \omega_{eff}^2}\right)^{1/3} \\ z_1^{R,0} &= z_2^{R,0} = 0 \end{aligned} \quad (58)$$

We notice the similarity with the two-ion chain and define an analogous radial length scale:

$$\ell_\rho = \left(\frac{e^2}{4\pi\epsilon_0 m \omega_{eff}^2}\right)^{1/3} \quad (59)$$

as well as the dimensionless coordinates $u_j = x_j^R/\ell_\rho$ and $v_j = y_j^R/\ell_\rho$. Figure II.6 shows the equilibrium distance between two ions calculated as a function of the rotation frequency and the effective radial frequency with parameters typical of our experiment. This distance has a minimum on the range $]\omega_-, \omega_+[$ for $\omega_r = \omega_c/2$ where ω_{eff} is maximum.

In general the condition on the total potential $\mathcal{V}$ which is the sum of the potentials

³The coordinate system is chosen so that the x coordinates are zero. Being in the radial plane, the z coordinates are zero by definition.

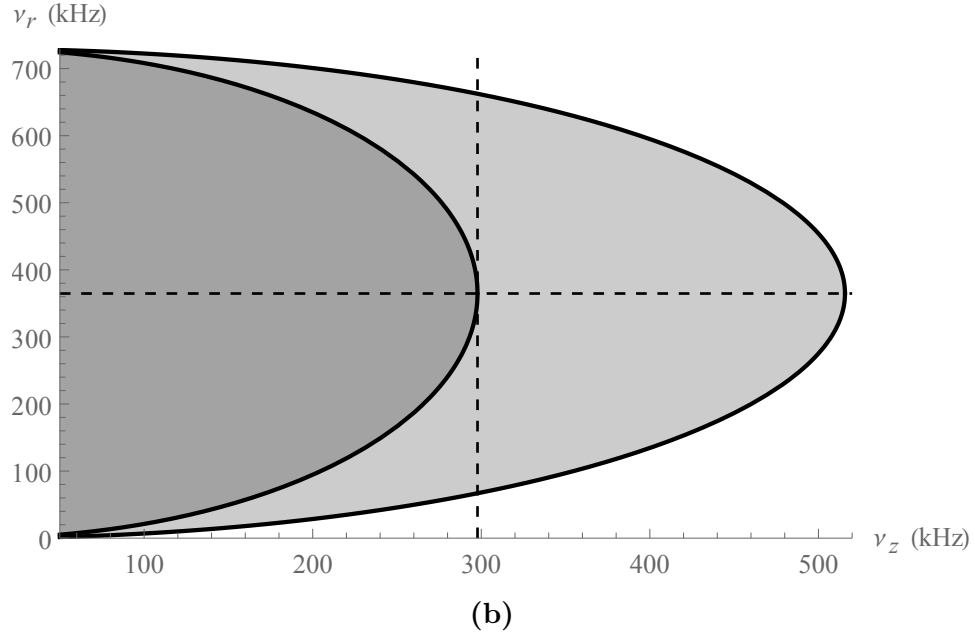
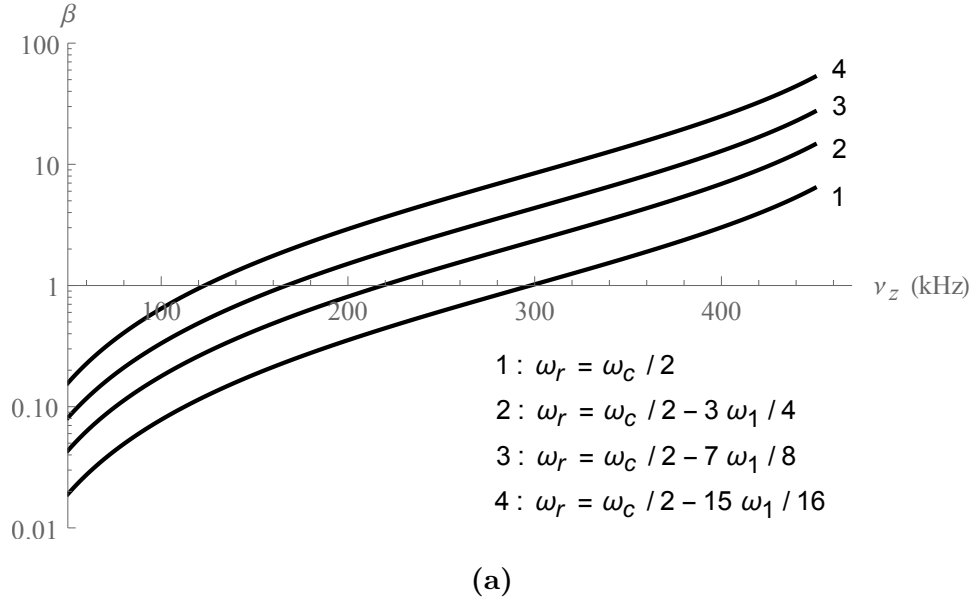


Figure II.5: (a) β as a function of the axial frequency (between 50 and 450 kHz) for different values of the rotation frequency. The planar configuration is more energetically favourable for high axial and low rotation frequencies. Calculated for a cyclotron frequency of 729.4 kHz. (b) Regions of stability of the axial (dark grey) and planar (light grey) configurations of a two-ion crystal. The line between the two regions corresponds to $\beta = 1$, the horizontal dashed line is at $\nu_r = \nu_c/2$ and the vertical one at $\nu_z = \sqrt{\nu_c^2/6}$. The outer solid line, respectively above and below the horizontal line, corresponds to the modified cyclotron and magnetron frequencies as a function of ν_z .

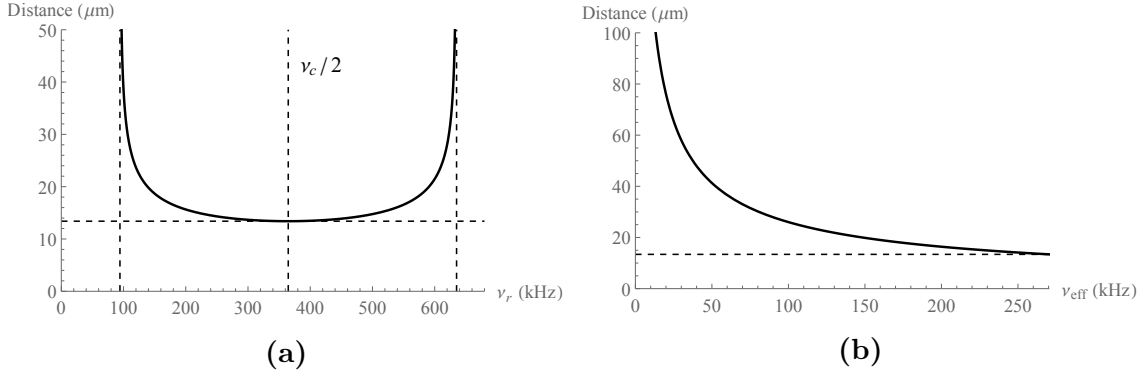


Figure II.6: (a) Inter-ionic distance for a two-ion crystal as a function of the rotation frequency (solid line). Calculated for singly charged calcium ions with a cyclotron frequency of 729.4 kHz and an axial frequency of 346 kHz. The rotation frequency spans the range between the magnetron and the modified cyclotron frequencies (about 94 kHz to 636 kHz, outer dashed vertical lines). The value of the distance diverges at these extremities. The minimum distance is reached at a rotation frequency equal to $\nu_c/2 = 364.7$ kHz. (b) Inter-ionic distance as a function of the effective radial frequency (solid line). The largest value of ν_{eff} is attained for $\nu_r = \nu_c/2$ which corresponds to the minimal achievable inter-ionic distance (13.4 μm here).

$e\phi_j^R$ for equilibrium can be expressed as:

$$\left[\frac{\partial \mathcal{V}^R}{\partial \lambda_j} \right]_{\lambda_j = \lambda_j^0} = 0 \quad , \quad \forall j, \quad \lambda \in \{x^R, y^R\} \quad (60)$$

which is explicitly, in terms of u_j and v_j (derivation in appendix A.3):

$$\begin{aligned} u_j - \sum_{k=1, k \neq j}^N \frac{u_j - u_k}{((u_j - u_k)^2 + (v_j - v_k)^2)^{3/2}} &= 0 \\ v_j - \sum_{k=1, k \neq j}^N \frac{v_j - v_k}{((u_j - u_k)^2 + (v_j - v_k)^2)^{3/2}} &= 0 \end{aligned} \quad (61)$$

This system can be solved numerically to determine the equilibrium coordinates in the rotating frame. Note that this system has an infinity of solutions because of the rotational invariance of the potential which is not a problem if one is only interested in the positions of the ions relative to each other. We can however set for example $u_1 = 0$ to circumvent this issue; the other coordinates are determined accordingly. This is equivalent to choosing a convenient initial phase for the rotating frame. Numerical solutions for planar crystals of two to seventeen ions are given in appendix B.1. We can also notice that, as for the ion chain, no physical parameter appears in the system of equations 61 which means that the dimensionless equilibrium coordinates and so the crystal configuration do not depend on the trapping parameters. Similarly, we can construct the matrix K^ρ to find the axial normal modes of the

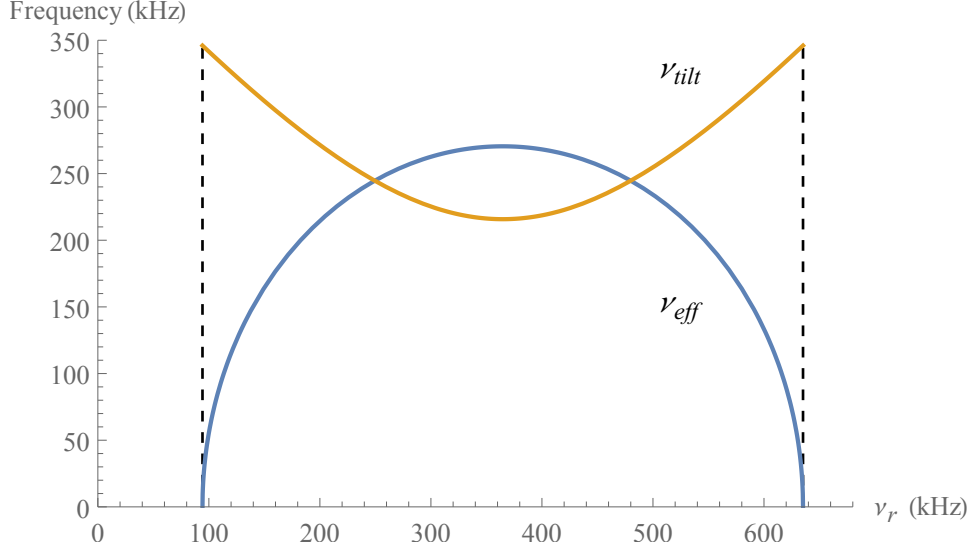


Figure II.7: Effective radial frequency (blue) and tilt frequency (yellow) as a function of the rotation frequency. Calculated for calcium ions with a cyclotron frequency of 729.4 kHz and an axial frequency of 346 kHz.

planar crystal:

$$K_{jk}^\rho = \begin{cases} \omega_z^2 - \omega_{eff}^2 \sum_{n=1, n \neq j}^N \frac{1}{((u_j - u_n)^2 + (v_j - v_n)^2)^{3/2}} & \text{if } j = k \\ \frac{\omega_{eff}^2}{((u_j - u_k)^2 + (v_j - v_k)^2)^{3/2}} & \text{if } j \neq k \end{cases} \quad (62)$$

The eigenvalues can be derived analytically for $N = 2$ and the frequencies of the two axial modes of a two-ion planar crystal are ω_z for the centre of mass mode and ω_{tilt} for the *tilt* mode where

$$\omega_{tilt}^2 = \omega_z^2 - \omega_{eff}^2. \quad (63)$$

The *tilt* mode is the equivalent of the *breathing* mode for the chain i.e. the ions move in opposite directions along the z axis. Figure II.7 shows the tilt and effective frequencies for a two-ion crystal with the same (realistic) parameters as for figure II.6. The tilt frequency reaches a minimum for $\omega_r = \omega_c/2$ which corresponds to a maximum of the effective radial frequency ω_{eff} and a minimum of the inter-ionic distance as seen above. For larger crystals, the eigenvalues of $\mathcal{K}^\rho$ can be calculated numerically. Let us consider the examples of a four and six-ion planar crystal. In equilibrium, the minimum energy configuration (as found numerically) for these crystals are respectively a square and a regular pentagon with an ion in the centre (see table II.1).

For $N = 4$, the matrix $\mathcal{K}^\rho$ has four eigenvalues, two of them are degenerate so the crystal is characterised by three oscillation frequencies. The eigenvectors below are simple enough to understand the motion of the individual ions for the different

Ion	4 ions	6 ions
1	(0 , 0.98549)	(0 , 0)
2	(0.98549 , 0)	(0 , -1.33446)
3	(0 , -0.98549)	(-1.26915 , -0.41237)
4	(-0.98549 , 0)	(-0.78438 , 1.07960)
5	NA	(0.78438 , 1.07960)
6	NA	(1.26915 , -0.41237)

Table II.1: Equilibrium coordinates of a four and a six-ion planar crystal in units of ℓ_p calculated numerically. The numbering of the ions is arbitrary.

modes. We write the μ eigenvector for a N -ion crystal $\vec{b}_\mu^N$, ordered so that the first vector corresponds to the highest eigenvalue and the subsequent ones to increasingly lower eigenvalues. For $N = 4$:

$$\begin{aligned}
\vec{b}_1^4 &= \frac{1}{\sqrt{4}} \begin{pmatrix} 1 \\ 1 \\ 1 \\ 1 \end{pmatrix}, & \vec{b}_2^4 &= \frac{1}{\sqrt{2}} \begin{pmatrix} -1 \\ 0 \\ 1 \\ 0 \end{pmatrix} \\
\vec{b}_3^4 &= \frac{1}{\sqrt{2}} \begin{pmatrix} 0 \\ 1 \\ 0 \\ -1 \end{pmatrix}, & \vec{b}_4^4 &= \frac{1}{\sqrt{4}} \begin{pmatrix} 1 \\ -1 \\ 1 \\ -1 \end{pmatrix}
\end{aligned} \tag{64}$$

We recognise in $\vec{b}_1^4$ the centre of mass modes with a frequency ω_z where all the ions move together along the axial direction. $\vec{b}_2^4$ and $\vec{b}_3^4$ correspond to two degenerate modes where two diagonally opposed ions move in opposite directions while the two others remain stationary in the radial plane. The last mode, at the lowest frequency, corresponds to two diagonally opposed ions moving up while the two others are moving down with an equal amplitude.

In the six ion case, the eigenvectors are:

$$\begin{aligned}
\vec{b}_1^6 &= \frac{1}{\sqrt{6}} \begin{pmatrix} 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \end{pmatrix}, & \vec{b}_2^6 &= \begin{pmatrix} 0 \\ -0.511667 \\ -0.511667 \\ 0.19544 \\ 0.632456 \\ 0.19544 \end{pmatrix}, & \vec{b}_3^6 &= \begin{pmatrix} 0 \\ 0.371748 \\ -0.371748 \\ -0.601501 \\ 0 \\ 0.601501 \end{pmatrix} \\
\vec{b}_4^6 &= \begin{pmatrix} 0 \\ 0.19544 \\ 0.19544 \\ -0.511667 \\ 0.632456 \\ -0.511667 \end{pmatrix}, & \vec{b}_5^6 &= \begin{pmatrix} 0 \\ 0.601501 \\ -0.601501 \\ 0.371748 \\ 0 \\ -0.371748 \end{pmatrix}, & \vec{b}_6^6 &= \frac{1}{\sqrt{30}} \begin{pmatrix} 5 \\ -1 \\ -1 \\ -1 \\ -1 \\ -1 \end{pmatrix}.
\end{aligned} \tag{65}$$

The corresponding modes are represented in figure II.8 (other examples in appendix B.2); the modes 2 and 3 are degenerate and so are the modes 4 and 5. We shall refer to the modes 2 and 3 as the *tilt* modes and to the modes 4 and 5 as the *folding* modes. Unlike the four-ion case, the modes of the six-ion crystals do not accept real eigenvalues over the whole range of rotation frequencies from ω_- to ω_+ . The frequency of mode 6 tends to zero for a certain rotation frequency as can be seen in figure II.9. Above this point, the eigenvalues for $\vec{b}_6^6$ are complex which means they do not correspond to a physical motional frequency and the planar configuration therefore cannot exist. The crystal would then change to a different (three-dimensional) configuration.

More generally, numerical results suggest that for crystals with more than four ions, the lowest frequency mode has a cut-off for a certain rotation frequency lower than $\omega_c/2$. This was observed by calculating the eigenvalues of the modes of crystals of different sizes (up to 17 ions). A higher rotation frequency corresponds to a stronger radial confinement of the crystal as we saw explicitly for two ions. Intuitively, it makes sense that if the radial confinement becomes too strong, the planar configuration becomes unstable. The general pattern is, for rotation frequencies between ω_- and $\omega_c/2$, the frequencies of all the modes decreases with ω_r bar the COM mode which has a constant frequency ω_z .

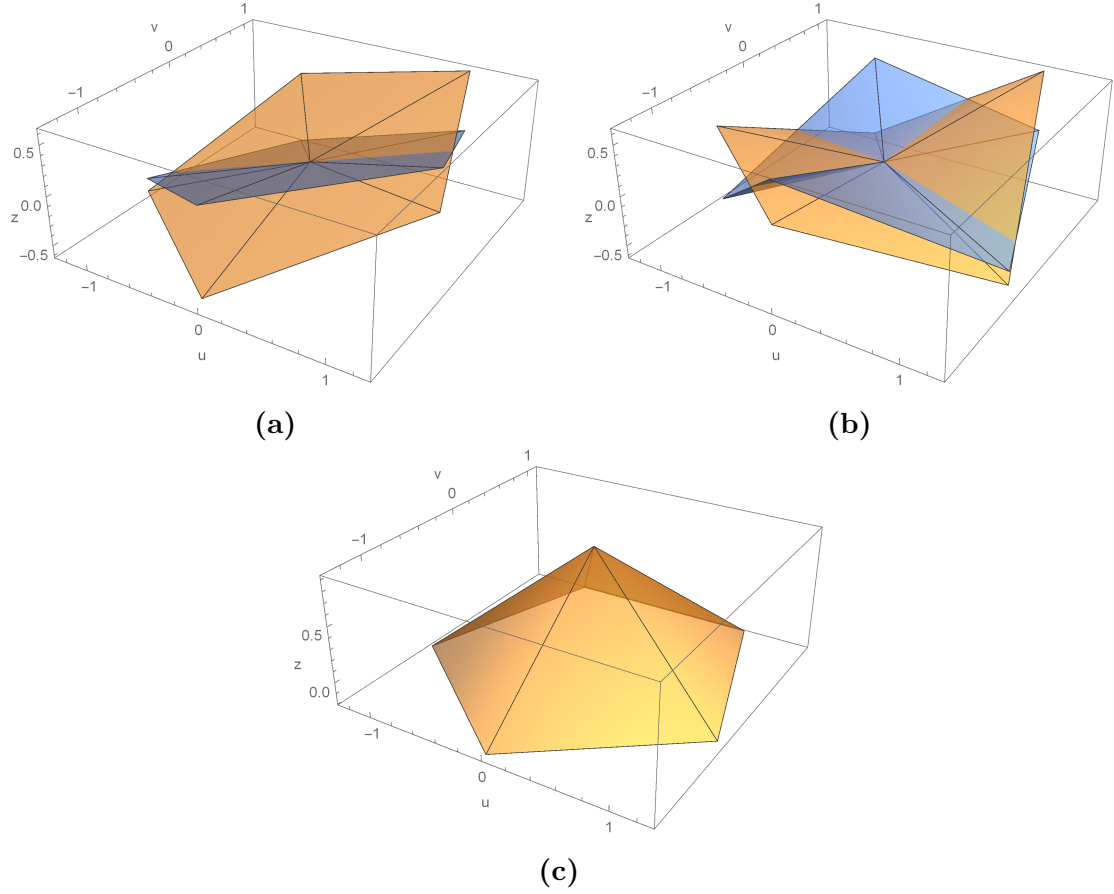
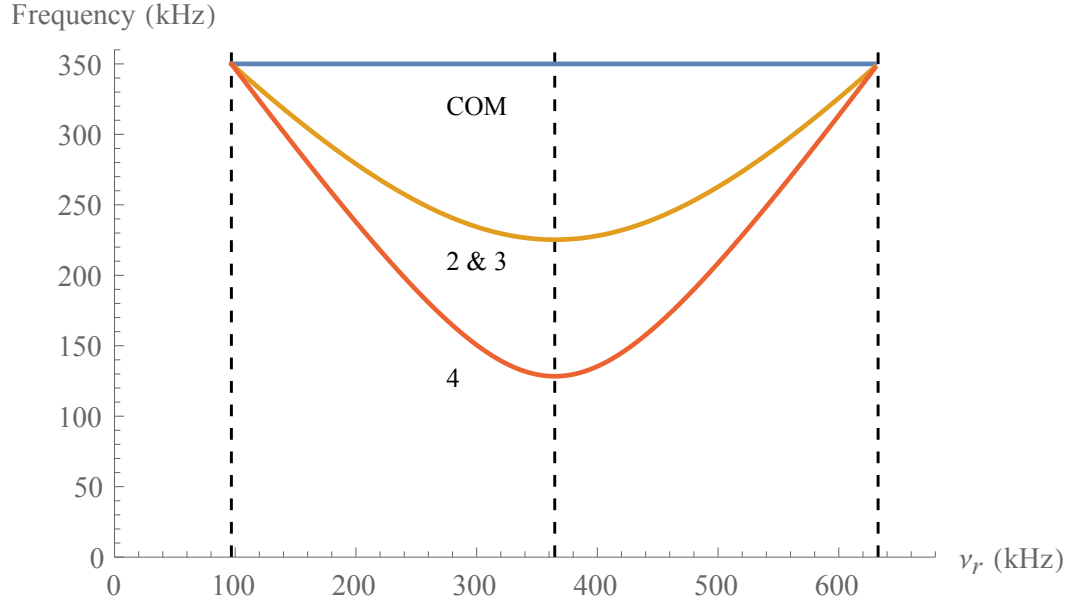
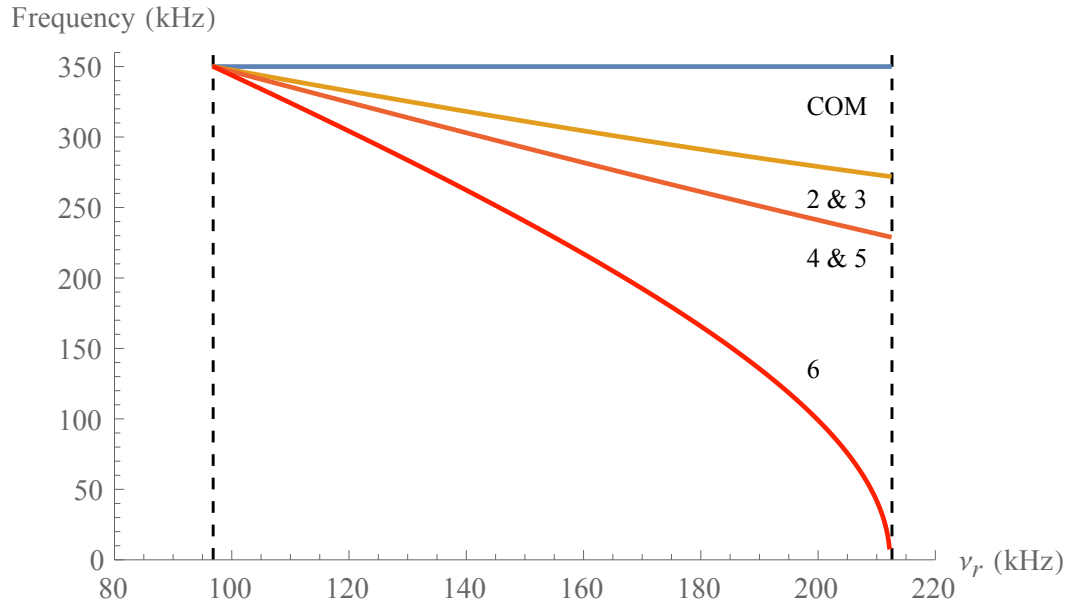


Figure II.8: Representation of the axial modes (except COM) of a six ion planar crystal. The u and v coordinates are taken directly from table II.1 and the z coordinates from the components of the normalised eigenvectors (arbitrary unit). **(a)** Modes 2 and 3 (*tilt*, degenerate). **(b)** Modes 4 and 5 (*folding*, degenerate). **(c)** Mode 6.



(a)



(b)

Figure II.9: (a) Frequency of the axial modes of a four-ion planar crystal as a function of the rotation frequency. The rotation frequency spans the range between the magnetron and the modified cyclotron frequencies (about 94 kHz to 636 kHz). (b) Frequency of the axial modes of a six-ion planar crystal as a function of the rotation frequency. The frequency of mode 6 tends to zero around 212.6 kHz. Plots generated by numerical solving for different rotation frequencies and a fixed axial frequency of 350 kHz.

Quantum description

To the classical normal modes described above correspond quantum phonon modes. A quantum description is especially relevant to treat the interaction of an external field with the motion of the trapped ions. This is the object of the next chapter. The normal modes of the crystal are uncoupled so we can write the (axial) motional Hamiltonian of the N -ion crystal as:

$$H_z^{Xtal} = \sum_{\mu=1}^N \hbar \omega_{\mu} \left(a_{\mu}^{\dagger} a_{\mu} + \frac{1}{2} \right) \quad (66)$$

where the summation is done over the N normal modes. The annihilation and creation operators for the mode μ are given by [62]:

$$a_{\mu} = \sqrt{\frac{m\omega_{\mu}}{2\hbar}} \left(\xi_{\mu} + \frac{iP_{\mu}^z}{m\omega_{\mu}} \right) \quad (67a)$$

$$a_{\mu}^{\dagger} = \sqrt{\frac{m\omega_{\mu}}{2\hbar}} \left(\xi_{\mu} - \frac{iP_{\mu}^z}{m\omega_{\mu}} \right) \quad (67b)$$

with ξ_{μ} and P_{μ}^z the generalised coordinates and momenta such that, for an ion j :

$$z_j^R = \sum_{\mu=1}^N b_{\mu,j}^N \xi_{\mu} \quad , \quad P_{\mu}^z = m \dot{\xi}_{\mu} \quad (68)$$

$b_{\mu,j}^N$ being the j -th component of the eigenvector $\vec{b}_{\mu}^N$. In terms of the ladder operators, the axial coordinate of the ion j is:

$$z_j^R = \sum_{\mu=1}^N b_{\mu,j}^N \sqrt{\frac{\hbar}{2m\omega_{\mu}}} (a_{\mu} + a_{\mu}^{\dagger}). \quad (69)$$

This quantum description is also valid for ion chains.

III

Interaction of Light with a Two-Level System

In this chapter, we use a semi-classical approach to describe the interaction between an atomic particle and a monochromatic electromagnetic radiation, like the one emitted by a narrow linewidth laser. We represent the atom as a quantised two-level system while the laser light is defined by its classical wave description.

III.1 Free Atom

We consider a system of two atomic states denoted $|g\rangle$ (for ground) of energy E_g and $|e\rangle$ (for excited) of energy E_e and a monochromatic laser field of angular frequency ω and phase φ . We define the atomic transition angular frequency ω_0 as:

$$\hbar\omega_0 = E_e - E_g. \quad (70)$$

Taking the zero point of energy between the two levels, the Hamiltonian of the free atom is written:

$$H_0 = \frac{\hbar\omega_0}{2} (|e\rangle\langle e| - |g\rangle\langle g|) = -\frac{\hbar\omega_0}{2}\sigma_z \quad (71)$$

where σ_z is the usual Pauli matrix. Note that we chose the basis $\{|g\rangle, |e\rangle\}$. Using the long wavelength approximation, the electric field of the laser radiation is considered homogeneous over the extent of the atom and evaluated at the nucleus of the atom, at the coordinate $\vec{r}_0$; that is:

$$\vec{E}(t, \vec{r}_0) = \vec{E}(\vec{r}_0) \cos(\omega t + \varphi). \quad (72)$$

The interaction Hamiltonian therefore is:

$$H_i = -\hat{D} \cdot \vec{E}(t, \vec{r}_0), \quad (73)$$

$\hat{D}$ being the dipole operator. We define the Rabi frequency from the matrix element of the interaction Hamiltonian:

$$\Omega_0 = | -\frac{1}{\hbar} \langle e | \hat{D} \cdot \vec{E}(\vec{r}_0) | g \rangle | \quad (74)$$

such that we can rewrite the interaction Hamiltonian as

$$H_i = \hbar \Omega_0 \cos(\omega t + \varphi) \sigma_x. \quad (75)$$

Let $|\psi_S\rangle$ be a general state of the system in the Schrödinger picture i.e. an arbitrary superposition of $|g\rangle$ and $|e\rangle$, $|\psi_S\rangle = c_g(t) |g\rangle + c_e(t) |e\rangle$. The Schrödinger equation is:

$$i\hbar \frac{\partial}{\partial t} |\psi_S\rangle = (H_0 + H_i) |\psi_S\rangle. \quad (76)$$

The most convenient way to solve the Schrödinger equation is to switch to the interaction picture. It becomes:

$$i\hbar \frac{\partial}{\partial t} |\psi_I\rangle = H_I |\psi_I(t)\rangle \quad (77)$$

where the quantum state in the interaction picture is defined by:

$$|\psi_I\rangle = e^{iH_0 t/\hbar} |\psi_S\rangle \quad (78)$$

and the Hamiltonian H_I by:

$$H_I = e^{iH_0 t/\hbar} H_i e^{-iH_0 t/\hbar}. \quad (79)$$

Using the Taylor expansion of the exponential function, we calculate:

$$e^{iH_0 t/\hbar} = \cos\left(\frac{\omega_0 t}{2}\right) \mathbb{1} - i \sin\left(\frac{\omega_0 t}{2}\right) \sigma_z \quad (80)$$

allowing us to express $|\psi_I\rangle$ and H_I more explicitly:

$$\begin{aligned} |\psi_I\rangle &= e^{-i\omega_0 t/2} c_g(t) |g\rangle + e^{i\omega_0 t/2} c_e(t) |e\rangle \\ \Rightarrow |\psi_I\rangle &= \tilde{c}_g(t) |g\rangle + \tilde{c}_e(t) |e\rangle, \end{aligned} \quad (81)$$

with

$$\tilde{c}_g(t) = e^{-i\omega_0 t/2} c_g(t) \quad , \quad \tilde{c}_e(t) = e^{i\omega_0 t/2} c_e(t) \quad (82)$$

and

$$H_I = \hbar\Omega_0 \begin{pmatrix} 0 & \cos(\omega t + \varphi)(\cos(\omega_0 t) - i \sin(\omega_0 t)) \\ \cos(\omega t + \varphi)(\cos(\omega_0 t) + i \sin(\omega_0 t)) & 0 \end{pmatrix}. \quad (83)$$

We now make the rotating wave approximation i.e. we neglect the terms in $\omega + \omega_0$ since they oscillate much more rapidly than the terms in $\omega - \omega_0$ and have a negligible average contribution [63]. We also introduce the laser frequency detuning $\delta = \omega - \omega_0$. The above Hamiltonian simplifies to:

$$H_I = \frac{\hbar\Omega_0}{2} \begin{pmatrix} 0 & e^{i(\delta t + \varphi)} \\ e^{-i(\delta t + \varphi)} & 0 \end{pmatrix}. \quad (84)$$

It is worth noting the presence of the following parameters of the laser radiation in the Hamiltonian: the intensity, through the Rabi frequency, the detuning, in other words the frequency, and the phase. The Schrödinger equation gives the system of coupled equations:

$$\dot{\tilde{c}}_g(t) = -i\frac{\Omega_0}{2}e^{i(\delta t + \varphi)}\tilde{c}_e(t) \quad (85a)$$

$$\dot{\tilde{c}}_e(t) = -i\frac{\Omega_0}{2}e^{-i(\delta t + \varphi)}\tilde{c}_g(t). \quad (85b)$$

The general solution of this system is:

$$\tilde{c}_g(t) = \alpha e^{i(\delta + \Omega)t/2} + \beta e^{i(\delta - \Omega)t/2} \quad (86a)$$

$$\tilde{c}_e(t) = -\alpha \frac{\delta + \Omega}{\Omega_0} e^{-i((\delta - \Omega)t/2 + \varphi)} - \beta \frac{\delta - \Omega}{\Omega_0} e^{-i((\delta + \Omega)t/2 + \varphi)} \quad (86b)$$

where α and β are constants and we have introduced the generalised Rabi frequency $\Omega = \sqrt{\Omega_0^2 + \delta^2}$. In terms of the initial values of $\tilde{c}_g$ and $\tilde{c}_e$, these constants are:

$$\begin{aligned} \alpha &= \frac{1}{2\Omega} \left((\Omega - \delta)\tilde{c}_g(0) - \Omega_0 e^{i\varphi} \tilde{c}_e(0) \right) \\ \beta &= \frac{1}{2\Omega} \left((\Omega + \delta)\tilde{c}_g(0) + \Omega_0 e^{i\varphi} \tilde{c}_e(0) \right) \end{aligned} \quad (87)$$

For the initial conditions $\tilde{c}_g(0) = 1$ and $\tilde{c}_e(0) = 0$ which correspond to the atom initially in the ground state, the solution is:

$$\tilde{c}_g(t) = e^{i\delta t/2} \left[\cos\left(\frac{\Omega t}{2}\right) - i\frac{\delta}{\Omega} \sin\left(\frac{\Omega t}{2}\right) \right] \quad (88a)$$

$$\tilde{c}_e(t) = -ie^{-i(\delta t/2 + \varphi)} \frac{\Omega_0}{\Omega} \sin\left(\frac{\Omega t}{2}\right). \quad (88b)$$

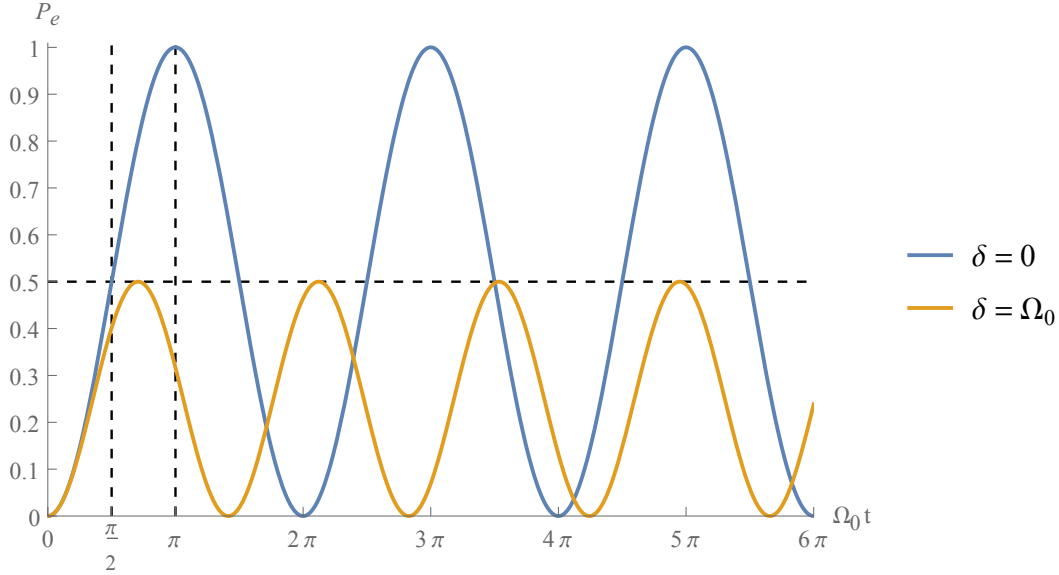


Figure III.1: Probability of excitation as a function of time showing Rabi oscillations on resonance (blue) and for a detuning equal to the Rabi frequency (orange). Rabi frequency normalised to $\Omega_0 = 1$.

The probability of excitation of an atom initially in the ground state by a near-resonant laser radiation is therefore:

$$P_e(t) = |c_e(t)|^2 = \frac{\Omega_0^2}{\Omega^2} \sin^2\left(\frac{\Omega t}{2}\right) \quad (89)$$

The probability of excitation has a sinusoidal evolution with time hence the appearance of Rabi oscillations (see figure III.1). In the resonant case, a pulse of duration π/Ω_0 transfers all the population from the ground to the excited state and is therefore often called a “ π pulse”. A pulse of duration $\pi/2\Omega_0$ creates an equal superposition of $|g\rangle$ and $|e\rangle$. In the spectral domain (figure III.2), for a π pulse, the probability of excitation as a function of the laser detuning is the product of oscillations due to the squared sine and a Lorentzian envelope coming from the prefactor Ω_0^2/Ω^2 . The resulting shape is characteristic of a coherent process.

III.2 Trapped Ion

We now consider a trapped ion interacting with a near resonant laser with only two electronic states for the ion denoted $|g\rangle$ and $|e\rangle$ as previously. Unlike the free atom, considered at rest, we need to take into account the motional Hamiltonian. As a result of the quantum nature of the ion’s motion, the laser light is resonant with the internal levels of the ion only for particular frequencies and not for a continuous Doppler broadened line as one could expect classically. Consequently, the absorption spectrum will be composed of a peak at the atomic transition frequency ω_0 referred to as the carrier and a set of equally spaced fringes called sidebands centred

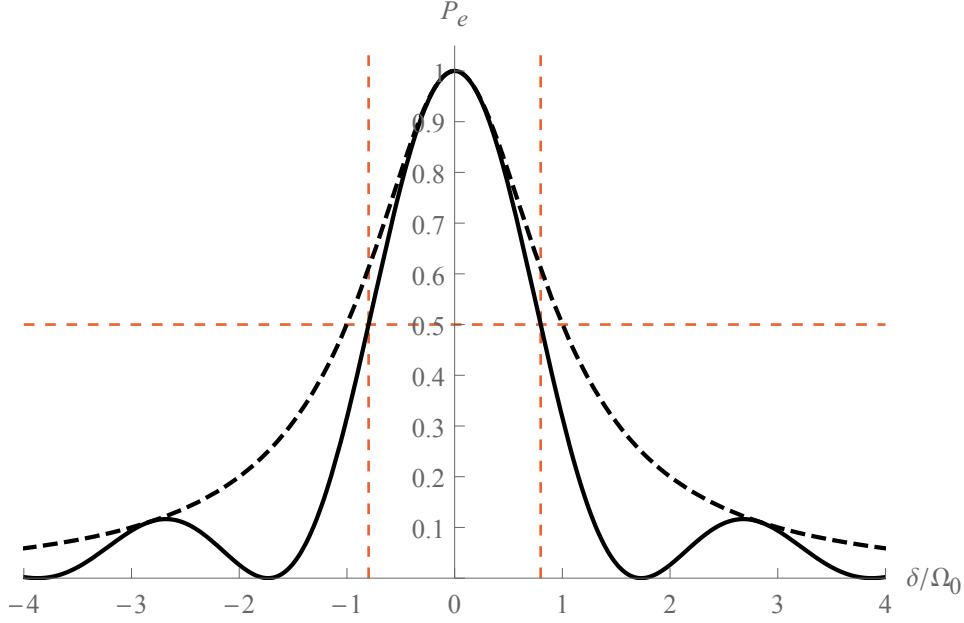


Figure III.2: Probability of excitation for a π pulse as a function the ratio of the laser detuning and the Rabi frequency (solid line) and its Lorentzian envelope (dashed). The full width at half maximum is roughly 1.6.

around the carrier. Intuitively, one can see that the resonant coupling of the laser light with the ion will occur for laser detunings equal to integer multiples of the quantum harmonic oscillator frequency as a consequence of Doppler shift. This will be shown mathematically below.

Axial direction

Let us consider the axial motion of the ion. For the trapped ion the unperturbed Hamiltonian is [60, 63]:

$$\begin{aligned} H'_0 &= H_0 + H_z \\ H'_0 &= -\frac{\hbar\omega_0}{2}\sigma_z + \hbar\omega_z\left(a_z^\dagger a_z + \frac{1}{2}\right) \end{aligned} \quad (90)$$

and the interaction Hamiltonian:

$$H_i = \hbar\Omega_0 \cos(kz - \omega t + \varphi)\sigma_x \quad (91)$$

where we have assumed that the laser field propagates along $\vec{z}$. Let us rewrite this by substituting z with its expression in terms of ladder operators (equation 25a):

$$H_i = \hbar\Omega_0 \cos\left(\sqrt{\frac{\hbar k^2}{2m\omega_z}}(a_z^\dagger + a_z) - \omega t + \varphi\right)\sigma_x. \quad (92)$$

We define the Lamb-Dicke parameter as:

$$\eta = \sqrt{\frac{\hbar k^2}{2m\omega_z}} \quad (93)$$

giving:

$$H_i = \hbar\Omega_0 \cos(\eta(a_z^\dagger + a_z) - \omega t + \varphi) \sigma_x \quad (94)$$

This Hamiltonian shows that the laser field can have an influence on the electronic state of the ion via σ_x as well as its motional state via the ladder operators. The Lamb-Dicke parameter is the product of the light's wavenumber k and the extension along $\vec{z}$ of the ground-state wave function of the harmonic oscillator; it can be seen as a measure of the confinement of the ion with respect to the light's wavelength. Like for the free atom, we look for the Hamiltonian in the interaction picture. Let the propagator $U'_0 = \exp(-iH'_0 t/\hbar)$. The transformed interaction Hamiltonian is given by:

$$H_I = U_0'^\dagger H_i U_0' \quad (95)$$

Because H_0 and σ_x commute with H_z , a_z and $a_z^\dagger$, we can write:

$$H_I = \hbar\Omega_0 e^{iH_0 t/\hbar} \sigma_x e^{-iH_0 t/\hbar} e^{iH_z t/\hbar} \cos(\eta(a_z^\dagger + a_z) - \omega t + \varphi) e^{-iH_z t/\hbar} \quad (96)$$

and one can easily verify, using equation 80, that:

$$e^{iH_0 t/\hbar} \sigma_x e^{-iH_0 t/\hbar} = e^{i\omega_0 t} |e\rangle \langle g| + e^{-i\omega_0 t} |g\rangle \langle e| \quad (97)$$

and so:

$$H_I = \frac{\hbar\Omega_0}{2} \left[e^{i\omega_0 t} |e\rangle \langle g| + e^{-i\omega_0 t} |g\rangle \langle e| \right] \times e^{iH_z t/\hbar} \left[e^{i(\eta(a_z^\dagger + a_z) - \omega t + \varphi)} + e^{-i(\eta(a_z^\dagger + a_z) - \omega t + \varphi)} \right] e^{-iH_z t/\hbar}. \quad (98)$$

The expression of the Hamiltonian above contains time-dependent terms with frequencies $\omega + \omega_0$ and $\omega - \omega_0$. By applying the rotating wave approximation, we retain only the slowly varying terms, leading to:

$$H_I \simeq \frac{\hbar\Omega_0}{2} \left[|e\rangle \langle g| e^{iH_z t/\hbar} e^{i(\eta(a_z^\dagger + a_z) - \delta t + \varphi)} e^{-iH_z t/\hbar} + |g\rangle \langle e| e^{iH_z t/\hbar} e^{-i(\eta(a_z^\dagger + a_z) - \delta t + \varphi)} e^{-iH_z t/\hbar} \right]. \quad (99)$$

Using the transformation for the ladder operators:

$$\tilde{a}_z = a_z e^{-i\omega_z t} \quad , \quad \tilde{a}_z^\dagger = a_z^\dagger e^{i\omega_z t} \quad (100)$$

which is equivalent to a transformation from the Schrödinger picture to the Heisenberg one, we can further write [64]:

$$H_I \simeq \frac{\hbar\Omega_0}{2} \left[|e\rangle \langle g| e^{i(\eta(\tilde{a}_z^\dagger + \tilde{a}_z) - \delta t + \varphi)} + |g\rangle \langle e| e^{-i(\eta(\tilde{a}_z^\dagger + \tilde{a}_z) - \delta t + \varphi)} \right]. \quad (101)$$

Let $|n\rangle$ and $|m\rangle$ be Fock states of the harmonic oscillator. We seek to evaluate the quantity

$$\langle e, m | H_I | g, n \rangle = \frac{\hbar\Omega_0}{2} \langle m | e^{i(\eta(\tilde{a}_z^\dagger + \tilde{a}_z) - \delta t)} | n \rangle. \quad (102)$$

Using the Baker-Campbell-Hausdorff formula (the Zassenhaus one in particular), we can express the exponential term above as:

$$e^{i(\eta(\tilde{a}_z^\dagger + \tilde{a}_z) - \delta t)} = e^{-i\delta t} e^{i\eta\tilde{a}_z^\dagger} e^{i\eta\tilde{a}_z} e^{-[i\eta\tilde{a}_z^\dagger, i\eta\tilde{a}_z]/2}. \quad (103)$$

The commutator can be calculated directly:

$$\begin{aligned} [i\eta\tilde{a}_z^\dagger, i\eta\tilde{a}_z] &= -\eta^2 [a_z^\dagger, a_z] \\ &= \eta^2. \end{aligned} \quad (104)$$

Besides, by writing the Taylor expansion of the exponential, we have

$$\exp(i\eta\tilde{a}_z) |n\rangle = \sum_{k=0}^{\infty} \frac{(i\eta)^k}{k!} e^{-ik\omega_z t} (a_z)^k |n\rangle \quad (105)$$

and

$$(a_z)^k |n\rangle = \begin{cases} \sqrt{\frac{n!}{(n-k)!}} |n-k\rangle & \text{if } k \leq n \\ 0 & \text{if } k > n \end{cases} \quad (106)$$

so

$$\exp(i\eta\tilde{a}_z) |n\rangle = \sum_{k=0}^n \frac{(i\eta)^k}{k!} e^{-ik\omega_z t} \sqrt{\frac{n!}{(n-k)!}} |n-k\rangle. \quad (107)$$

Similarly,

$$\langle m | \exp(i\eta\tilde{a}_z^\dagger) = \sum_{l=0}^m \frac{(i\eta)^l}{l!} e^{il\omega_z t} \sqrt{\frac{m!}{(m-l)!}} \langle m-l|. \quad (108)$$

Using these results, we obtain:

$$\begin{aligned} \langle e, m | H_I | g, n \rangle &= \frac{\hbar\Omega_0}{2} e^{-i\delta t} e^{-\eta^2/2} \sqrt{n!m!} \times \\ &\sum_{k=0}^n \sum_{l=0}^m \frac{(i\eta)^{k+l}}{k!l! \sqrt{(n-k)!} \sqrt{(m-l)!}} e^{i(l-k)\omega_z t} \langle m-l | n-k \rangle. \end{aligned} \quad (109)$$

Considering the orthogonality of the eigenvectors of the harmonic oscillator and introducing the notation: $s = m - n$, $n_{<} = \min\{n, m\}$, $n_{>} = \max\{n, m\}$:

$$\langle e, m | H_I | g, n \rangle = \frac{\hbar\Omega_0}{2} e^{i(s\omega_z - \delta)t} e^{-\eta^2/2} (i\eta)^{|s|} \sqrt{n_{<}! n_{>}!} \sum_{k=0}^{n_{<}} \frac{(-1)^k \eta^{2k}}{k! (k + |s|)! (n_{<} - k)!}. \quad (110)$$

In terms of the generalised Laguerre polynomials defined by

$$L_{n_{<}}^{(|s|)}(\eta^2) = \sum_{k=0}^{n_{<}} (-1)^k \binom{n_{<} + |s|}{n_{<} - k} \frac{\eta^{2k}}{k!}, \quad (111)$$

we have:

$$\langle e, m | H_I | g, n \rangle = \frac{\hbar\Omega_0}{2} e^{i(s\omega_z - \delta)t} e^{-\eta^2/2} (i\eta)^{|s|} \sqrt{\frac{n_{<}!}{n_{>}!}} L_{n_{<}}^{(|s|)}(\eta^2). \quad (112)$$

If the detuning of the laser is such that $\delta = s\omega_z$, the time dependent term in the expression above drops. This situation corresponds to the resonant coupling of the states $|g, n\rangle$ and $|e, n + s\rangle$ by the laser and the strength of this coupling is

$$\Omega_{n, n+s} = \Omega_0 e^{-\eta^2/2} \eta^{|s|} \sqrt{\frac{n_{<}!}{n_{>}!}} L_{n_{<}}^{(|s|)}(\eta^2). \quad (113)$$

Note that this quantity corresponds to and is also referred to as the Rabi frequency of the transition $|g, n\rangle \longleftrightarrow |e, n + s\rangle$. If the linewidth of the laser is sufficiently narrow ($\Delta\omega \ll \omega_z$) and the intensity small enough such that $\Omega_0 \ll \omega_z$, the coupling will be significant only for the transition $|g, n\rangle \longleftrightarrow |e, n + s\rangle$. From these results, it is understood that a resonant coupling occurs only at particular values of laser detuning $\delta = s\omega_z$, with s an integer. If the linewidth of the electronic transition excited by the laser is smaller than the oscillation frequency of the ion, the absorption spectrum of the trapped ion is made of bands, similar to that shown in figure III.2, separated by ω_z and with a width determined by $\Omega_{n, n+s}$ (and the linewidth of the laser). This situation is called the strong-binding regime as opposed to the weak-binding regime where the electronic transition linewidth is larger than the oscillation frequency. In an analogy with the field of signal processing, the central band at zero laser detuning is called the carrier and the other bands for $s \neq 0$ are referred to as

blue sidebands for positive detunings and red sidebands for negative detunings. Another consequence of equation 113 is that, for a given sideband, there are motional state numbers such that $\Omega_{n,n+s}$ tends to zero i.e. the transition $|g, n\rangle \longleftrightarrow |e, n+s\rangle$ is extremely unlikely to occur. This is clear from the mathematical expression of the coupling strength which contains a generalised Laguerre polynomial. To give a physical explanation, Stig Stenholm writes in his 1986 review on laser cooling [65]:

“The rate of transfer between quantum numbers n and m depends on the overlap of two momentum wave functions, one of which is displaced by the photon momentum. The shape of the momentum wave functions of the harmonic oscillator equals the shape of the position wave functions [...]. The argument is very similar to that giving a Franck-Condon factor for the molecular transitions only here it operates in momentum space.”

This has important implications for laser cooling as discussed in section IV.4. The Rabi frequencies for the carrier, first red sideband (RSB, $\delta = -\omega_z$) and first blue sideband (BSB, $\delta = \omega_z$) as a function of the motional state number (Fock state) are plotted in figure III.3.

If the ion is cold enough, i.e. if its average motional quantum number ($\bar{n}$) is low and η sufficiently small, coupling between the laser field and the ion only occurs significantly for a laser detuning of 0, ω_z or $-\omega_z$. The corresponding spectrum would only show a peak at the carrier frequency and the first blue and red sidebands. This defines the Lamb-Dicke regime where the following criterion must be fulfilled:

$$\eta\sqrt{2\bar{n}+1} \ll 1. \quad (114)$$

In this case, the Hamiltonian H_I can be approximated by its Taylor expansion truncated to the first order:

$$H_I \simeq \frac{\hbar\Omega_0}{2} \left((1 + i\eta(a_z^\dagger e^{i\omega_z t} + a_z e^{-i\omega_z t})) e^{-i(\delta t - \varphi)} |e\rangle \langle g| + (1 - i\eta(a_z^\dagger e^{i\omega_z t} + a_z e^{-i\omega_z t})) e^{i(\delta t - \varphi)} |g\rangle \langle e| \right). \quad (115)$$

By setting the laser detuning at the aforementioned resonant values, the Rabi frequencies are:

$$\Omega_{n,n} = \Omega_0 \left(1 - \eta^2 \left(n + \frac{1}{2} \right) \right) \quad \text{for } \delta = 0 \quad (116a)$$

$$\Omega_{n,n-1} = \Omega_0 \eta \sqrt{n} \quad \text{for } \delta = -\omega_z \quad (116b)$$

$$\Omega_{n,n+1} = \Omega_0 \eta \sqrt{n+1} \quad \text{for } \delta = \omega_z \quad (116c)$$

These expressions are only valid for the first motional states and quickly deviate from the correct values of the Rabi frequencies. However they clearly show that the Rabi frequency at $n = 0$ for the first red sideband is zero but $\Omega_0 \eta$ for the blue one.

This asymmetry is used as an indicator to determine whether a trapped ion is in its motional ground state where there is no state to be coupled to with a red detuned laser ($|n\rangle = |-1\rangle$ doesn't exist).

Often, the ion is not in a well defined Fock state but rather in some statistical distribution of different motional states. In this case, if $P(n)$ is the probability of the ion to be in the motional state $|n\rangle$, the average probability of excitation of the electronic transition on a given sideband is:

$$P_{e,s}(t) = |c_e(t)|^2 = \sum_{n=0}^{\infty} P(n) \sin^2 \left(\frac{\Omega_{n,n+s} t}{2} \right) \quad (117)$$

assuming the laser is on resonance with the sideband. A common distribution is the so-called thermal state (which follows a Boltzmann distribution) where the probability of occupation of a given motional state follows:

$$P(n) = \frac{\bar{n}^n}{(\bar{n} + 1)^{n+1}} \quad (118)$$

with $\bar{n}$ the average motional state. Figure III.4 shows the probability of excitation when the laser is tuned to different sideband frequencies for a thermal state and for the motional ground state.

Radial direction

We can follow the same treatment for the radial motion of the ion. We now assume that the laser propagates in the radial plane (along the y axis for example) and so the interaction Hamiltonian in the Schödinger picture is:

$$\begin{aligned} H_i &= \hbar\Omega_0 \cos(ky - \omega t) \\ H_i &= \hbar\Omega_0 \cos \left(k\sqrt{\frac{\hbar}{4m\omega_1}} (a_+^\dagger + a_+ - (a_-^\dagger + a_-)) - \omega t \right), \end{aligned} \quad (119)$$

where all the parameters have been defined previously. We define a radial Lamb-Dicke parameter:

$$\eta_\rho = \sqrt{\frac{\hbar k^2}{4m\omega_1}}. \quad (120)$$

The unperturbed Hamiltonian for the radial motion is:

$$\begin{aligned} H_0'' &= H_0 + H_\rho \\ H_0'' &= -\frac{\hbar\omega_0}{2}\sigma_z + \hbar\omega_+ \left(a_+^\dagger a_+ + \frac{1}{2} \right) - \hbar\omega_- \left(a_-^\dagger a_- + \frac{1}{2} \right). \end{aligned} \quad (121)$$

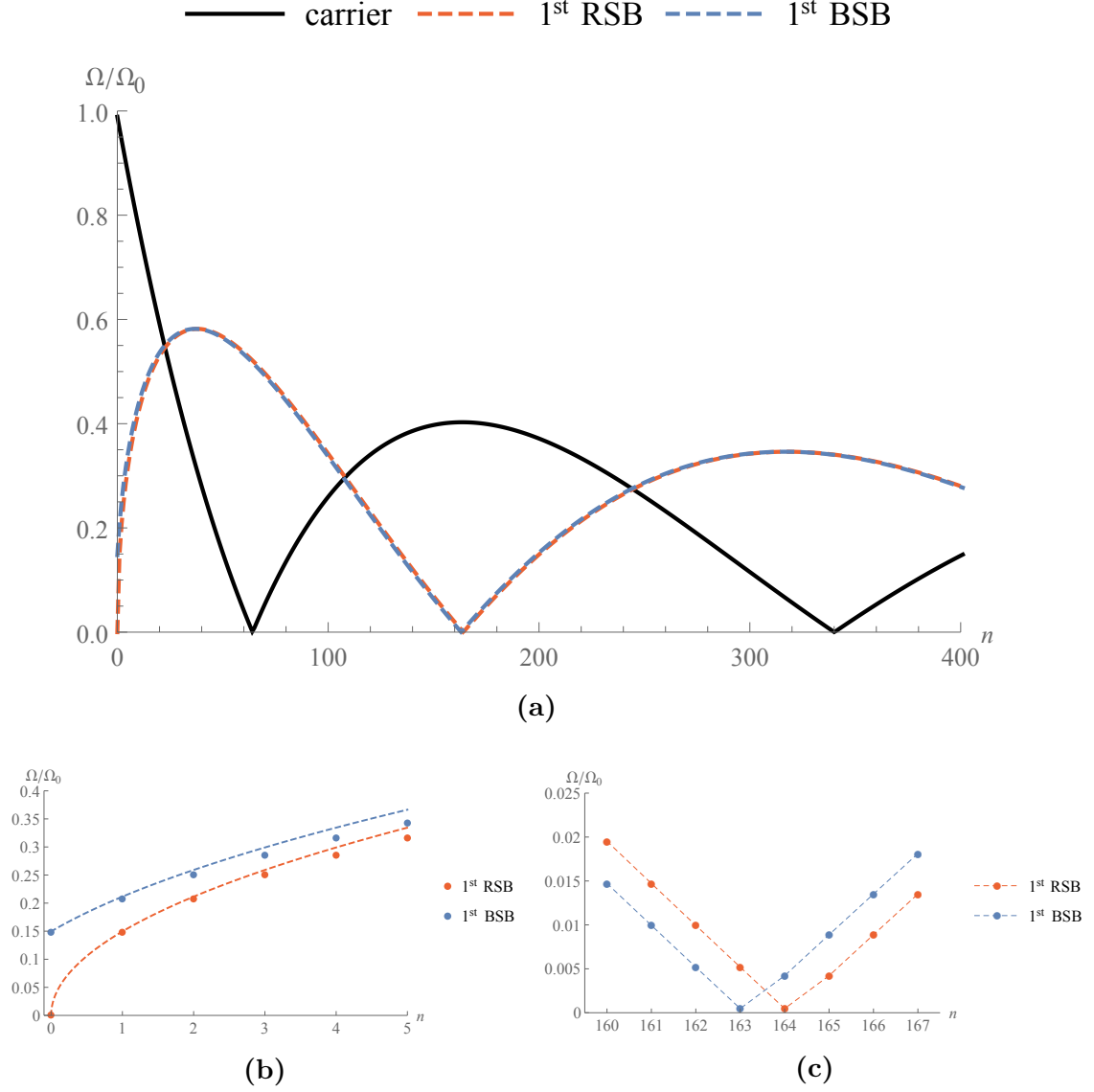


Figure III.3: (a) Normalised Rabi frequencies for the carrier, first blue sideband (BSB) and first red sideband (RSB) as a function of the motional state number for a Lamb-Dicke parameter $\eta = 0.15$. (b) At $n = 0$, the Rabi frequency for the first RSB (and all higher order RSB) is strictly zero but not for the BSB. The dashed lines are plots of equations 116, corresponding to the Lamb-Dicke approximation. (c) View of a region where the first BSB and RSB coupling strengths tend to zero. Although very small, the Rabi frequency is not exactly zero. The minimum value for the BSB is reached for a motional state number smaller by 1 (163 here) than for the RSB (164). The dashed lines just link the dots and do not correspond to a physical model.

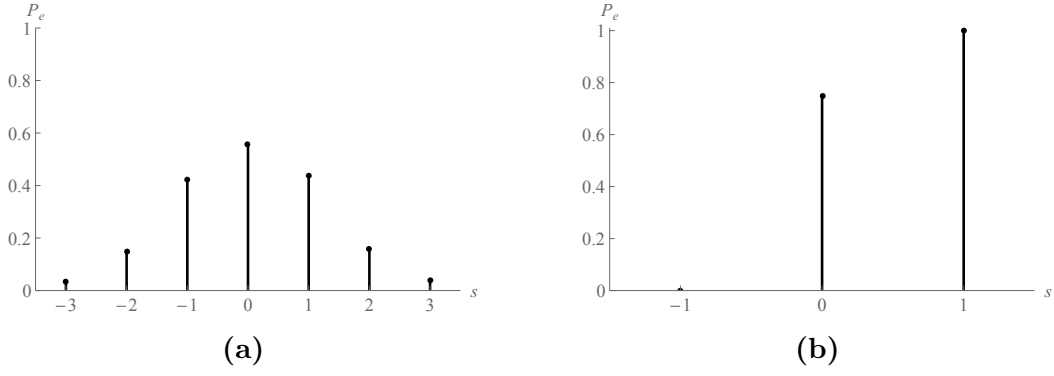


Figure III.4: (a) Probability of excitation for different sidebands when the ion is in a thermal state. Calculated for $\bar{n} = 30$ and $\eta = 0.15$ (realistic values in a Penning trap after Doppler cooling) and an interaction time $t = \pi/\Omega_0$. (b) Probability of excitation on the first RSB, carrier and first BSB for an ion in the motional ground state with probability 1. Calculated for $\eta = 0.15$ and $t = \pi/\Omega_{0,1}$. No excitation is possible on the first RSB hence the zero probability. N.B: the numbers on the abscissa for both graphs represent the sidebands: positive numbers (positive laser detuning) correspond to blue sidebands, negative numbers to red sidebands. Zero corresponds to the carrier.

There is no difficulty specific to the radial motion to find the Rabi frequency of a particular transition, we can follow the same procedure as for the axial motion and so the derivations are not detailed here to avoid repetition.

The quantity $\langle e, m_+, m_- | H_I | g, n_+, n_- \rangle$ where m_+ and n_+ are modified cyclotron states and m_- and n_- are magnetron states is given by:

$$\langle e, m_+, m_- | H_I | g, n_+, n_- \rangle = \frac{\hbar\Omega_0}{2} e^{i(s_+\omega_+ + s_-\omega_- - \delta)t} e^{-\eta_\rho^2} (i\eta_\rho)^{|s_+|+|s_-|} (-1)^{|s_-|} \times \sqrt{\frac{n_+^{<}|}{n_+^{>}|}} \sqrt{\frac{n_-^{<}|}{n_-^{>}|}} L_{n_+^{<}|}^{(|s_+|)}(\eta_\rho^2) L_{n_-^{<}|}^{(|s_-|)}(\eta_\rho^2) \quad (122)$$

with $s_\pm = m_\pm - n_\pm$, $n_\pm^{<} = \min\{n_\pm, m_\pm\}$ and $n_\pm^{>} = \max\{n_\pm, m_\pm\}$. In the resonant case where $\delta = s_+\omega_+ + s_-\omega_-$, this yields the Rabi frequency of the transition $|g, n_+, n_- \rangle \longleftrightarrow |e, n_+ + s_+, n_- + s_- \rangle$:

$$\Omega_{n_+, n_+ + s_+}^{n_-, n_- + s_-} = \Omega_0 e^{-\eta_\rho^2} \eta_\rho^{|s_+|+|s_-|} \sqrt{\frac{n_+^{<}|}{n_+^{>}|}} \sqrt{\frac{n_-^{<}|}{n_-^{>}|}} L_{n_+^{<}|}^{(|s_+|)}(\eta_\rho^2) L_{n_-^{<}|}^{(|s_-|)}(\eta_\rho^2). \quad (123)$$

In the strong-binding regime, the absorption spectrum of the ion when the laser propagates in the radial plane features sidebands corresponding to both motional modes. The double modulation produces modified cyclotron sidebands spaced by ω_+ flanked by magnetron sidebands separated by ω_- .

The coupling between two states of a radial motional mode is a two-dimensional problem as it depends on the states of both modes (see figure III.5). A consequence is that the coupling strength between say, two cyclotron states can be zero even if the corresponding Laguerre polynomial is large. Imagine the laser is tuned to

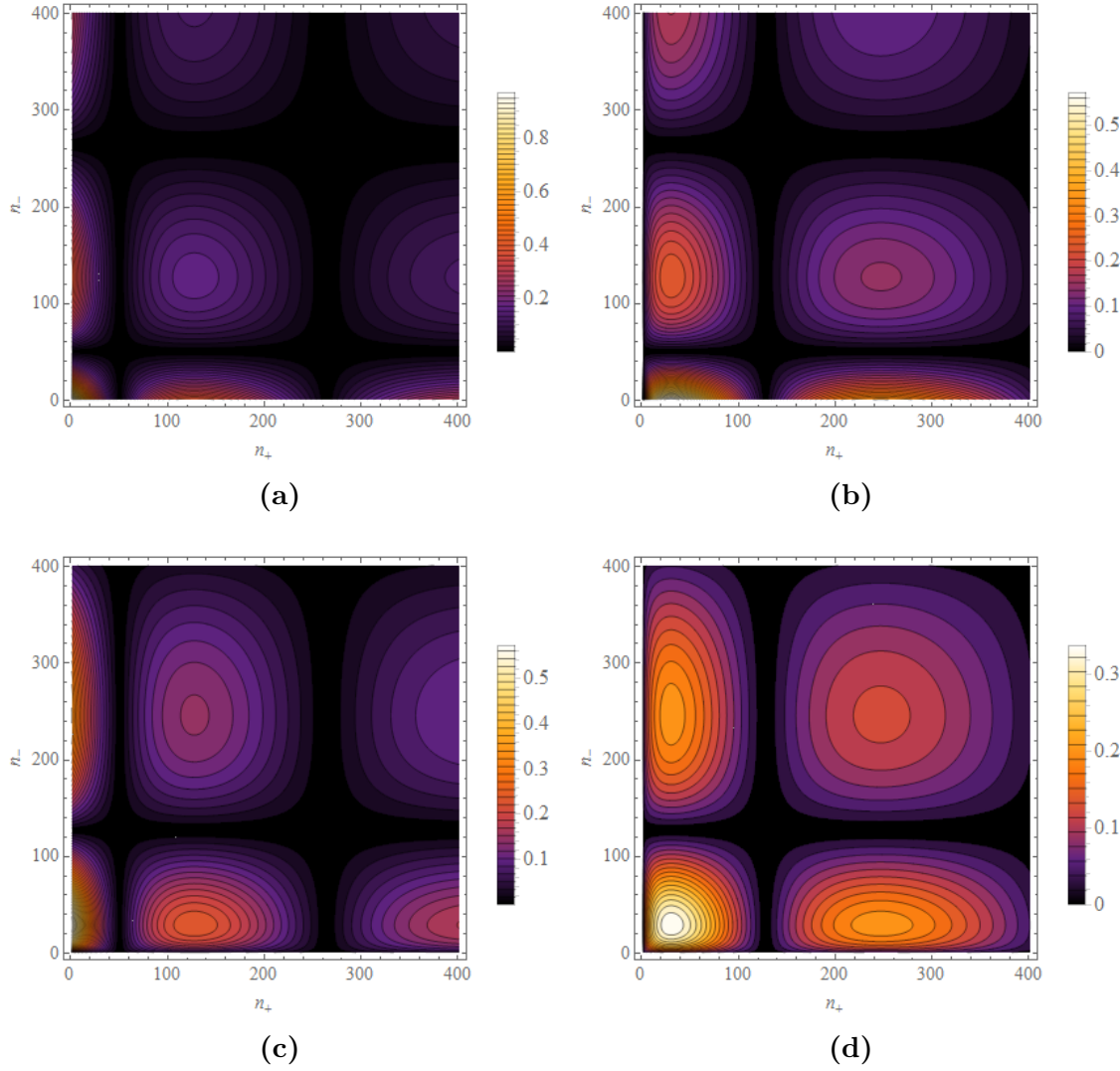


Figure III.5: Rabi frequency in units of Ω_0 for a radial Lamb-Dicke parameter $\eta_\rho = 0.17$. (a) Carrier ($s_+ = 0, s_- = 0$). (b) First cyclotron red sideband ($s_+ = -1, s_- = 0$). (c) First magnetron blue sideband ($s_+ = 0, s_- = +1$). (d) First magnetron blue sideband of first cyclotron red sideband ($s_+ = -1, s_- = +1$).

a cyclotron sideband: $\delta = s_+ \omega_+$. Here, the magnetron mode is not affected by the interaction with the laser but it contributes to determine the Rabi frequency of the transition $|g, n_+, n_- \rangle \longleftrightarrow |e, n_+ + s, n_- \rangle$. If the Laguerre polynomial $L_{n_-}^{(0)}(\eta_\rho^2)$ is vanishingly small then the Rabi frequency for that transition will tend to zero, regardless of n_+ and s_+ .

Coulomb crystals

The case of Coulomb crystals is significantly more complicated since the laser light may interact differently with each ion and each mode. Here we shall only consider the axial modes of the crystal and therefore assume that the laser propagates along the z axis. We further suppose that the crystal, which may be in linear or planar configuration, is illuminated uniformly by the laser beam. The interaction Hamiltonian for a N -ion crystal is the sum of the contributions from the individual

ions:

$$H_i = \sum_{j=1}^N \hbar \Omega_0 \cos(kz_j - \omega t) \sigma_x^j. \quad (124)$$

Recalling equation 69, we have:

$$H_i = \sum_{j=1}^N \hbar \Omega_0 \cos \left(k \sum_{\mu=1}^N b_{\mu,j}^N \sqrt{\frac{\hbar}{2m\omega_\mu}} (a_\mu + a_\mu^\dagger) - \omega t \right) \sigma_x^j. \quad (125)$$

If we define a Lamb-Dicke parameter in a similar way as for the single ion case:

$$\eta_\mu^j = b_{\mu,j}^N \sqrt{\frac{\hbar k^2}{2m\omega_\mu}}, \quad (126)$$

the dependency on the mode and the ion is apparent. This quantity is zero for certain ions in certain modes, for example for the central ion of a six-ion planar crystal for the tilt and folding modes as defined in section II.3. Particular cases where the interaction Hamiltonian can be simplified are discussed in chapter VII.

IV

Theory of Laser Cooling in a Penning Trap

Laser cooling of ions was proposed in 1975 by David Wineland and Hans Dehmelt [66] and demonstrated for the first time in 1978 [67]. This was soon followed by thorough studies of laser cooling [68, 69]. Today, laser cooling is widely used to cool ions [46], neutral atoms and even molecules [70] down to very low temperatures. In this chapter, we focus on laser cooling of ions in a Penning trap and in particular of $^{40}\text{Ca}^+$ as it is the ion species used at Imperial College. The descriptions made below are however, for a large part, valid for other ion species. This chapter only offers a theoretical treatment of laser cooling. Experimental results are discussed in chapter VII.

IV.1 The Energy Structure of Singly Ionised Calcium-40

The singly charged calcium-40 cation is a suitable medium for ion trapping experiments. Its sole valence electron confers it an alkali-like electron configuration (ground state $[\text{Ar}]4s^1$) with a fairly simple energy structure which possesses electronic transitions in the visible and near infrared domains, directly accessible with lasers. The energy levels are shown in figure IV.1. Ignoring the Zeeman sub-levels, there are five accessible levels for the valence electron: the ground state $S_{1/2}$ (orbital 4s), two short-lived excited states $P_{1/2}$ and $P_{3/2}$ (orbital 4p) and two metastable states $D_{3/2}$ and $D_{5/2}$ (orbital 3d). The dipole allowed transitions $S_{1/2} \longleftrightarrow P_{1/2}$ and $D_{3/2} \longleftrightarrow P_{1/2}$ form a closed cycle which can be used for Doppler cooling. Indeed, the selection rules state that the excited state $P_{1/2}$ can decay either to $S_{1/2}$ or $D_{3/2}$. In a Penning trap however, because of the large magnetic field, an additional transition is necessary to close the loop. This is discussed below. Owing to its narrow linewidth, much smaller than the typical motional frequencies of trapped ions, the electric quadrupole transition $S_{1/2} \longleftrightarrow D_{5/2}$ at 729 nm is suitable for resolved sideband spectroscopy and cooling. Besides, the relatively long life-time of the $D_{5/2}$ state (about 1.2 s) makes it possible to form a quantum bit with $S_{1/2}$ and $D_{5/2}$. The

Transition	Wavelength (nm)	Type	Transition Rate
$S_{1/2} \longleftrightarrow P_{1/2}$	397	Dipole (E1)	$2\pi \times 21 \text{ MHz}$
$S_{1/2} \longleftrightarrow P_{3/2}$	393	Dipole (E1)	$2\pi \times 21.5 \text{ MHz}$
$D_{3/2} \longleftrightarrow P_{1/2}$	866	Dipole (E1)	$2\pi \times 1.4 \text{ MHz}$
$D_{5/2} \longleftrightarrow P_{3/2}$	854	Dipole (E1)	$2\pi \times 1.35 \text{ MHz}$
$S_{1/2} \longleftrightarrow D_{5/2}$	729	Quadrupole (E2)	$2\pi \times 0.136 \text{ Hz}$

Table IV.1: Relevant electronic transitions for laser cooling of $^{40}\text{Ca}^+$. Data from ref. [71, 72, 73] and the Handbook of Basic Atomic Spectroscopic Data of NIST.

electronic transitions relevant for laser cooling and other experiments are shown in table IV.1.

Zeeman effect

The magnetic field used in Penning traps is the origin of a Zeeman effect splitting the energy levels of the trapped ions. This is of importance as it changes the resonant frequencies needed to drive the electronic transitions of the ions. Although significant, the effect on calcium ions is weak enough in comparison to the spin-orbit coupling to be treated with perturbation theory. The potential associated with the magnetic field is:

$$V_m = -\vec{\mu} \cdot \vec{B} \quad (127)$$

and the magnetic moment is given by:

$$\vec{\mu} = -\frac{\mu_B(g_l\vec{L} + g_s\vec{S})}{\hbar} \quad (128)$$

with μ_B the Bohr magnetron, $g_l = 1$ and $g_s \simeq 2.0023192$. Given an unperturbed spin-orbit wavefunction $|\psi^{(0)}\rangle$, the first order Zeeman energy shift is:

$$\begin{aligned} \Delta E_\psi^{(1)} &= \langle V_m \rangle \\ &= \frac{\mu_B B}{\hbar} \langle \psi^{(0)} | (g_l L_z + g_s S_z) | \psi^{(0)} \rangle \\ &= \mu_B B g_j m_j \end{aligned} \quad (129)$$

where we have introduced the Landé g -factor defined by:

$$g_j = g_l \frac{j(j+1) - s(s+1) + l(l+1)}{2j(j+1)} + g_s \frac{j(j+1) + s(s+1) - l(l+1)}{2j(j+1)} \quad (130)$$

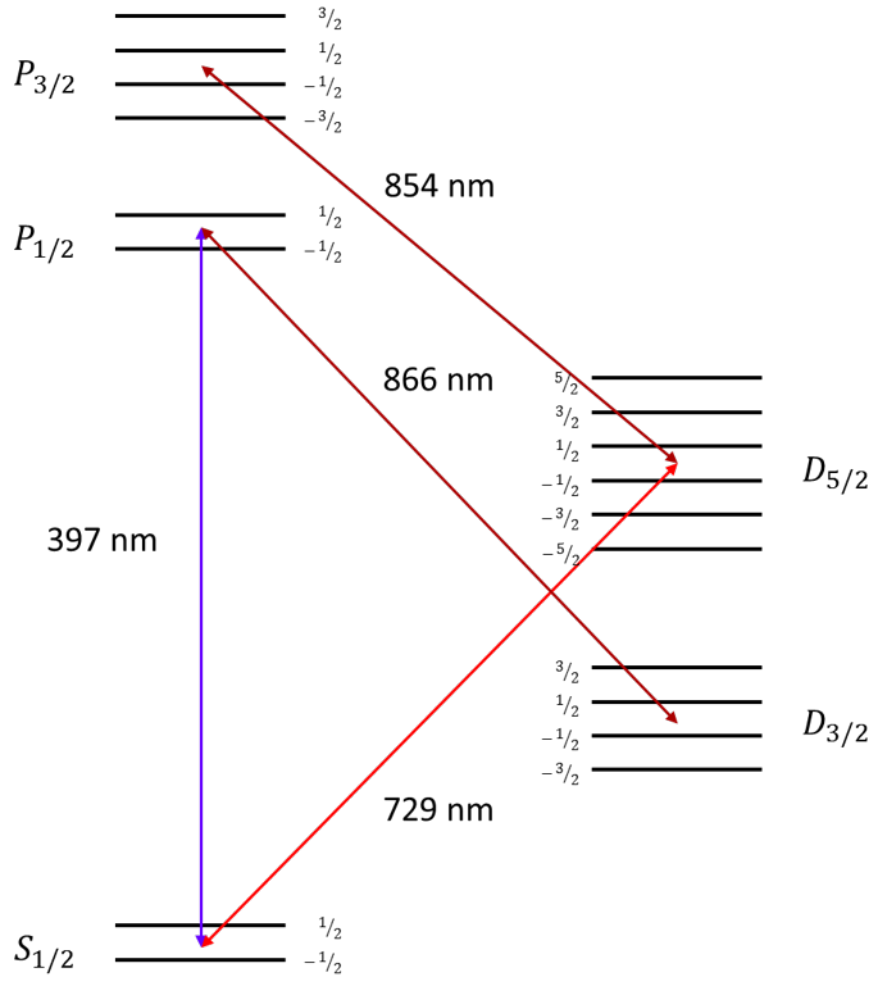


Figure IV.1: Energy levels of $^{40}\text{Ca}^+$ showing the Zeeman sub-levels (not to scale) in the presence of a magnetic field and the relevant electronic transitions for laser cooling.

and m_j the secondary total angular momentum quantum number. j , l and s are respectively the main total angular momentum, orbital and spin quantum numbers as usually denoted. The values of the Zeeman shifts for the different energy levels are given in table IV.2. With a typical magnetic field of 2 T, they are of the order of ten to hundred gigahertz. The consequence of these large splittings is that a laser can only drive an optical transition between two specific Zeeman sub-levels e.g. $S_{1/2,1/2} \longleftrightarrow P_{1/2,-1/2}$.

It is also necessary to consider the second order Zeeman effect as it produces an energy shift with a corresponding frequency comparable to the linewidth of the dipole transitions. The second order energy shift is:

$$\Delta E_{\psi}^{(2)} = \sum_{\phi \neq \psi} \frac{\mu_B^2 B^2 |\langle \phi^{(0)} | (g_l L_z + g_s S_z) | \psi^{(0)} \rangle|^2}{\hbar^2 (E_{\psi}^{(0)} - E_{\phi}^{(0)})}. \quad (131)$$

The terms of this sum can be calculated in the $|l, s, m_l, m_s\rangle$ basis with Clebsch-Gordan coefficients which can be found for example in ref. [74]. They are non zero for sub-levels with identical azimuthal quantum number l and m_j but different j . We obtain:

$$\begin{aligned} \Delta E^{(2)}(D_{3/2,\pm 1/2}) &= -\Delta E^{(2)}(D_{5/2,\pm 1/2}) = \epsilon_{1/2}^2 (E^{(0)}(D_{3/2}) - E^{(0)}(D_{5/2})) \\ \Delta E^{(2)}(D_{3/2,\pm 3/2}) &= -\Delta E^{(2)}(D_{5/2,\pm 3/2}) = \epsilon_{3/2}^2 (E^{(0)}(D_{3/2}) - E^{(0)}(D_{5/2})) \\ \Delta E^{(2)}(P_{1/2,\pm 1/2}) &= -\Delta E^{(2)}(D_{3/2,\pm 1/2}) = \epsilon_P^2 (E^{(0)}(P_{1/2}) - E^{(0)}(P_{3/2})) \end{aligned} \quad (132)$$

where we have defined mixing coefficients:

$$\begin{aligned} \epsilon_{1/2} &= \frac{\langle D_{3/2,1/2} | -\vec{\mu} \cdot \vec{B} | D_{5/2,1/2} \rangle}{E^{(0)}(D_{5/2}) - E^{(0)}(D_{3/2})} = \frac{\sqrt{6}}{5} \frac{\mu_B B}{E^{(0)}(D_{5/2}) - E^{(0)}(D_{3/2})} \\ \epsilon_{3/2} &= \frac{\langle D_{3/2,3/2} | -\vec{\mu} \cdot \vec{B} | D_{5/2,3/2} \rangle}{E^{(0)}(D_{5/2}) - E^{(0)}(D_{3/2})} = \frac{2}{5} \frac{\mu_B B}{E^{(0)}(D_{5/2}) - E^{(0)}(D_{3/2})} \\ \epsilon_P &= \frac{\langle P_{3/2,1/2} | -\vec{\mu} \cdot \vec{B} | P_{1/2,1/2} \rangle}{E^{(0)}(P_{1/2}) - E^{(0)}(P_{3/2})} = \frac{\sqrt{2}}{3} \frac{\mu_B B}{E^{(0)}(P_{1/2}) - E^{(0)}(P_{3/2})}. \end{aligned} \quad (133)$$

The numerical values are given in table IV.2 in terms of frequency shifts. Note that the sign of the shift does not depend on that of m_j which means that the total shift (first plus second order) of a pair of sub-levels $\pm|m_j|$ from the unperturbed level is asymmetric, provided the second order shift is non zero.

***j*-mixing**

The interaction that leads to the second order Zeeman shift is also responsible for an admixture of the states with different j but equal l and m_j called *j*-mixing. The magnitude of this admixture is given by the mixing coefficients in equation 133. The eigenstates of the system are not those of the unperturbed system (at $B = 0$)

Term	Level (THz)	m_j	1 st order (GHz T ⁻¹)	2 nd order (MHz T ⁻²)
S _{1/2}	0	$\pm 1/2$	± 14.01	0
D _{3/2}	409.223	$\pm 1/2$	± 5.59	-25.8
		$\pm 3/2$	± 16.8	-17.2
D _{5/2}	411.042	$\pm 1/2$	± 8.40	25.8
		$\pm 3/2$	± 25.2	17.2
		$\pm 5/2$	± 42.0	0
P _{1/2}	755.223	$\pm 1/2$	± 4.66	-6.51
P _{3/2}	761.905	$\pm 1/2$	± 9.34	6.51
		$\pm 3/2$	± 28.0	0

Table IV.2: First and second order Zeeman shifts for $^{40}\text{Ca}^+$.

described by the total angular momentum j but rather superpositions of the latter. A notable consequence of this mixing is that electronic transitions that are forbidden at zero field become allowed albeit weakly. For example, the mixing of P_{1/2} with P_{3/2} and of D_{3/2} with D_{5/2} renders the transition D_{5/2} $\longleftrightarrow$ P_{1/2} possible as both D_{3/2} $\longleftrightarrow$ P_{1/2} and D_{5/2} $\longleftrightarrow$ P_{3/2} are dipole allowed. The decay rate as a function of the magnetic field (expressed in tesla) from P_{1/2} to D_{5/2} can be calculated using perturbation theory [74, 75]:

$$\Gamma_{\text{P}_{1/2} \leftrightarrow \text{D}_{5/2}} = \Gamma_{\text{P}_{1/2} \leftrightarrow \text{S}_{1/2}} \times 4.2 \times 10^{-7} \times B^2. \quad (134)$$

Exact calculations (non perturbative) yield very similar results [76]. It is now understood that the transitions S_{1/2} $\longleftrightarrow$ P_{1/2} and D_{3/2} $\longleftrightarrow$ P_{1/2} do not form a closed cycle as the P_{1/2} state can decay to D_{5/2} as well.

IV.2 Doppler Cooling

Doppler cooling consists in shining a laser with a frequency close to resonance with an atomic electronic transition. It relies on the momentum transfer between the photons of the laser beam and the atom. Here, we call cooling the reduction of the velocity of the atom. Let us denote $\hbar\vec{K}$ the momentum of the atom and $\hbar\vec{k}$ the momentum of a photon. When the atom absorbs a photon, its momentum becomes $\hbar\vec{K} + \hbar\vec{k}$. Reducing the problem to one dimension, it is clear that the momentum of the atom is either increased by $\hbar k$ if the photons and the atom propagate in the same direction or reduced by $\hbar k$ if they are counter-propagating. To make sure absorption occurs only in the counter-propagating case, the frequency of the laser is tuned to a

frequency slightly smaller than the frequency of the atomic transition (red-detuning) so that the laser light is resonant only when the atom moves towards the source of the beam due to Doppler shift; hence the name of the technique. After absorption, the atom is in an excited electronic state and will spontaneously re-emit a photon which will impart a recoil momentum to the atom. However, because spontaneous emission is isotropic to a good approximation, the net change in momentum after a few events is nil. When the laser light is resonant with an electric dipole allowed transition, the scattering rate i.e. the number of absorption and re-emission events per unit of time may be very high (tens of MHz) resulting in a strong deceleration of the atom. The momentum kicks during spontaneous emission however prevent reaching a zero temperature with Doppler cooling.

In this section, we focus on the case of a single ion in a Penning trap and we seek to determine the minimum temperature achievable. This limit will be expressed in terms of the motional states of the quantum harmonic oscillators introduced in section II.2 which are the most relevant quantities for a trapped ion. The description of Doppler cooling in a Penning trap has been carried out in detail by Wayne Itano and David J. Wineland in their 1982 paper [69] as well as in previous theses of our group [77, 78]. The important results are given below.

The natural linewidths of electric dipole allowed transitions involved in Doppler cooling are much larger than typical motional frequencies in a Penning trap (tens of MHz versus hundreds of kHz); a situation that corresponds to the weak binding regime where the sideband structure described in section III.2 can be ignored. Following the method of ref. [69], we consider the absorption of a photon with momentum $\hbar\vec{k}$ followed by the spontaneous emission of a photon with momentum $\hbar\vec{k}_s$ such that the change in velocity of the ion is $\Delta\vec{v} = \hbar(\vec{k} - \vec{k}_s)/m$. We recall the equations of motion:

$$\begin{aligned} x(t) &= R_+ \cos(\omega_+ t + \phi_+) + R_- \cos(\omega_- t + \phi_-) \\ y(t) &= -R_+ \sin(\omega_+ t + \phi_+) - R_- \sin(\omega_- t + \phi_-) \\ z(t) &= Z \cos(\omega_z t + \phi_z) \end{aligned} \tag{135}$$

and we write x' , y' and z' the coordinates after the absorption-emission event. We assume the absorption and emission to occur instantaneously at t_0 such that:

$$\begin{aligned} j(t_0) &= j'(t_0) \\ \dot{j}(t_0) &= \dot{j}'(t_0) + \Delta v_j \quad , \quad j \in \{x, y, z\}. \end{aligned} \tag{136}$$

This is a reasonable assumption considering the absorption and emission occur on a time scale much shorter than the oscillation periods of the ion. With these equations,

we can calculate the variation of the squared amplitudes:

$$\Delta Z^2 = \left(\frac{\Delta v_z}{\omega_z} \right)^2 - \frac{2Z\Delta v_z}{\omega_z} \sin(\omega_z t_0 + \phi_z) \quad (137a)$$

$$\Delta R_+^2 = \frac{(\Delta v_x)^2 + (\Delta v_y)^2}{4\omega_1^2} - \frac{R_+}{\omega_1} (\Delta v_x \sin(\omega_+ t_0 + \phi_+) + \Delta v_y \cos(\omega_+ t_0 + \phi_+)) \quad (137b)$$

$$\Delta R_-^2 = \frac{(\Delta v_x)^2 + (\Delta v_y)^2}{4\omega_1^2} + \frac{R_-}{\omega_1} (\Delta v_x \sin(\omega_- t_0 + \phi_-) + \Delta v_y \cos(\omega_- t_0 + \phi_-)) \quad (137c)$$

The equation for the axial motion may be rewritten as:

$$\Delta Z^2 = \left(\frac{\Delta v_z}{\omega_z} \right)^2 + \frac{2\Delta v_z \dot{z}(t_0)}{\omega_z^2}. \quad (138)$$

For the amplitude to decrease, it is necessary that Δv_z and $\dot{z}(t_0)$ have opposite signs. Assuming $\dot{z}(t_0)$ is negative, the condition is $(\vec{k} - \vec{k}_s) \cdot \hat{z} > 0$. If the laser light propagates along the axial direction, then $\vec{k} \cdot \hat{z} \geq \vec{k}_s \cdot \hat{s}$ and so the condition for cooling becomes $k > 0$ i.e. the light and the ion have to propagate in opposite directions. As mentioned above, detuning the laser to a lower frequency (red detuning) ensures that absorption occurs preferentially when the light and the ion propagate in opposite directions.

Cooling the radial motion is more complicated: to reduce the amplitude of the magnetron mode, one needs to increase its energy while the energy of the modified cyclotron mode needs to be diminished. A uniform red detuned laser beam propagating in the radial plane ($\vec{k} \in (x, y)$) will cool the cyclotron mode but increase the amplitude of the magnetron motion until the ion goes out of the laser beam or crashes in the trap's electrodes [69].

The solution to this issue is called intensity gradient cooling. Both radial modes can be cooled by using a non-uniform beam (still with $\vec{k} \in (x, y)$) with a greater intensity on the side of the magnetron orbit where the ion and the light propagate in the same direction (see figure IV.2). We choose the radial cooling beam to be along the x direction and consider a Gaussian intensity profile in the y direction (see figure IV.3) such that the intensity at the centre of the trap ($y = 0$) is I_1 and the gradient at this point is positive and equal to I_1/y_0 . The ion is supposed to be sufficiently confined so that the intensity profile can be approximated to a linear model around $y = 0$ given by the equation $I(y) = I_1(1 + y/y_0)$. Meanwhile another red detuned laser beam, assumed uniform with intensity I_2 , propagates along the axial direction to cool the axial motion.

The rate of change of the motional amplitudes is found by multiplying the scattering

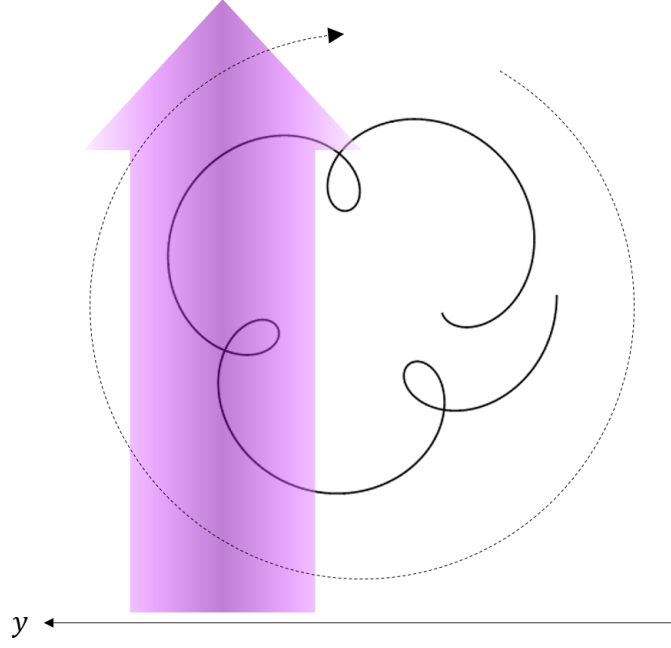


Figure IV.2: Intensity gradient cooling. The black line represents the trajectory of the ion in the radial plane and the dotted line indicates the direction of the magnetron orbit. The laser beam (purple arrow) is offset from the centre of the trap resulting in a higher intensity where the light and the magnetron motion travel in the same direction. The focusing of the laser beam is exaggerated here; in reality the ion is illuminated by the laser at any point of its trajectory, provided it is cold enough.

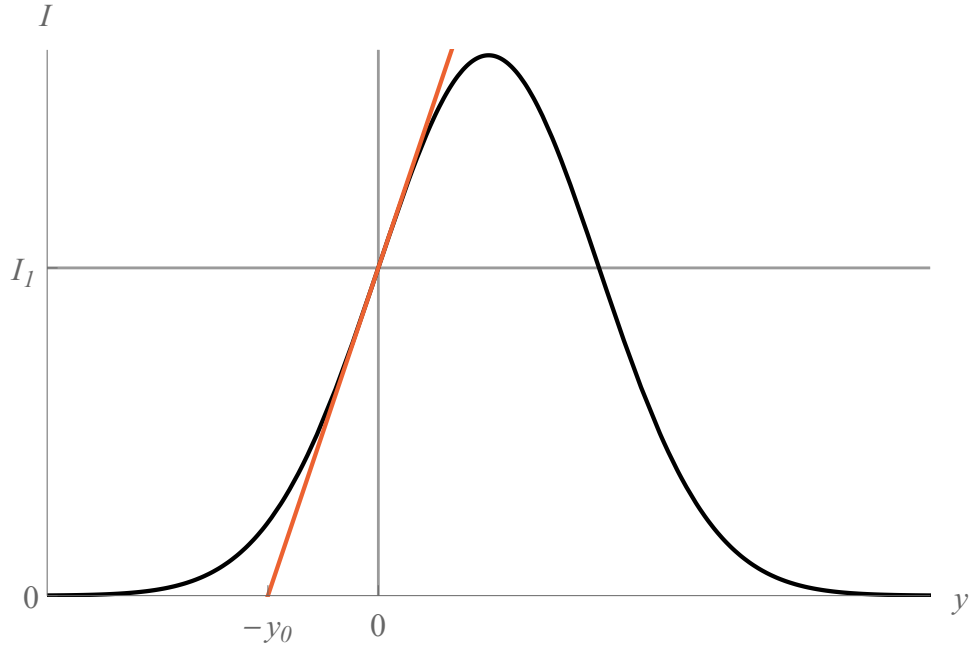


Figure IV.3: Gaussian intensity profile of the radial cooling beam and linear approximation in $y = 0$ with equation $I(y) = I_1(1 + y/y_0)$. The coordinate $y = 0$ corresponds to the centre of the trap.

rate and the change in amplitude per scattering event (equations 137):

$$\frac{dX^2}{dt} = \frac{I(\vec{r})}{\hbar\omega} \sigma(\omega, \vec{v}) \Delta X^2 \quad , \quad X \in \{Z, R_+, R_-\} \quad (139)$$

with $\sigma(\omega, \vec{v})$ the scattering cross section for a laser frequency ω . If we call γ_1 and γ_2 the average scattering rates due to the radial and axial beams respectively and Γ the spontaneous decay rate of the optical transition, the average rates are found to be:

$$\frac{d\langle Z^2 \rangle}{dt} = \gamma_2 \left(\frac{\hbar^2 k^2 (1 + (1 + \gamma_1/\gamma_2) f_z)}{m^2 \omega_z^2} + \frac{8\hbar k^2 \delta \langle Z^2 \rangle}{m(\Gamma^2 + 4\delta^2)} \right) \quad (140a)$$

$$\frac{d\langle R_+^2 \rangle}{dt} = \gamma_1 \left(\frac{\hbar^2 k^2 (1 + (1 + \gamma_2/\gamma_1)(f_x + f_y))}{4m^2 \omega_1^2} + \frac{4\hbar k^2 \delta \omega_+ \langle R_+^2 \rangle}{m\omega_1(\Gamma^2 + 4\delta^2)} + \frac{\hbar k \langle R_+^2 \rangle}{2m\omega_1 y_0} \right) \quad (140b)$$

$$\frac{d\langle R_-^2 \rangle}{dt} = \gamma_1 \left(\frac{\hbar^2 k^2 (1 + (1 + \gamma_2/\gamma_1)(f_x + f_y))}{4m^2 \omega_1^2} - \frac{4\hbar k^2 \delta \omega_- \langle R_-^2 \rangle}{m\omega_1(\Gamma^2 + 4\delta^2)} - \frac{\hbar k \langle R_-^2 \rangle}{2m\omega_1 y_0} \right) \quad (140c)$$

where

$$f_j = \int P \left(\frac{\vec{k}_s}{\|\vec{k}_s\|} \right) \frac{\vec{k}_s \cdot \hat{j}}{\|\vec{k}_s\|} d\Omega \quad , \quad j \in \{x, y, z\} \quad (141)$$

are geometrical factors determined by the probabilities of spontaneous emission in given directions of space ($d\Omega$ represents the solid angle). The first term in the equations 140 corresponds to the heating due to the spontaneous re-emission. The second term depends on the detuning and, for a negative (red) detuning ($\omega < \omega_0$), leads to cooling for the axial and cyclotron modes and heating for the magnetron mode. The third term in equations 140b and 140c arises from the radial beam offset and yields heating of the cyclotron and cooling of the magnetron mode. The condition for simultaneous cooling of both radial modes i.e. for the sum of the two last terms in equations 140b and 140c to be negative is:

$$\omega_- < \frac{c(\Gamma^2 + 4(\omega_0 - \omega)^2)}{8\omega(\omega_0 - \omega)y_0} < \omega_+ \quad (142)$$

with c the speed of light in vacuum. From this inequality, it is apparent that if the laser beam is tightly focused, the central term may become greater than the modified cyclotron frequency which leads to heating of this mode. Conversely, a loosely focused beam will heat the magnetron mode. Note that the condition is more easily met for low axial frequencies and therefore low trapping potentials where the splitting between the magnetron and the modified cyclotron frequencies are larger. The Doppler cooling limits are determined by the steady state solutions of the differential equations 140. To find the average occupation numbers of the quantum

harmonic oscillators, we use the correspondence principle:

$$\begin{aligned}\frac{1}{2}m\omega_z^2 \langle Z^2 \rangle &= \hbar\omega_z \left(\bar{n}_z + \frac{1}{2} \right) \\ m\omega_1\omega_{\pm} \langle R_{\pm}^2 \rangle &= \hbar\omega_{\pm} \left(\bar{n}_{\pm} + \frac{1}{2} \right)\end{aligned}\tag{143}$$

Choosing the laser detuning to be $\delta = -\Gamma/2$, the cooling limits are:

$$\bar{n}_z = \frac{\Gamma(1 + (1 + \gamma_1/\gamma_2)f_z)}{4\omega_z} - \frac{1}{2}\tag{144a}$$

$$\bar{n}_+ = \frac{(\omega_0 - \Gamma/2)\Gamma y_0(1 + (1 + \gamma_2/\gamma_1)(f_x + f_y))}{4(\omega_0 - \Gamma/2)\omega_+ y_0 - 2\Gamma c} - \frac{1}{2}\tag{144b}$$

$$\bar{n}_- = \frac{(\omega_0 - \Gamma/2)\Gamma y_0(1 + (1 + \gamma_2/\gamma_1)(f_x + f_y))}{2\Gamma c - 4(\omega_0 - \Gamma/2)\omega_- y_0} - \frac{1}{2}\tag{144c}$$

Neglecting the $-1/2$ term ($\Gamma \gg \omega_z$), the cooling limit of the axial motion is simply proportional to the inverse of the axial trapping frequency. Equation 144c has a singularity for a certain ω_- where $\bar{n}_-$ goes to infinity signifying that the trapping potential needs to remain under a certain value¹ (ω_- increases with potential) for the magnetron motion to be cooled. The cooling limit of the modified cyclotron mode also increases with the trapping potential but it is much lower than the magnetron's and is unlikely to reach a singularity on the stability region of the trap, unless the radial beam focus is very tight (small y_0). The total kinetic energy can be minimised by setting the cooling laser powers such that the ratio $\gamma_1/\gamma_2 = \sqrt{2(f_x + f_y)/f_z}$ which is 2 for an isotropic spontaneous emission. In such case, the cooling limit of the axial motion is

$$\bar{n}_z = \frac{\Gamma}{2\omega_z} - \frac{1}{2}\tag{145}$$

which corresponds to the well-known Doppler limit for the temperature T given by [79]:

$$k_B T = \frac{\hbar\Gamma}{2} = \hbar\omega_z \left(\bar{n}_z + \frac{1}{2} \right).\tag{146}$$

For a calcium ion, where $\Gamma = 2\pi \times 21$ MHz, the Doppler limit is approximately $T = 0.5$ mK.

Doppler cooling of $^{40}\text{Ca}^+$

The transition used for Doppler cooling of $^{40}\text{Ca}^+$ is the dipole allowed $S_{1/2} \longleftrightarrow P_{1/2}$ with a wavelength of roughly 397 nm and a $2\pi \times 21$ MHz transition rate. This

¹This value is only valid for the linear intensity profile model. Experimentally, we find that the magnetron motion can be cooled at higher trapping potentials than theoretically predicted.

transition can be addressed directly with a near resonant laser. As seen in section IV.1, the $P_{1/2}$ state decays predominantly to the ground state $S_{1/2}$ but also to the metastable $D_{3/2}$ via a dipole allowed transition and in a Penning trap, due to j -mixing, to the $D_{5/2}$ with a smaller probability. In order to achieve efficient Doppler cooling, the scattering rate must be high and so the D metastable states cannot be allowed to be populated. Any population that decays to these states must be pumped out. This can be done with “repump” lasers tuned to the resonant frequencies of the transitions $D_{3/2} \longleftrightarrow P_{1/2}$ at 866 nm and $D_{5/2} \longleftrightarrow P_{3/2}$ at 854 nm. Because the Zeeman splitting is larger than the typical linewidth of the lasers used for Doppler cooling, the different sub-levels need to be pumped by individual laser radiations.

The polarisations of the cooling beams also have to be considered. In our set-up, described in chapter V, a 397 nm laser propagates along the magnetic field axis – and the quantisation axis – to cool the axial motion and another one perpendicularly in the radial plane to cool the radial modes. The repump laser beams at 866 nm and 854 nm all propagate along the magnetic field. For technical reasons, we require the beams to be linearly polarised and the axial and radial 397 nm beams to be at the same frequency. To obtain a significant interaction with both blue beams, the selection rules state that σ transitions (m_j changed by ± 1) should be driven and that the polarisation of the radial beam should be orthogonal to the quantisation axis i.e. it should be in the radial plane. The (linear) polarisation of the axial beam does not matter. Detailing the sub-levels, the cooling transitions are thus $S_{1/2,1/2} \longleftrightarrow P_{1/2,-1/2}$ and $S_{1/2,-1/2} \longleftrightarrow P_{1/2,1/2}$; each addressed with different lasers due to the large Zeeman splitting. Similarly the repump lasers should be resonant with σ transitions as the beams are collinear with the quantisation axis. The transitions between the Zeeman sub-levels of $D_{3/2}$ and $P_{1/2}$ and $D_{5/2}$ and $P_{3/2}$ are shown in tables IV.3 and IV.4.

IV.3 Axialisation

Axialisation or azimuthal quadrupolar excitation is a technique consisting in applying a radial quadrupolar electric field oscillating at the true cyclotron frequency (ω_c) in order to couple the magnetron motion to the modified cyclotron motion. This coupling allows the former to benefit from the efficient laser cooling of the latter without the need to satisfy the condition described by equation 142. To implement this technique, the ring electrode has to be split into four electrically separated segments on which are applied oscillating voltages with a π phase shift between adjacent segments. Axialisation has previously been studied in the ion trapping group of Imperial College with experimental realisations reported in ref. [80, 81] and analytical treatments made in ref. [82] and the PhD theses of Hebe Powell [83] and Richard Hendricks [84]. An analysis of the effects of various forms of additional

$D_{3/2} m_j$	$P_{1/2} m'_j$	Type	$\Delta\nu$ (MHz) (B in T)
$-3/2$	$-1/2$	$\sigma+$	$12140 \times B + 10.7 \times B^2$
$-1/2$	$-1/2$	π	$930 \times B + 19.3 \times B^2$
$-1/2$	$1/2$	$\sigma+$	$10250 \times B + 19.3 \times B^2$
$1/2$	$-1/2$	$\sigma-$	$-10250 \times B + 19.3 \times B^2$
$1/2$	$1/2$	π	$-930 \times B + 19.3 \times B^2$
$3/2$	$1/2$	$\sigma-$	$-12140 \times B + 10.7 \times B^2$

Table IV.3: Transitions between the Zeeman sub-levels of $D_{3/2}$ and $P_{1/2}$. The last column gives the difference with the transition frequency at zero magnetic field.

$D_{5/2} m_j$	$P_{3/2} m'_j$	Type	$\Delta\nu$ (MHz) (B in T)
$-5/2$	$-3/2$	$\sigma+$	$14000 \times B$
$-3/2$	$-3/2$	π	$-2800 \times B - 17.2 \times B^2$
$-3/2$	$-1/2$	$\sigma+$	$15860 \times B - 10.7 \times B^2$
$-1/2$	$-3/2$	$\sigma-$	$-19600 \times B - 25.8 \times B^2$
$-1/2$	$-1/2$	π	$-940 \times B - 19.3 \times B^2$
$-1/2$	$1/2$	$\sigma+$	$17740 \times B - 19.3 \times B^2$
$1/2$	$-1/2$	$\sigma-$	$-17740 \times B - 19.3 \times B^2$
$1/2$	$1/2$	π	$940 \times B - 19.3 \times B^2$
$1/2$	$3/2$	$\sigma+$	$19600 \times B - 25.8 \times B^2$
$3/2$	$1/2$	$\sigma-$	$-15860 \times B - 10.7 \times B^2$
$3/2$	$3/2$	π	$2800 \times B - 17.2 \times B^2$
$5/2$	$3/2$	$\sigma-$	$-14000 \times B$

Table IV.4: Transitions between the Zeeman sub-levels of $D_{5/2}$ and $P_{3/2}$. The last column gives the difference with the transition frequency at zero magnetic field.

electric fields, including axialisation, in a Penning trap can be found in ref. [85].

In terms of energy, the effect of axialisation can be illustrated by the absorption of a quantum of the axialisation field (a photon) with the energy transferred to the radial motion. We will assume that the axialisation field is weak enough such that the energy transfer between the field and the ion is small over the period of the ion's oscillations compared to its motional energies. Given a set of non-zero motional states n_+ and n_- , the conservation of energy requires that after the absorption of a photon with frequency ω_c the radial energy is:

$$E_\rho = \hbar\omega_+(n_+ + 3/2) - \hbar\omega_-(n_- - 1/2) \quad (147)$$

or, equivalently:

$$E_\rho = \hbar\omega_+(n'_+ + 1/2) - \hbar\omega_-(n'_- + 1/2) \quad (148)$$

where $n'_+ = n_+ + 1$ and $n'_- = n_- - 1$. Similarly, after the emission of a photon at ω_c , the radial energy is:

$$E_\rho = \hbar\omega_+(n_+ - 1/2) - \hbar\omega_-(n_- + 3/2) \quad (149)$$

The evolution of the energy of the radial modes are therefore linked by the relationship:

$$\frac{dE_+}{dt} = \frac{\omega_+}{\omega_-} \frac{dE_-}{dt} \quad (150)$$

which in terms of motional amplitudes translates to:

$$R_+ \frac{dR_+}{dt} = -R_- \frac{dR_-}{dt} \quad (151)$$

The electric potential created by the radio-frequency signal applied on the segments of the ring electrode is:

$$\phi_a = V_a \frac{x^2 - y^2}{2r_0^2} \sin(\omega_c t) \quad (152)$$

where r_0 is the geometrical parameter introduced in chapter II and V_a the amplitude of the radio-frequency voltage on the electrodes. This potential exerts an electric force ($\vec{F}$) on the ion with components:

$$\begin{aligned} F_x &= -\frac{eV_a}{r_0^2} \sin(\omega_c t) x \\ F_y &= \frac{eV_a}{r_0^2} \sin(\omega_c t) y. \end{aligned} \quad (153)$$

This extra force modifies the x and y components in Newton's second law:

$$\begin{aligned}\ddot{x} &= \frac{\omega_z^2}{2}x - \frac{eV_a}{mr_0^2}\sin(\omega_c t)x + \omega_c \dot{y} \\ \ddot{y} &= \frac{\omega_z^2}{2}y + \frac{eV_a}{mr_0^2}\sin(\omega_c t)y - \omega_c \dot{x}\end{aligned}\tag{154}$$

These differential equations do not have analytical solutions but can be solve numerically. A solution is shown in figure IV.4. Instead, let us consider the instantaneous power absorption by the ion from the axialisation force given by $\mathcal{P} = \vec{F} \cdot \vec{v}$ i.e.

$$\begin{aligned}\mathcal{P} &= F_x \dot{x} + F_y \dot{y} \\ \Leftrightarrow \mathcal{P} &= \frac{eV_a}{r_0^2}\sin(\omega_c t)(y\dot{y} - x\dot{x}).\end{aligned}\tag{155}$$

Using the equations of motion (equation 16) in the unperturbed potential of the Penning trap and their derivatives yields:

$$\mathcal{P} = \frac{eV_a}{r_0^2}\sin(\omega_c t)[\omega_+ R_+^2 \sin(2\omega_+ t) + \omega_- R_-^2 \sin(2\omega_- t) + \omega_c R_+ R_- \sin(\omega_c t)]\tag{156}$$

which after reduction gives:

$$\mathcal{P} = \frac{eV_a}{2r_0^2}\omega_c R_+ R_- + \text{sinusoidal terms}.\tag{157}$$

Only the constant term will have a significant contribution over time and so we neglect the variable terms. This power is equal to the change in the ion's radial energy:

$$\mathcal{P} = \frac{dE_\rho}{dt} = \frac{dE_+}{dt} + \frac{dE_-}{dt}\tag{158}$$

or,

$$\mathcal{P} = \frac{\omega_c}{\omega_+} \frac{dE_+}{dt} = \frac{\omega_c}{\omega_-} \frac{dE_-}{dt}.\tag{159}$$

Rewriting this equation in terms of motional amplitudes yields a familiar system of differential equations:

$$\begin{aligned}\frac{dR_+}{dt} &= \frac{eV_a}{4mr_0^2\omega_1}R_- \\ \frac{dR_-}{dt} &= -\frac{eV_a}{4mr_0^2\omega_1}R_+.\end{aligned}\tag{160}$$

We recognise the equations of harmonic oscillators which accept the solutions:

$$\begin{aligned} R_+(t) &= R_+(0) \cos(\Omega_a t) + R_-(0) \sin(\Omega_a t) \\ R_-(t) &= R_-(0) \cos(\Omega_a t) - R_+(0) \sin(\Omega_a t) \end{aligned} \tag{161}$$

where we have introduced an axialisation coupling frequency (called the beat frequency in ref. [85])

$$\Omega_a = \frac{eV_a}{4mr_0^2\omega_1}. \tag{162}$$

Interestingly, this frequency not only depends on the strength of the electric force due to the axialisation field but also on the trapping frequencies via ω_1 . Ω_a increases with ω_z as the magnetron and modified cyclotron frequencies get closer to each other (see figure IV.5).

To find the radial trajectory of the ion under the influence of axialisation, the constant motional amplitudes in the equations of motion are replaced with the expressions of equations 161.

Assuming a laser beam is shone in an appropriate way to cool the modified cyclotron mode, the magnetron mode will be cooled as well thanks to this coupling. Axialisation therefore allows the somewhat stringent requirements of intensity gradient cooling to be overlooked. In practice, in our experiment, we almost always use a combination of axialisation and intensity gradient as axialisation alone is not enough to cool the magnetron motion close to the Doppler limit; it only prevents the motion from expanding too much.

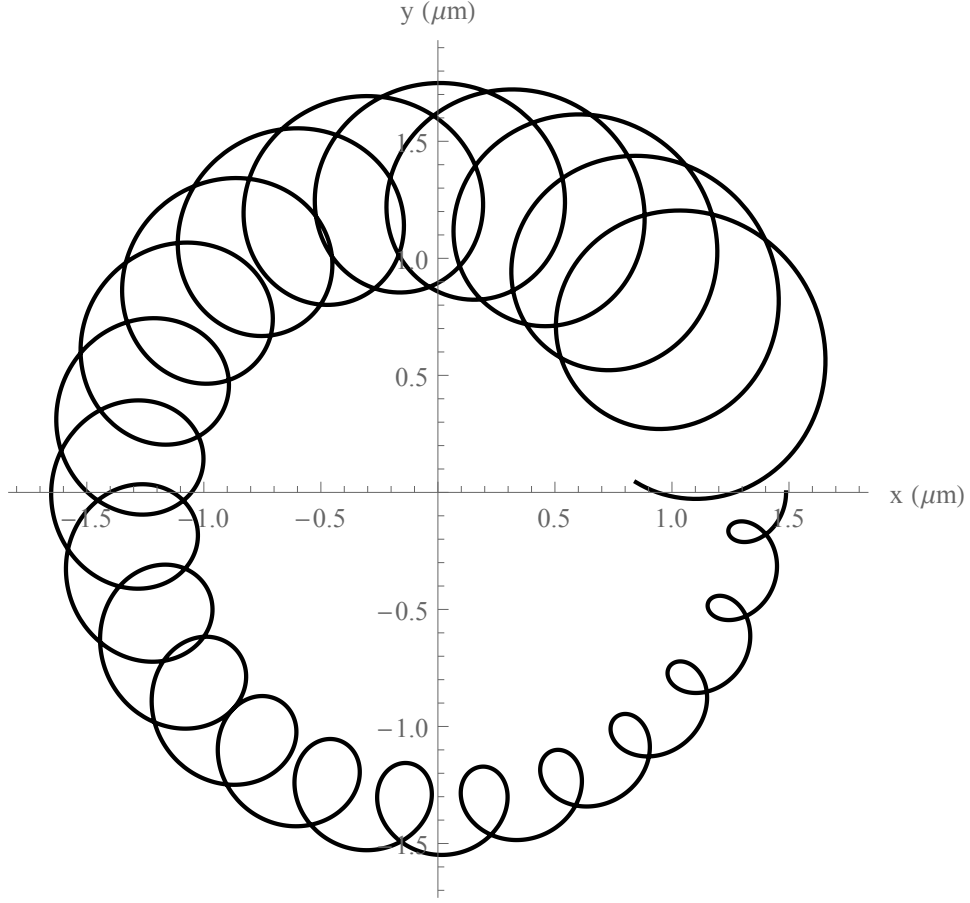


Figure IV.4: Radial trajectory of a single ion in the presence of an axialisation field. Calculated by numerical solving of equations 154 for $^{40}\text{Ca}^+$ with an axial frequency of 200 kHz, a cyclotron frequency of 729.4 kHz and an axialisation potential of 5 V. The initial conditions are taken from the equations of motion without axialisation and the cooling limits for $y_0 = 20 \mu\text{m}$.

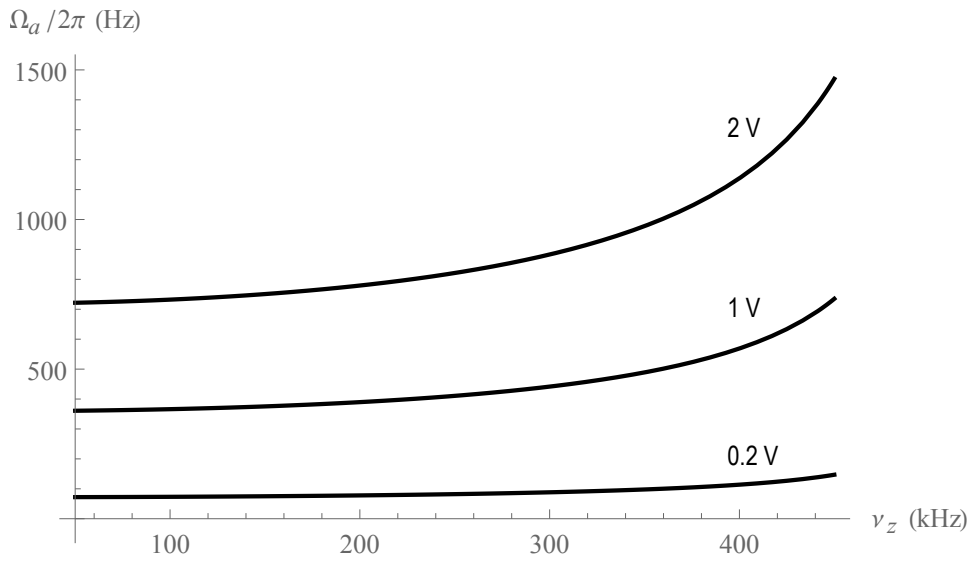


Figure IV.5: Axialisation coupling frequency as a function of the axial frequency for different values of the voltage applied on the ring electrode. Calculated for a cyclotron frequency of 729.4 kHz and a ring electrode inner radius $r_0 = 10.8 \text{ mm}$.

IV.4 Optical Resolved Sideband Cooling

Principle

Optical sideband cooling (SBC) is a technique used to overcome the limitations of Doppler cooling by exciting individual sidebands with a narrow linewidth laser and therefore necessitates being in the strong binding regime. In other words, there must be an accessible optical transition with a decay rate smaller than the trapping frequency. Dipole allowed transitions usually have a linewidth of a few tens of MHz (e.g. 21 MHz for the $S_{1/2} \longleftrightarrow P_{1/2}$ transition of $^{40}\text{Ca}^+$) which is larger than the typical trapping frequencies for both Penning and Paul traps. Although very high trapping frequencies can be attained in RF microtraps, the strong binding regime is nearly impossible to reach in Penning traps for a dipole allowed transition as it would require an extremely large magnetic field². Instead, a stimulated Raman transition between two ground states or a dipole-forbidden transition can be used. We will focus on the second option as it is the one we use at Imperial College ($S_{1/2} \longleftrightarrow D_{5/2}$ transition of $^{40}\text{Ca}^+$ at 729 nm). Sideband cooling was first implemented in a radio-frequency trap in 1989 at NIST [86] on a single mercury ion via direct optical excitation and in 1995, ground state cooling in three dimensions of a single beryllium ion was achieved with Raman sideband cooling [47]. Ground state cooling of the axial motion of a single calcium ion in a Penning trap was demonstrated in the Ion Trapping Group of Imperial College in 2014 [46].

The basic principle of sideband cooling is rather straightforward. Let us consider a harmonic motion like the axial motion in the Penning trap. A laser tuned to the frequency of the first red motional sideband i.e. detuned by $-\omega_z$ from the (carrier) frequency of the atomic transition is shone on the ion. Because of the detuning of the laser, when the ion absorbs a photon, a quantum of motion (a phonon) is lost in order to conserve energy. Meanwhile, spontaneous emission can occur on different sidebands with probabilities depending on the Lamb-Dicke parameter but on average, the vibrational state is conserved and so a scattering event decreases the motional number by one. Repeating the operation several times leads the ion towards the ground state. A representation of the process is shown in figure IV.6.

Sideband cooling with a three-level ion

This simple technique using only one laser to excite a dipole-forbidden transition between the ground state ($|g\rangle$) and a metastable excited state ($|e\rangle$) is highly inefficient because the low decay rate from the metastable state yields a low scattering rate and therefore an impractically slow cooling process. Heating of the ion's motion due to background electromagnetic noise would be likely to outpace sideband cooling.

²It would be about 12 T for $^{40}\text{Ca}^+$.

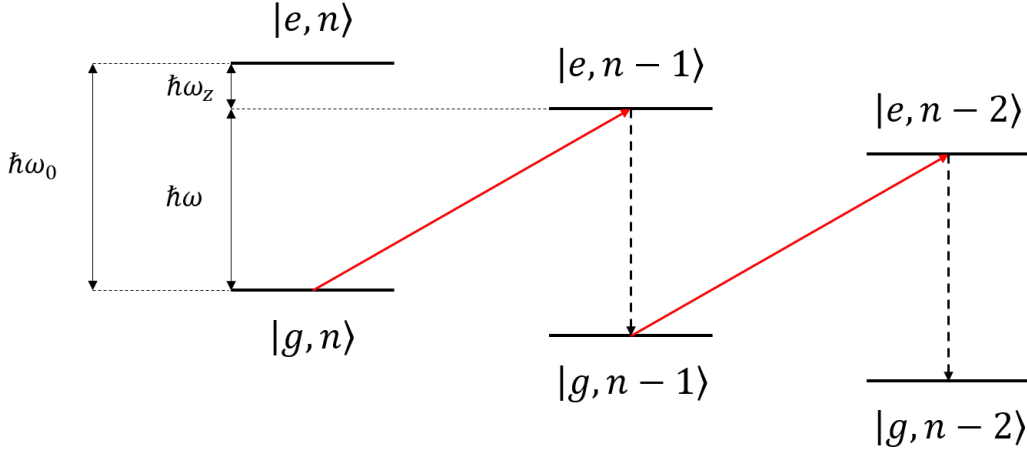


Figure IV.6: Principle of sideband cooling: the ion absorbs a photon from a laser beam tuned to the first red sideband (frequency $\omega = \omega_0 - \omega_z$). The absorption process takes the ion to the excited electronic state but decreases the motional state by one (transition $|g, n\rangle \rightarrow |e, n-1\rangle$, red solid arrow). Spontaneous emission then puts the ion back into the electronic ground state but leaves the vibrational state unchanged on average (transition $|e, n-1\rangle \rightarrow |g, n-1\rangle$, black dashed arrow).

In order to increase the cooling rate, an additional “quench” laser resonantly couples the metastable state with a third auxiliary state ($|a\rangle$) by driving the transition between the (metastable) excited state and the auxiliary state. The transition is assumed to be dipole allowed. This auxiliary state is also assumed to have a fast decay rate to the ground state. In this section, we are interested in the cascade configuration where the auxiliary level is at a higher energy than the excited state such that spontaneous emission can occur from $|a\rangle$ to $|e\rangle$ (see figure IV.7). If the rate at which the state $|a\rangle$ is populated by the laser driven transitions is small in comparison to the decay rate to the ground state, then the three-level system can be reduced to an effective two-level system. This condition is quantified by the saturation parameter of the $|a\rangle \leftrightarrow |g\rangle$ being much smaller than one [87]:

$$\frac{\Omega_{ae}^2}{(\Gamma_{ag} + \Gamma_{ae})^2 + 4\delta_{ae}^2} \ll 1 \quad (163)$$

where Ω_{ij} denotes the Rabi frequency of the transition $|i\rangle \leftrightarrow |j\rangle$ ($i, j \in \{a, e, g\}$), δ_{ij} the detuning of the laser from the transition and Γ_{ij} the associated decay rate. Assuming this condition satisfied, the effective decay rate from $|e\rangle$ to $|g\rangle$ is:

$$\tilde{\Gamma} = \frac{\Omega_{ae}^2}{(\Gamma_{ag} + \Gamma_{ae})^2 + 4\delta_{ae}^2} \Gamma_{ag}. \quad (164)$$

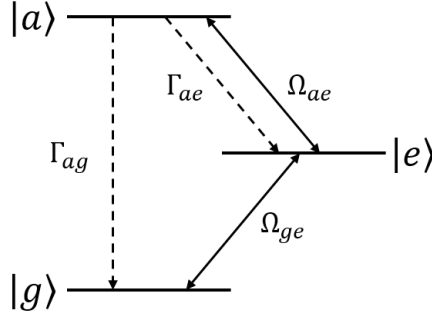


Figure IV.7: Three-level system in cascade configuration. The transition driven by lasers are indicated by the solid double-ended arrows. The dashed arrows represent the (fast) spontaneous emissions on the dipole-allowed transitions. The $|e\rangle$ state is metastable and has a slow decay rate to the ground state which we neglect.

This shows that the effective decay rate can be adjusted by changing the intensity and detuning of the quench laser for efficient cooling. A consequence of the additional laser is an AC Stark shift which modifies the frequency at which the laser exciting the quadrupole transition must be tuned. The carrier frequency moves by:

$$\delta_{AC} = -\frac{\Omega_{ae}^2}{(\Gamma_{ag} + \Gamma_{ae})^2 + 4\delta_{ae}^2}\delta_{12} \quad (165)$$

which means that in order to excite the first red sideband (of the axial motion) to carry out sideband cooling, the detuning of the laser with respect to the unshifted carrier must be $-\omega_z + \delta_{AC}$.

The narrow laser driving the transition $|g\rangle \leftrightarrow |e\rangle$ also induces a shift which depends on the coupling strength of the carrier transition and therefore the vibrational state of the ion. The amplitude of the shift is (for the axial motion) [88]:

$$\delta_n = \frac{\Omega_{n,n}}{2\omega_z}. \quad (166)$$

This is only significant when the trapping frequency is low.

Cooling limit in the Lamb-Dicke regime for $^{40}\text{Ca}^+$

We now seek to determine the limit of sideband cooling, that is, the maximum probability of occupation of the ground state achievable. We do so for the axial mode of a single ion in our particular system. For the purpose of sideband cooling, the states $S_{1/2}$, $D_{5/2}$ and $P_{3/2}$ of $^{40}\text{Ca}^+$ can be seen respectively as the ground, excited and auxiliary states of a cascade three-level system. A narrow linewidth laser drives

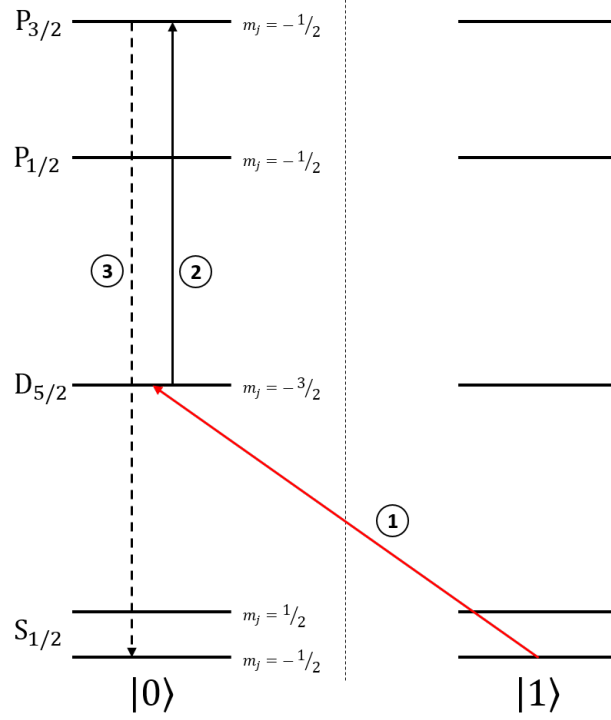
the transition between the sub-levels $S_{1/2,-1/2}$ and $D_{5/2,-3/2}$. This $\sigma-$ transition is chosen because it can be driven by a laser travelling along or perpendicular to the magnetic field thus allowing us to address both the axial and radial modes of the ion. A sideband cooling cycle therefore has to start and finish with the ion being in the sub-level $m_j = -1/2$ of the ground state. As shown in table IV.4 of section IV.2, the 854 nm laser pumps the population in $D_{5/2,-3/2}$ to $P_{3/2,-1/2}$ via a $\sigma+$ transition. The latter state can decay to either sub-levels of the ground state, to $m_j = -1/2$ with probability $2/3$ and to $m_j = 1/2$ with probability $1/3$ [46, 77]. Note that we neglect here a possible decay to $D_{5/2}$ or $D_{3/2}$. In the first case (decay to $m_j = -1/2$), the sideband cooling cycle is complete and the ion can be excited again by the 729 nm laser. If however, the ion is in the sub-level $m_j = 1/2$, it must be optically pumped to $m_j = -1/2$. This is done with one of the 397 nm Doppler cooling lasers which drives the transition $S_{1/2,1/2} \longleftrightarrow P_{1/2,-1/2}$. The ion then decays to $S_{1/2,-1/2}$ with a probability of $1/3$ (again neglecting decay to the D states) so that it takes on average three scattering events on the 397 nm transition to pump the ion to $S_{1/2,-1/2}$ and complete the sideband cooling cycle.

The difficulty of finding the cooling limit resides in the fact that each optical transition in this cooling cycle may change the phonon state of the ion. Even if the 729 nm laser is tuned to a particular sideband, off-resonant excitation on other sidebands cannot be ignored. On dipole transitions too, absorption and decay do not always occur on the carrier transition. Let us consider some phonon state $|n\rangle$ with a probability of occupation $P(n)$. To find the evolution of this probability during sideband cooling, we need to add the contributions of the events that populate $|n\rangle$ and subtract those that deplete it. We get:

$$\begin{aligned} \frac{dP(n)}{dt} = & - \sum_{k=0, k \neq n}^{\infty} \sum_{j=0}^{\infty} \frac{\tilde{\Gamma} \Omega_{n,j}}{\tilde{\Gamma}^2 + 4(\delta + (n-j)\omega_z)^2 + 2\Omega_{n,j}^2} \left(\frac{2}{3} D_{j \rightarrow k} + \frac{1}{3} \tilde{D}_{j \rightarrow k} \right) P(n) \\ & + \sum_{k=0, k \neq n}^{\infty} \sum_{j=0}^{\infty} \frac{\tilde{\Gamma} \Omega_{k,j}}{\tilde{\Gamma}^2 + 4(\delta + (k-j)\omega_z)^2 + 2\Omega_{k,j}^2} \left(\frac{2}{3} D_{j \rightarrow n} + \frac{1}{3} \tilde{D}_{j \rightarrow n} \right) P(k) \end{aligned} \quad (167)$$

where $\Omega_{n,j}$ is the Rabi frequency as defined by equation 113, δ is the laser detuning from the carrier. $D_{j \rightarrow k}$ and $\tilde{D}_{j \rightarrow k}$ denote diffusion coefficients which represent a change in the phonon state from $|j\rangle$ to $|k\rangle$ when the ion decays to the ground state. The former when the ion decays directly to $S_{1/2,-1/2}$ from $P_{3/2,-1/2}$ and the latter when it cycles on the $S_{1/2,1/2} \longleftrightarrow P_{1/2,-1/2}$ transition.

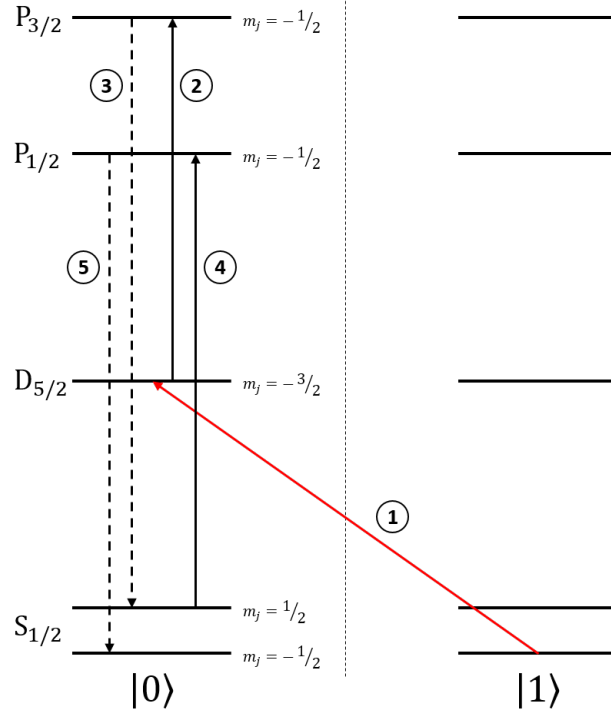
To find an analytical expression of the cooling limit, we need to make several assumptions and approximations. We place the ion in the Lamb-Dicke regime such that the phonon number can only change by 0 or ± 1 during an optical transition. We further assume that the ion is close to the ground state and its average phonon number $\bar{n}$ is much lower than 1. Only the phonon states $|0\rangle$ and $|1\rangle$ may be populated. We



(a)

Figure IV.8

thus shall consider two types of terms in the differential equation: the cooling terms where the ion is initially in $|1\rangle$ and finishes in $|0\rangle$ at the end of the cycle and the heating ones where it starts in $|0\rangle$ and ends in $|1\rangle$. The events where the phonon state is unchanged can be ignored. A representation of two possible cooling events is shown in figure IV.8 showing the relevant energy levels in our approximation.



(b)

Figure IV.8: Two possible sideband cooling cycles of $^{40}\text{Ca}^+$ in the Lamb-Dicke regime. Here, a phonon from the 729 nm laser is absorbed on the red sideband changing the phonon state from $|1\rangle$ to $|0\rangle$. In these examples, the other transitions do not change the phonon state. Solid arrows represent absorption and dashed arrows spontaneous emission. The numbers next to the arrows indicate the chronological order of the transitions. In (a), the ion decays directly from $P_{3/2,-1/2}$ to $S_{1/2,-1/2}$ finishing the cooling cycle while in (b) an extra cycle of absorption and spontaneous emission on the $S_{1/2} \longleftrightarrow P_{1/2}$ transition is needed to take the ion to the $m_j = -1/2$ sub-level of the ground state.

Four transitions are possible from the absorption of a photon of the 729 nm laser which is tuned to the red sideband. Only $|g, 1\rangle \rightarrow |e, 0\rangle$ is resonant. Their scattering rates are:

$$\begin{aligned}
 & \frac{\tilde{\Gamma}\Omega_0^2\eta_{729}^2}{\tilde{\Gamma}^2 + 2\Omega_0^2\eta_{729}^2} \quad \text{for } |g, 1\rangle \rightarrow |e, 0\rangle, \\
 & \frac{\tilde{\Gamma}\Omega_0^2(1 - 3\eta_{729}^2/2)^2}{4\omega_z^2 + \tilde{\Gamma}^2 + 2\Omega_0^2(1 - 3\eta_{729}^2/2)^2} \quad \text{for } |g, 1\rangle \rightarrow |e, 1\rangle, \\
 & \frac{\tilde{\Gamma}\Omega_0^2(1 - \eta_{729}^2/2)^2}{4\omega_z^2 + \tilde{\Gamma}^2 + 2\Omega_0^2(1 - \eta_{729}^2/2)^2} \quad \text{for } |g, 0\rangle \rightarrow |e, 0\rangle, \\
 & \frac{\tilde{\Gamma}\Omega_0^2\eta_{729}^2}{16\omega_z^2 + \tilde{\Gamma}^2 + 2\Omega_0^2\eta_{729}^2} \quad \text{for } |g, 0\rangle \rightarrow |e, 1\rangle.
 \end{aligned} \tag{168}$$

Being in the Lamb-Dicke regime, $\eta_{729}^2 \ll 1$. The subscript ‘729’ is here to specify that

it is the Lamb-Dicke parameter for the transition $S_{1/2} \longleftrightarrow D_{5/2}$ at 729 nm. Besides, typically $\Omega_0 \leq \tilde{\Gamma}$ (tens of kHz) so $\tilde{\Gamma}^2 \gg \eta^2 \Omega_0^2$. The axial frequency ω_z is usually several hundred kHz so $\omega_z^2 \gg \tilde{\Gamma}^2$. Note that in a Penning trap, the axial frequency can be quite low, of the same order as $\tilde{\Gamma}$ in which case this latter inequality would not be true any more. This however corresponds to a regime where the Lamb-Dicke approximation does not hold and so where the treatment made here is not valid anyway. With these inequalities, we can approximate the scattering rates to:

$$\begin{aligned}
& \frac{\Omega_0^2 \eta_{729}^2}{\tilde{\Gamma}} \quad \text{for} \quad |g, 1\rangle \rightarrow |e, 0\rangle, \\
& \frac{\tilde{\Gamma} \Omega_0^2}{4\omega_z^2} \quad \text{for} \quad |g, 1\rangle \rightarrow |e, 1\rangle, \\
& \frac{\tilde{\Gamma} \Omega_0^2}{4\omega_z^2} \quad \text{for} \quad |g, 0\rangle \rightarrow |e, 0\rangle, \\
& \frac{\tilde{\Gamma} \Omega_0^2 \eta_{729}^2}{16\omega_z^2} \quad \text{for} \quad |g, 0\rangle \rightarrow |e, 1\rangle.
\end{aligned} \tag{169}$$

The diffusion coefficient is calculated by taking into account the relative coupling strength on the different sidebands. It is 1 for the carrier i.e. when the phonon state is not changed and equal to the square of the Lamb-Dicke parameter for the blue and red sidebands. This Lamb-Dicke parameter is a priori different for each transition. We neglect terms in $\mathcal{O}(\eta^4)$ which correspond to cooling cycles where the phonon state changes more than once. This leaves us with the differential equation:

$$\begin{aligned}
\frac{dP(1)}{dt} = & - \left[\frac{\Omega^2 \eta_{729}^2}{\tilde{\Gamma}} + \frac{\tilde{\Gamma} \Omega_0^2}{4\omega_z^2} \left(\frac{2}{3} (\eta_{854}^2 + \tilde{\eta}^2) + \frac{1}{3} (\eta_{854}^2 + \tilde{\eta}^2 + 3\eta_r^2 + 3\tilde{\eta}_r^2) \right) \right] P(1) \\
& + \left[\frac{\tilde{\Gamma} \Omega^2 \eta_{729}^2}{16\omega_z^2} + \frac{\tilde{\Gamma} \Omega_0^2}{4\omega_z^2} \left(\frac{2}{3} (\eta_{854}^2 + \tilde{\eta}^2) + \frac{1}{3} (\eta_{854}^2 + \tilde{\eta}^2 + 3\eta_r^2 + 3\tilde{\eta}_r^2) \right) \right] P(0)
\end{aligned} \tag{170}$$

where $\tilde{\eta}$ is the Lamb-Dicke parameter for the decay from $P_{3/2}$ to $S_{1/2}$ while η_r and $\tilde{\eta}_r$ are the parameters respectively for the absorption and emission on the $S_{1/2} \longleftrightarrow P_{1/2}$ transition. There is a factor of 3 in front of these last two to take into account that it takes on average three cycles to pump the ion to $S_{1/2,-1/2}$. Given that $P(0) = 1 - P(1)$, the steady state solution is (see also the supplementary material of ref. [46]):

$$P(1)_{lim} = \frac{\tilde{\Gamma}^2}{4\omega_z^2} \left(\frac{\eta_{854}^2 + \tilde{\eta}^2 + \eta_r^2 + \tilde{\eta}_r^2}{\eta_{729}^2} + \frac{1}{4} \right). \tag{171}$$

Because only $|0\rangle$ and $|1\rangle$ can be populated, the average phonon number is simply equal to $P(1)$ so this equation gives the sideband cooling limit. The cooling limit decreases with increasing trapping frequencies which can be understood intuitively

by the fact that a higher frequency corresponds to a lower Lamb-Dicke parameter and thus a lower chance of non-resonant excitation of $S_{1/2} \longleftrightarrow D_{5/2}$ on the carrier or blue sideband. Outside the Lamb-Dicke regime, the cooling limit can be calculated numerically and is higher than predicted by equation 171. To obtain numerical values for the Lamb-Dicke parameters, one has to take into account the emission patterns of the transitions and, for the absorption at 397 nm, the probability to absorb an axial or a radial photon. The approximations made in this section are quite rough so the expression of the cooling limit above should not be used for quantitative analyses but it gives the general trend.

Sideband cooling outside the Lamb-Dicke regime

As mentioned above, the motional frequencies in a Penning trap can be low so that the ion is not in the Lamb-Dicke regime after Doppler cooling. Sideband cooling remains however possible but has to face an additional difficulty which we discuss here and how to overcome it. After Doppler cooling, an ion is in a thermal state with a probability to occupy a phonon state $|n\rangle$ given by equation 118. Sideband cooling requires a significant coupling between the electronic ground and excited states. We know from section III.2 that this coupling depends on the initial phonon state and on the change in phonon state i.e. the sideband and that the coupling strength (the Rabi frequency) can take values very close to zero. Suppose an ion after Doppler cooling is left at a phonon state $|n\rangle$ and suppose the Rabi frequency of the red sideband is very close to zero for some phonon state $|n_0\rangle$ with $0 < n_0 < n$. Driving the red sideband will progressively decrease the phonon state of the ion until it reaches $|n_0\rangle$ at which point the ion cannot absorb a photon from the red detuned laser because the Rabi frequency is zero. The cooling process therefore stops at this point and the ion's final phonon state is $|n_0\rangle$. Experimentally this signifies that the ion cannot be prepared in the motional ground state with a high probability. If a fraction x of the initial thermal distribution is above the first zero-coupling point of the red sideband, the occupation probability of the ground state after SBC will be at most $1 - x$. We call this situation 'population trapping', referring to the fraction x stuck at $|n_0\rangle$.

To circumvent this issue we make use of higher order sidebands. Because the ion is outside the Lamb-Dicke regime, it is possible to drive the second red sideband (or even higher depending on the case) with an appreciable Rabi frequency. The second red sideband also has points where the coupling is vanishingly small but fortunately these points do not match those of the first red sideband (see figure IV.9) so that for any phonon state, it is possible to drive either the first or the second sideband. Population trapping can thus be avoided by tuning the laser alternately to the first and second red sideband. To achieve (near) ground state cooling, the sideband cooling process should end with the laser tuned to the first red sideband since the

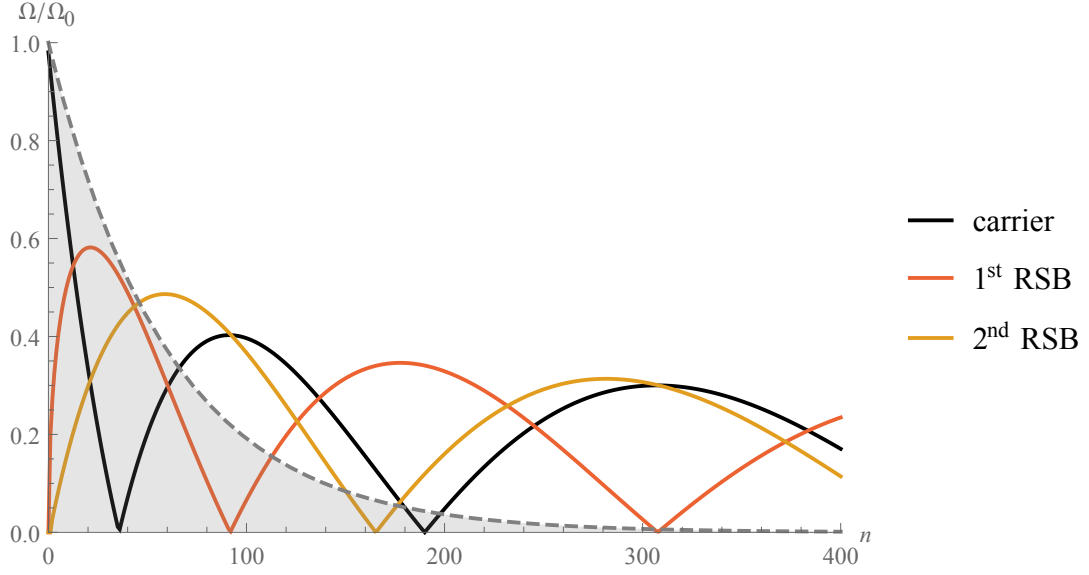


Figure IV.9: Normalised Rabi frequencies for the carrier, first and second red sidebands as a function of the motional state number for a Lamb-Dicke parameter $\eta = 0.2$. The dashed grey line represents the probability of occupation of a given motional state for a thermal distribution (normalised to 1 for $n = 0$) with an average motional state $\bar{n} = 60$.

second one has zero coupling at the phonon state $|1\rangle$. We refer to the technique where several sidebands are addressed as multiple-stage sideband cooling (MSSBC).

V

Apparatus and Experimental Techniques

Most of the current experimental set-up was developed before my time in the group and it has been described in detail in previous theses [77, 78, 89, 90]. The emphasis of this chapter will be on modifications and improvements that were made during my time in the laboratory. An overview of the whole set-up is given however for the sake of completeness. The experiment is based on three main components: the trap with the superconducting magnet and associated equipment, the laser system for cooling, spectroscopy and coherent manipulation and a control system centred around a field programmable gate array (FPGA) and a computer.

V.1 The Penning Trap at Imperial College

Our trap is a cylindrical Penning trap with open endcaps known as SPECTRAP. The original design of SPECTRAP was made by Manuel Vogel for the eponymous experiment at GSI Helmholtzzentrum für Schwerionenforschung in Darmstadt, Germany. It is described in detail in Shailen Bharadia's PhD thesis [89]. The trap consists of a stack of seven cylindrical electrodes with an internal diameter of 21.6 mm made of oxygen-free high conductivity copper. The trapping is ensured by a ring electrode at the centre of the structure and two end caps. The latter are separated from the former by two compensation electrodes whose role is to compensate for the anharmonicities of the electric potential arising from the cylindrical design of the trap. The outermost electrodes are required to capture externally generated ions as done in the GSI experiment. Since the ions are internally produced in our experiment, the capture electrodes are not in use and therefore held to electrical ground. The ring electrode is split into four segments to allow an axialisation field to be applied. Each segment has a 4 mm aperture to ensure radial optical access. Figure V.1 shows a schematic cross section of SPECTRAP.

The trap is maintained at ultra high vacuum (10^{-9} to 10^{-11} mbar) in a vacuum chamber connected to an ion pump and with windows at the top and bottom as

well as on the side, aligned with the apertures of the trap. The chamber holds the trap and rests inside the bore of a solenoid superconducting magnet, mounted on a breadboard below the magnet. The magnet, maintained at a temperature of 4.2 K by liquid helium, generates a magnetic field of 1.898 T within the trap¹. Calcium ions are created by heating with an electric current a tantalum tube filled with solid calcium, called the oven, located at the bottom of the trap. The oven liberates a vapour of calcium atoms which are non-resonantly ionised by shooting a high power frequency doubled Nd:YAG pulsed laser² (wavelength of 532 nm) into the trap. The laser is left on for a duration between a few seconds to a couple of minutes. The longer it is on, the larger the ion cloud loaded will be. The ion cloud is immediately Doppler cooled after loading provided the cooling lasers are set correctly and a fluorescence signal can be observed.

The electrodes of the trap are connected to a home made device which provides the electric fields necessary for trapping, compensation and axialisation. This device was developed by Joe Goodwin; details can be found in his PhD thesis [78]. The DC originates from a low noise power supply manufactured by Delta Elektronika (E0300-0.1) which is used for the trapping field applied on the end caps as well as for small offset voltages (up to a few volts) on the compensation electrodes and the segments of the split ring electrode. The power supply delivers up to 300 V for the trapping voltage which remains in the stability region of the trap. The radio-frequency signal for axialisation comes from a Hewlett-Packard function generator and is fed to two non-inverting and two inverting channels before being sent to the ring. The device also features a pulse-down circuit which grounds the end caps when a logic low is sent. An Arduino micro-controller (Arduino Uno) is used to send short TTL³ pulses (5 to 50 μ s) to the pulse down circuit in order to switch off the trapping momentarily which has the effect of removing some ions from the trap. Indeed, the Coulomb repulsion between the ions make them move outwards quickly in the absence of a trapping potential causing some of them to leave the trap. This technique allows a capable experimenter to “pulse down” an ion cloud to a single or a few ions depending on the experiment to conduct.

V.2 Doppler Cooling Lasers

Four lasers are used for Doppler cooling, all of them are external-cavity diode lasers (ECDL). Two TuiOptics⁴ DL100 - called blue 1 and blue 2 - provide radiations at 397 nm to address the $S_{1/2} \longleftrightarrow P_{1/2}$ transition (one laser per Zeeman sub-level) with an output power of about 15 mW. For Doppler cooling their frequency is tuned

¹As calculated from the measured cyclotron frequency of $^{40}\text{Ca}^+$.

²Continuum Surelite II. The energy of the pulses is 300 mJ.

³Transistor-transistor logic. Here and in the rest of this document, TTL is used to indicate TTL compatible levels rather than TTL digital circuits.

⁴Now known as Toptica Photonics AG.

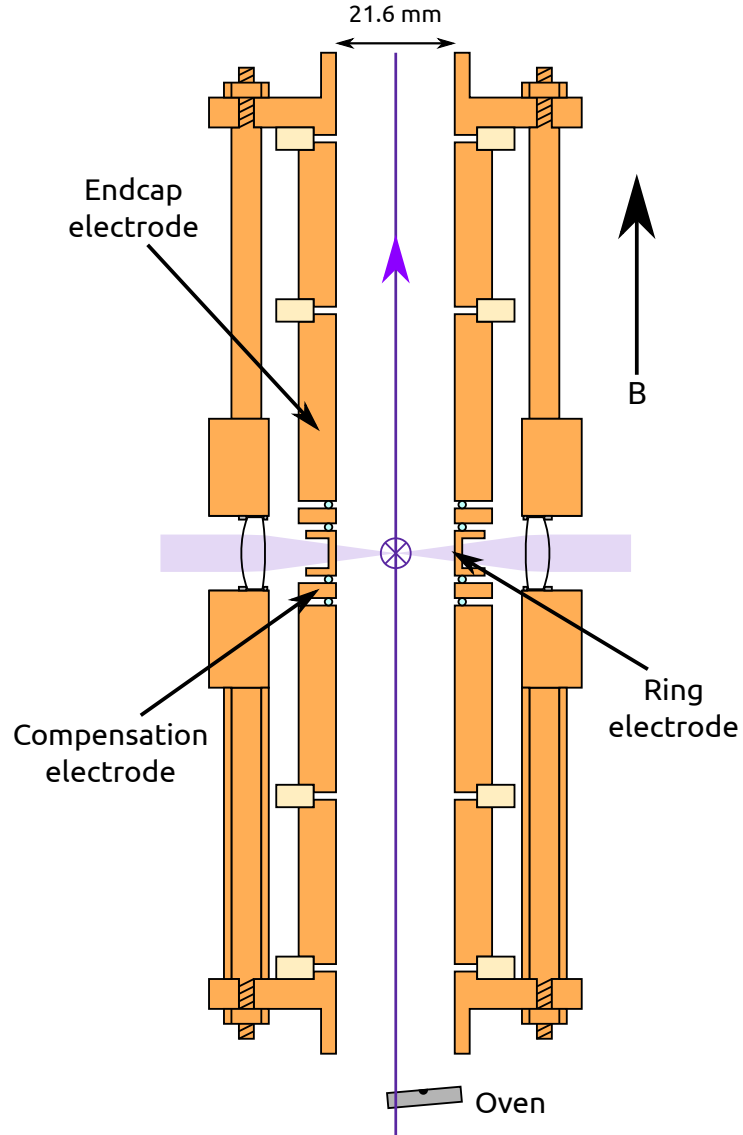


Figure V.1: Cross section of SPECTRAP showing the stacked cylindrical electrodes. To trap cations, a positive voltage is applied on the endcaps and the ring electrode is tied to ground. The purple arrows indicate the direction of propagation of the laser beams. Fluorescence from the ions is collected through apertures in the ring electrode via lenses with a 20 mm focal length. Reproduced from ref. [90].

to roughly 755.189 THz and 755.259 THz respectively (subject to the calibration of the wavemeter). Their linewidth and drift are unsatisfactory for efficient Doppler cooling and must be reduced via a locking system. The linewidth of the lasers is decreased by locking them to a pair of low finesse optical cavities thanks to a Pound-Drever-Hall (PDH) scheme which provides a feedback signal used to modulate the current of the laser diodes as well as the piezos of the ECDL. Long term drifts due to thermal fluctuation in the low finesse cavities are dealt with thanks to a second locking stage where the lasers are compared to a stable HeNe laser reference in a transfer scanning cavity. The scanning cavity is temperature locked to avoid any change of the refractive index of the air in the cavity due to thermal fluctuations. A feedback is provided to piezoelectric actuators which adjust the length of the low finesse cavities. Before the temperature control was installed, a slow drift of the laser frequencies could be observed which was particularly damaging for long experiments where they need to be stable for several hours. The schematic of the home-built temperature controller is given in appendix C. Details on these lasers and the locking systems can be found in Sandeep Mavadia's [77], Joe Goodwin's [78] and Graham Stutter's [90] theses. Before being sent to the trap, blue 1 and blue 2 are combined in a 50/50 beam splitter so that both frequencies are present in the same beam and split into two paths: one will ultimately propagate in the trap along the axis while the other will be in the radial plane.

An important addition (made by Pavel Hrmo and me) to the laser system was the installation of an extra AOM in the radial beam path (see figure V.2) which diffracts the beam when turned on, reducing the optical power sent to the trap. This has the advantage of drastically reducing scatter but at the expense of degraded radial cooling. It is especially useful for experiments on a single ion where the fluorescence level is low and hard to distinguish from background. Because the two beams coming from blue 1 and blue 2 have different divergences and foci and generally poor beam qualities, no less than five additional lenses were necessary to get a good diffraction efficiency and coupling to the fibre going to the trap.

The fibres carrying the light to the trap are polarisation maintaining (PM). As mentioned in section IV.2, the excitation of the optical transition $S_{1/2} \leftrightarrow P_{1/2}$ depends on the polarisation of the laser light and care is taken to send the correct one to the trap. PM fibres are however not perfect and introduce some ellipticity in the polarisation which may eventually affect the fluorescence level from the ions. We witnessed in particular a strong dependence on temperature when the fibre was heated artificially. Natural fluctuations in the room temperature do not seem to be a major problem however.

In addition to the 397 nm lasers are the two home-built repump lasers at 854 nm and 866 nm used to depopulate the $D_{5/2}$ and $D_{3/2}$ states respectively which output approximately 20 mW and 50 mW. Both are also locked to a transfer scanning cav-

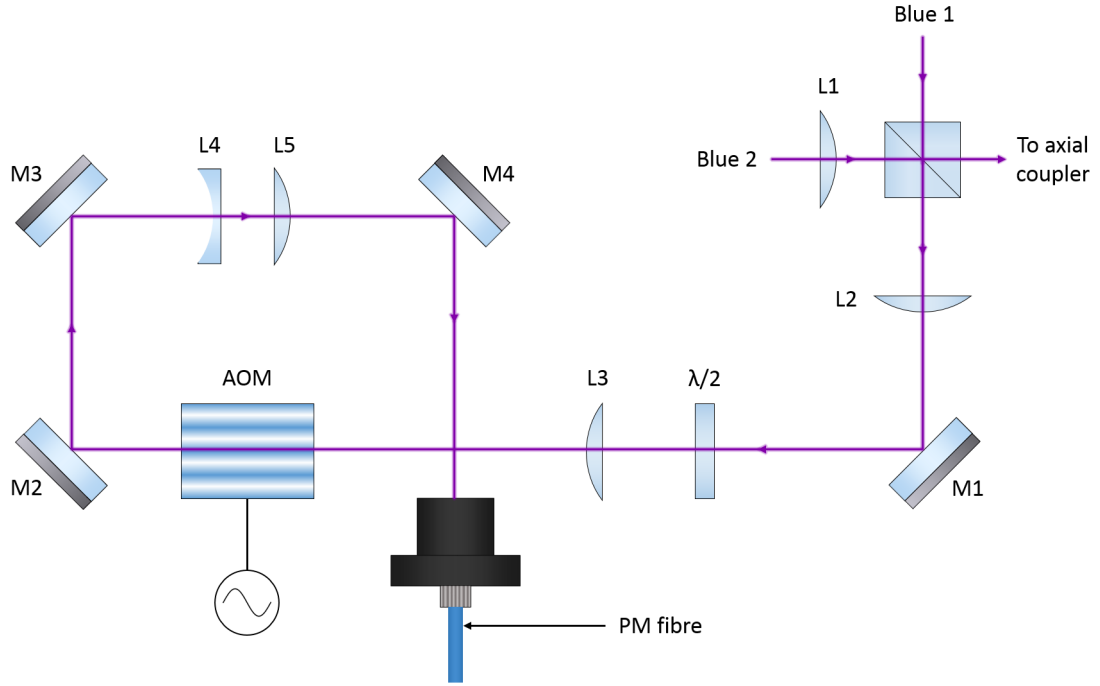


Figure V.2: Schematic of the optics (not to scale) on the radial beam path of the 397 nm cooling lasers featuring an AOM to reduce the intensity by diffracting up to 80% of the light. The diffracted beam is not coupled to the optical fibre and therefore does not go to the trap. The numerous lenses are required to shape the beam and obtain a good diffraction efficiency in the AOM and an efficient coupling to the fibre. Focal lengths of lenses: L1: 1000 mm, L2: 150 mm, L3: 100 mm, L4: -100 mm, L5: 100 mm. Distances: L1-cube: 3 cm, cube-L2: 7.5 cm, L2-M1: 3.5 cm, M1-L3: 24 cm, L3-AOM: 30 cm, AOM-M2: 11.5 cm, M2-M3: 9.5 cm, M3-L4: 7 cm, L4-L5: 12.5 cm, L5-M4: 12.5 cm, M4-coupler: 10 cm.

ity - with the same HeNe reference as for the blues - to prevent frequency drifts. The feedback is provided to the ECDL piezoelectric transducers. Again more details can be found in the theses cited above. The light from these lasers is sent to a fibre electro-optic modulator (EOM) to create a frequency comb in order to cover all the Zeeman sub-levels. All laser beams go through AOMs in double-pass configuration; the first order diffracted beams are coupled to fibres going to the trap allowing for fast switching of the lasers.

V.3 Spectroscopy Laser and Associated Devices

The laser used for spectroscopy, sideband cooling and coherent manipulation is perhaps the most critical element of the experiment. To be able to resolve the motional sidebands of the ions and drive coherently and repeatedly the transition $S_{1/2} \longleftrightarrow D_{5/2}$ imposes strong requirements on the frequency, linewidth and power of the laser light. The typical trapping frequency in our experiments is of the order of 100 kHz which means that the linewidth of the laser must be much smaller than this. For experiments with several ions, the spacing between the sidebands of different modes can be just a few kilohertz which further increases the constraints on the linewidth. The optical power at the trap must be high enough to efficiently sideband cool and to drive the ions faster than the coherence is lost. Besides, the power should be stable, at least over the course of an experiment, to be able to repeat a measurement. To scan a spectrum accurately and address a particular sideband requires the frequency to be tuned with sub-kilohertz precision. Figure V.3 gives an overview of the system.

Narrow linewidth spectroscopy laser

The laser system is based around an ECDL with a frequency of approximately 411.2 THz locked to a high finesse cavity ($F \approx 60000$) thanks to the PDH technique. The optical cavity is built with ultra-low expansion glass and maintained at a constant temperature where thermal fluctuations have no influence on the length of the cavity to first order. In order to avoid the detrimental effects of acoustic vibrations on the linewidth, the breadboard of the laser system rests on bicycle inner tubes; despite being a rather rustic technique, it is satisfactory for our purpose and the resulting linewidth is just under 1 kHz. This figure is estimated from a measurement of the minimum achievable absorption linewidth of the $S_{1/2} \longleftrightarrow D_{5/2}$ transition of $^{40}\text{Ca}^+$ which has a natural linewidth of about 0.136 Hz [73] and from Ramsey experiments detailed in chapter VIII. It is not a direct measurement of the laser linewidth like a self-heterodyne measurement and therefore may reflect other effects (magnetic field noise in the trap for example). The laser diode outputs nearly 20 mW which, considering the light has to go through several optical elements and

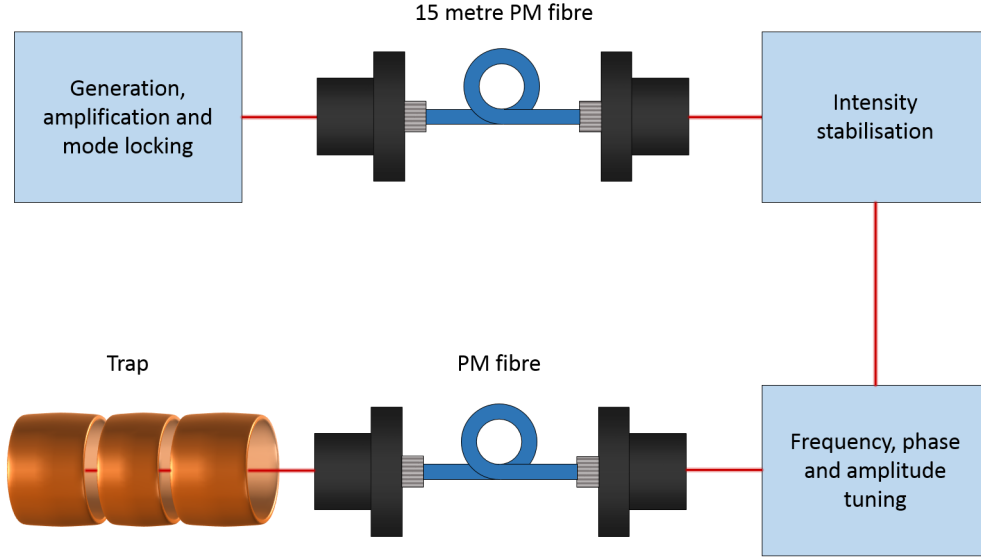


Figure V.3: Diagram of the spectroscopy laser system. The laser light is produced by an ECDL, locked to a high finesse cavity and amplified before it is sent via a 15 metre long polarisation maintaining fibre to the room where the Penning trap is located. There, the laser light goes through an intensity noise cancellation system and an acousto-optic modulator to tune the frequency, phase and amplitude of the light before being sent via another short fibre to the trap. Depending on the experiment, the AOM can be set-up in single or double pass configuration.

fibres before reaching the trap, is not enough to get an acceptable Rabi frequency. The laser light is therefore amplified with a tapered amplifier i.e a semiconductor gain medium which increases the laser power by close to 14 dB (10 to 250 mW) while preserving the spectral quality. Joe Goodwin describes this laser system in more details in his thesis [78]. There was however one major modification since: the system used to employ two laser diodes called master and slave; the latter was used as a gain medium to first amplify the light from the former. Optimisations on the optical set-up, including fine alignment of optical isolators and the replacement of optics, has made the slave diode redundant since.

Intensity stabilisation (noise eater)

A power stabilisation system or “noise eater” developed by Manoj Joshi and Pavel Hrmo ensures a power noise level below 0.5 %. This device works by measuring the intensity of the zeroth order (non diffracted) beam of an acousto-optic modulator and comparing it to a reference level. Any difference between the measured signal and the reference gives rise to an error signal fed to a proportional-integral controller which provides a feedback control signal used to modulate the output amplitude of the AOM radio-frequency driver. The power of the zeroth order beam decreases when that of the acoustic wave in the AOM increases. The acoustic power is determined

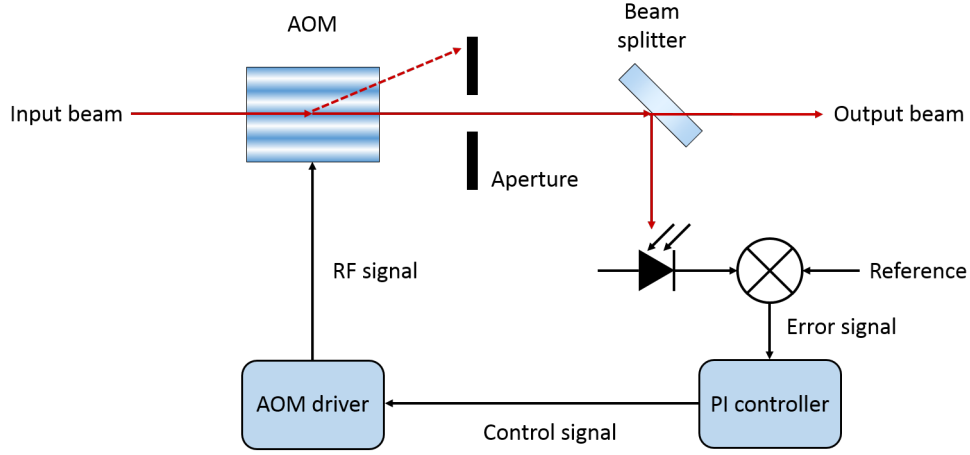


Figure V.4: Schematic of the noise eater. The incoming laser light goes through an AOM and a fraction of it illuminates a fast photodiode to measure its intensity. The voltage output of the photodiode is compared to a reference level to produce an error signal used to modulate the output of the radio-frequency generator that drives the AOM. Part of the light going to the AOM is diffracted in the first order to maintain the power in the non diffracted beam constant.

by the RF amplitude; hence the optical power at the AOM output can be maintained at a fixed level by modulating the RF amplitude. A schematic of the system is shown in figure V.4. Note that the intensity noise at the trap may be higher than what is measured at the output of the noise eater since the laser beam goes through several optical elements including a fibre afterwards.

Tunable AOM driver (direct digital synthesiser)

The properties of the laser light (frequency, phase, amplitude) can be adjusted thanks to an acousto-optic modulator to meet the requirements of our experiments, for instance to change the frequency to take a spectrum or address different sidebands for sideband cooling. The light scattered out of an AOM is the result of wave mixing between the incoming light and a sound wave travelling in the crystal of the AOM. The sound wave is produced by a piezoelectric transducer fed by a radio-frequency electric signal with a frequency typically of a few tens of MHz to a few hundreds of MHz.

I developed an AOM driver based on direct digital synthesis (DDS) capable of generating such radio-frequency signals (the full schematics are in appendix C). This driver replaced a system made of three function generators and RF switches previously in use. It was designed to be fully integrated with the rest of the experimental set-up and is controlled by the same software used to communicate with the field programmable gate array (FPGA) that controls the experiment as explained in section V.5. Thanks to DDS, the driver allows the frequency, the phase and the amplitude of its output signal to be finely tuned and the output can be switched

between different pre-set signals very rapidly. Both the computer and the FPGA exert some control on the AOM driver in a similar way as they did with the system that the driver replaced: the computer is used to set up the driver while the FPGA is responsible for the switching part.

Direct digital synthesis is a means to generate tunable signals (frequency, phase and amplitude) from a fixed frequency reference called the clock. The main elements of a direct digital synthesiser are a phase accumulator, a phase to amplitude converter and a digital to analogue converter (DAC). The clock is fed to the phase accumulator which is similar to a counter. At each clock cycle, the counter is incremented by an integer quantity called the frequency tuning word (FTW) until it overflows because it has reached its maximum value. The time needed to reach the maximum value of the counter corresponds to the period of the output signal. Its frequency is thus given by:

$$f_0 = \frac{\text{FTW} \cdot f_c}{2^N} \quad (172)$$

where f_c is the clock frequency and N is, in bits, the capacity of the phase accumulator. The frequency resolution is therefore given by $f_c/2^N$ and the maximum output frequency is $f_c/2$ as indicated by the Nyquist-Shannon theorem [91]. In practice it is often limited to 40 % of the clock frequency however. A lookup table is used to then transform the values of the phase accumulators into amplitude values fed to the DAC to generate the analogue output signal.

Our system is based on a AD9910/PCBZ board from Analog Devices and an Arduino Mega 2560 micro-controller. The board features an AD9910 DDS chip able to generate signals up to 400 MHz when using a 1 GHz clock with a resolution of 0.23 Hz ($N = 32$). For technical reasons explained below, the frequency range of our device is software limited to 100 MHz to 300 MHz and the resolution to 1 Hz which is more than enough given the linewidth of the spectroscopy laser.

The output power from the on-chip 14-bit DAC varies greatly and non linearly with frequency: about -33 dBm at 1 kHz to 0 dBm at 100 MHz. On the range 100 MHz to 300 MHz, the variation is limited to around 2 dBm. This is however still unsatisfactory given the power of the radio-frequency signal ultimately determines the power of the light sent to the trap which should not depend on frequency. Besides, before going to the AOM, the signal undergoes a two-stage amplification and goes through several radio-frequency components (switches, mixer, attenuators), all of them with different frequency responses. To solve this issue, we use the following method: the amplitude at the output of the DAC is given by:

$$A(f, \text{ASF}) = \frac{\text{ASF} \cdot A_{\text{max}}(f)}{2^{14} - 1} \quad (173)$$

where the amplitude scale factor (ASF) is an integer tuning word which takes values between 0 and $2^{14} - 1$. This is the parameter the DDS chip uses to change the output amplitude. $A_{max}(f)$ is the maximum output amplitude at a given frequency. We measure this maximum amplitude for the range 100 MHz - 300 MHz, look for the minimum and use it as the reference, or normalised output amplitude for the range; and we calculate the coefficient:

$$\alpha(f) = \frac{\min[A_{max}(f), \text{range}]}{A_{max}(f)} \times 2^{14}. \quad (174)$$

To obtain a signal with an amplitude that is a fraction β of the normalised amplitude, at a given frequency the ASF is given by $\alpha(f) \times \beta \times 2^{14}$. This is the value to supply the DDS with. The drawback of this method is that the amplitude resolution is lowered for frequencies that do not correspond to the minimum output amplitude because only a part of the possible 2^{14} values of the DAC is used. In practice, given the relatively small variation in the non normalised output over the range of interest, it is not an issue owing to the good resolution of the DAC. The maximum power of the signal going to the AOM is normalised to 33 dBm (2 W), enough to drive it at maximum diffraction efficiency.

The spectral purity of the signal at the output of the DAC is excellent: no spurious component can be seen on a spectrum analyser. After amplification, the first harmonic remains low at -30 dBc and is outside the frequency response range of the AOM anyway. The frequency stability is only limited by that of the reference clock (Hewlett-Packard HP8643A in our case).

The DDS is used in “single tone” mode which means it outputs a sinusoidal frequency with fixed frequency, phase and amplitude by contrast to e.g. a “ramp” mode where a parameter varies with time. It is however possible to switch very rapidly (a few ns) between eight single tone “profiles” where each profile may have different frequencies, phases and amplitudes. The profiles are loaded and stored in on-chip registers.

It is also possible to create bichromatic signals by modulating the DDS output with another signal (provided by an external source) in a mixer installed in the driver. This feature is however rarely used as we realised that the resulting signal always contain a carrier frequency that cannot be suppressed to low enough levels.

The other key element of the system is the Arduino micro-controller which makes the link between the DDS chip and the computer used to conduct the experiments. The micro-controller communicates with the DDS via a serial peripheral interface (SPI) bus and sends the appropriate signals to configure the DDS and the data containing the profiles information. On the computer end, a graphical user interface (GUI) written in C# allows the user to enter the desired parameters for each profile. The data is sent to the micro-controller via USB which transforms it in an understandable way for the DDS (figure V.5). The links between the computer, the

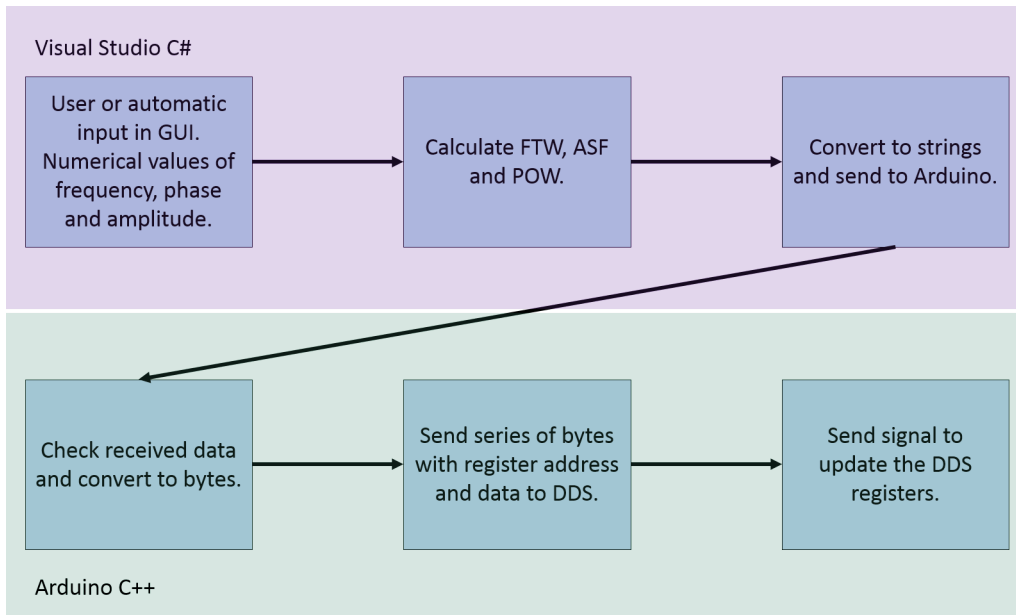


Figure V.5: Writing process of the DDS profiles. FTW: frequency tuning word, ASF: amplitude scale factor, POW: phase offset word.

Profile	P2	P1	P0
0	0	0	0
1	0	0	1
2	0	1	0
3	0	1	1
4	1	0	0
5	1	0	1
6	1	1	0
7	1	1	1

Table V.1: Logic levels to apply on pins P0, P1 and P2 of the AD9910 chip for each profile.

FPGA and the AOM driver are summarised in figure V.6.

An issue with profile switching was discovered after the AOM driver was commissioned and required a small modification of the system. To each profile corresponds a three-bit binary code written by three TTL signals (P0, P1, P2) sent from the FPGA (see table V.1). Changing the TTL logic levels changes the profile. For the profile switching to work correctly, the electric signals from the FPGA have to all arrive on the pins of the DDS chip within one cycle of the system clock of the DDS (250 MHz) i.e. 4 ns. This is a stringent timing requirement which is not met by our system. The consequence is occasional faulty switching where the profile activated is not the one desired. A possible way to avoid this is to change only one logic level at a time. In other words, when going through a sequence of profiles during an experiment, successive profiles should differ by only one bit e.g. 0 to 4 but not 1 to 2. This method is known as reflected binary code or Gray code. Although perfectly

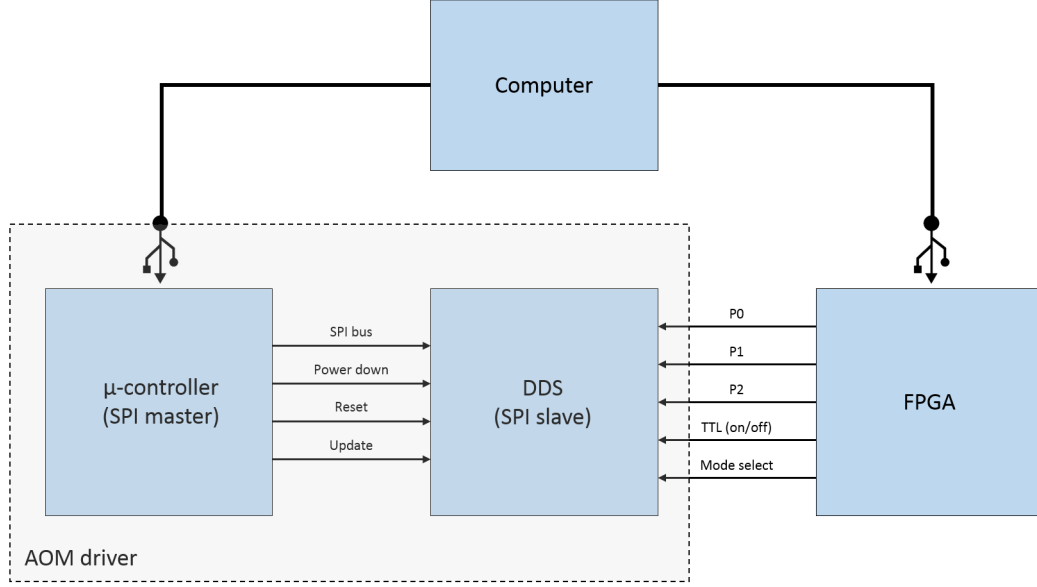


Figure V.6: Links between the AOM driver, the computer and the FPGA. The micro-controller and the FPGA are controlled from the computer via USB connections. The micro-controller communicates with the DDS thanks to a SPI bus as well as other signals to update, reset and power down the DDS. The TTL signals P0, P1 and P2 from the FPGA perform the profile switching (3 logic signals for $2^3 = 8$ profiles). Two other TTL signals are used to change the state of RF switches to turn on and off the radio-frequency output from the driver and change its mode of operation (modulated or non modulated signal).

reliable, this technique is rather inconvenient as one needs to make sure that the profile sequences are Gray coded when designing an experiment. The solution we adopted instead is to add delay circuits between the FPGA and the DDS: the P1 signal is delayed by 50 ns and the P2 by 100 ns. This gives enough time to change the logic level independently and so to switch profile reliably. This is effectively an automatic Gray coding: to go from profile 0 to 7, the DDS will first switch to profile 1, then 3 and finally 7 but the person designing the sequence does not have to worry about it. Even though the DDS switches to undesired profiles in the process (1 and 3 in our example), it is not a problem for all practical purposes as it happens on a short time scale; too short to have any significant detrimental effect.

V.4 Other Optical Systems

Light guiding to the trap

The laser beams are conveyed by single mode polarisation maintaining fibres from the optical table to the breadboard located under the superconducting magnet. Because of their large wavelength difference, the blue cooling lasers and the NIR repump lasers are carried by different fibres and are combined at the breadboard by a dichroic mirror. Axial and radial beams travel in separate fibres. The axial beams

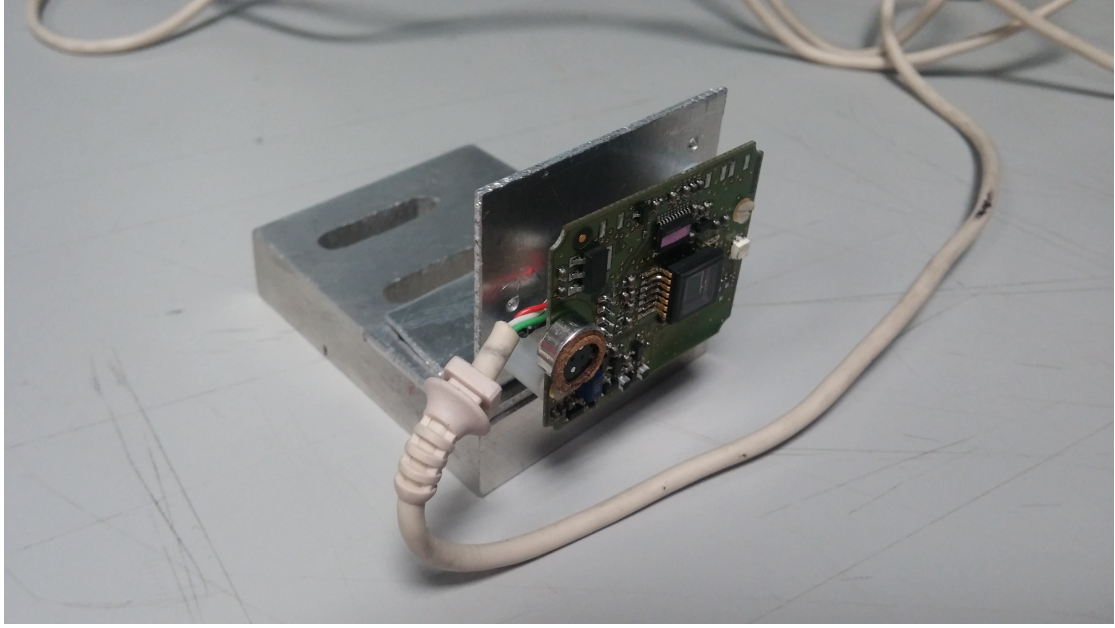


Figure V.7: Camera used for radial beam positioning. Philips webcam stripped of its case and lenses and mounted on an aluminium plate. The laser beam is directly impacting the CMOS photodetector. A Matlab code is used to create a graphical representation of the beam position on the detector.

are directed upwards directly to the trap centre and through the vacuum chamber by a mirror located underneath while the radial beams travel up the magnet bore outside the chamber before being bent at a right angle by a mirror to enter the trap radially through one of the apertures on the ring electrode. The position of the radial beam can be adjusted thanks to a motorised mirror mount located on the breadboard and monitored thanks to a modified webcam (figure V.7) hit by the beam after it exits the trap through the opposite aperture.

Fluorescence detection

Fluorescence light from the ions is detected thanks to two photomultiplier tubes (PMT) or alternatively one PMT and a EMCCD⁵ camera (Andor iXon Ultra 897). The light is collected from two of the circular apertures of the ring electrode and collimated by lenses. One path always leads to a PMT via a multimode optical fibre while the other gives the possibility to send the light to either another PMT, also via optical fibre, or the EMCCD camera. In the latter case, an imaging system directs the light to the camera which is installed away from the trap to avoid perturbations from the magnetic field. The magnification of the imaging system is such that one pixel on the camera corresponds to about $1.32\text{ }\mu\text{m}$ in the focal plane. This is determined by measuring the distance between two ions in a chain on the camera. For a given axial frequency, the inter-ionic distance can be calculated as we saw in section II.3 and used for calibration.

⁵Electron Multiplying Charge Coupled Device: a type of amplified CCD

In our set-up, fluorescence is the only way to confirm the presence of the ions as no electronic detection is available. The image from the camera as well as the fluorescence count from the PMTs is a great help to determine the number of ions in the trap, the goodness of Doppler cooling and to observe the spatial configuration of the Coulomb crystals. Fluorescence is also used to determine the electronic state of ions during spectroscopy as explained in section V.6.

V.5 Control Systems

Spectroscopy, sideband cooling and coherent manipulation experiments rely on a control system based on a computer and a field programmable gate array. The primary role of the FPGA is to control different elements of the experimental set-up by switching the state of its TTL outputs. It can also collect fluorescence information from the PMTs in the form of a photon count and store it on its SDRAM⁶. The TTL outputs are used to control the state of radio-frequency switches and attenuators to determine whether AOMs are powered or not and so whether laser beams are sent to the trap or not. They are also used to select the active profile of the DDS.

The computer features a graphical user interface (GUI) - the same as the one of section V.3 - where experiments are designed and conducted. The software, developed internally, is written in C# in the Microsoft Visual Studio environment and has undergone many updates since its initial release to adapt to the changes on the experiment and to improve its functionalities. It allows the user to configure the 729 AOM driver by writing in the DDS registers to specify the frequency, phase and amplitude of each profile; to control the EMCCD camera and visualise its image and to manually switch on and off the lasers by sending a command to the FPGA to change the state of its outputs.

All experiments consist of sequences of several stages where various actions are carried out. Such actions may be:

- turning on and off lasers (via AOMs)
- turning on and off the axialisation drive
- turning on and off the radial beam AOM
- switching the DDS profiles
- collecting fluorescence (counting).

Several actions can be done at every stage. Each sequence is usually repeated hundreds of times to get statistically significant results. This repetition of identical sequences constitutes one step of the experiment or, in other words, a data point.

⁶Synchronous dynamic random-access memory.

An experiment can have anything between less than ten steps to several hundred. Between two steps, a parameter, the variable of the experiment, is modified. This varying parameter can be for instance the frequency of the 729 nm laser for spectroscopy, its phase or the duration of a laser pulse e.g. for Rabi experiments. The quantity measured is however always the occupation probability of the $D_{5/2}$ state. The sequence to be performed is constructed by the user on the GUI, saved (.xml format) and uploaded to the FPGA in a readable format (.hex). It always contains stages where the photons detected by the PMTs (or EMCCD camera) are counted to determine the occupation probability of the $D_{5/2}$ state as explained in section V.6. The fluorescence data are stored on the SDRAM of the FPGA. Before moving to the next step, the data are sent to the computer and saved in a data file. The SDRAM is then cleared and the experiment moves on. Upon reception of a start signal from the computer at the beginning of an experiment, the FPGA goes through the steps until the experiment is completed or until the user stops it. Meanwhile the GUI on the computer displays the data point in real time. The number of steps and the change in the variable between the steps is set by the user. For example, for spectroscopy, start and stop frequencies as well as a frequency increment are specified in the GUI before starting the experiment. In that case, between each step, one of the profiles (profile 0) of the DDS is updated automatically with a new frequency. In order to avoid potential variations between the repeated sequences of a data point due to the oscillations of the mains electric supply, the FPGA can be configured to trigger the sequences at a given phase of the mains cycle. It is however unclear whether this has any significant effect. Alternatively, the trigger can come from the EMCCD camera, if used instead of one of the PMTs. This is done because for technical reasons internal to the camera (the way it refreshes its detector), the lowest noise in the fluorescence signal is achieved if the camera is used as the reference to synchronise the experiment. The FPGA also receives logic signals from the locking systems of the lasers so that if one or several lasers get unlocked during an experiment, the affected data points can be identified and discarded. These logic signals can also be used as triggers to stop the experiment if a laser unlocks.

Besides its role in running experiments, the computer controls directly various other devices via dedicated programs, not used during experiments and so not connected to the FPGA in any way. Those include the pulse box and the motorised mirror mentioned earlier. The Arduino micro-controller of the pulse box is also used to control servomotors used as mechanical shutters to block the different laser beams (axial blue, radial blue and NIR) going to the trap. These servomotors are slow and not suitable to create short pulses in an experiment but are convenient for diagnostics. The communication between this Arduino and the computer is done by means of a simple GUI, written in the C# language. The links between the different devices of the set-up are shown in figure V.8.

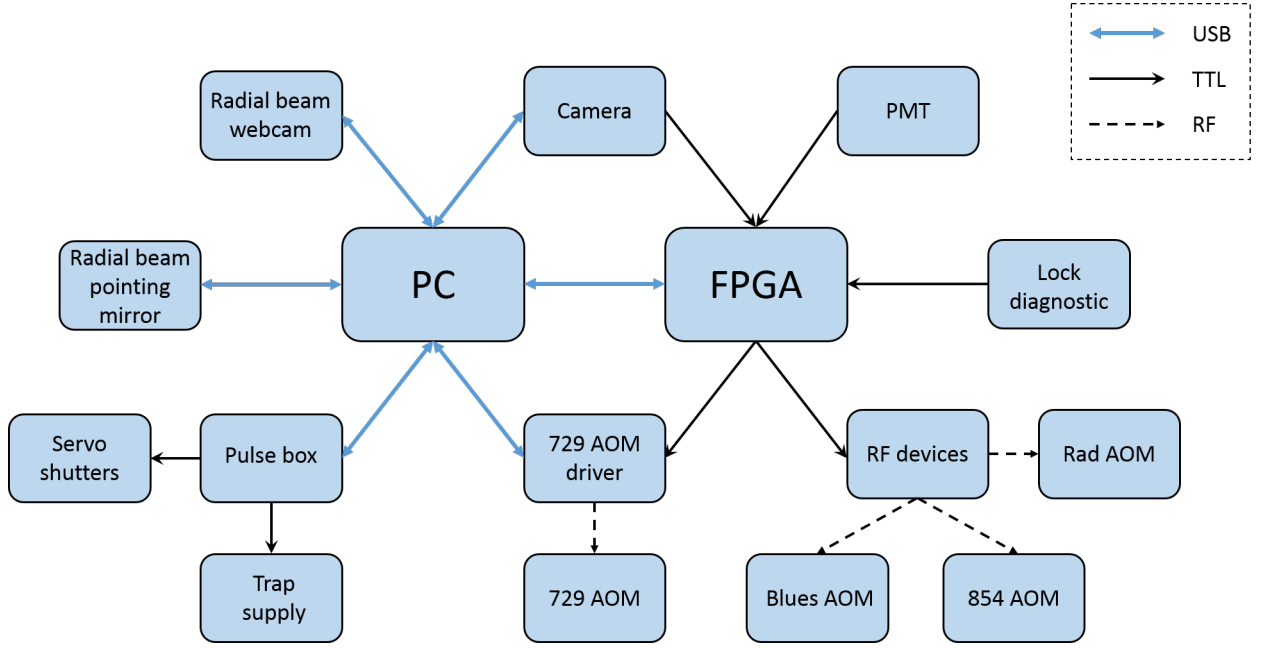


Figure V.8: Interactions between the elements of the experimental apparatus.

V.6 Resolved Sideband Spectroscopy

As an illustration, let us take the example of resolved sideband spectroscopy, which is one of the simplest types of experiment done in our laboratory. Spectroscopy on the $S_{1/2} \longleftrightarrow D_{5/2}$ transition is used to study the sideband structure of one or several ions to access valuable information on their vibrational state after Doppler cooling. It is also a prerequisite for sideband cooling where the vibrational sidebands must be targeted with a good accuracy on the frequency. Resolved sideband spectroscopy requires the ion to be in the strong binding regime. In our case, the typical trapping frequencies are hundreds of kHz for the axial and the modified cyclotron and some tens of kHz for the magnetron motion while the natural linewidth of the $S_{1/2} \longleftrightarrow D_{5/2}$ transition is 0.136 Hz and that of the laser about one kHz.

Spectroscopy experiments seek to determine the probability of excitation of the $S_{1/2} \longleftrightarrow D_{5/2}$ transition as a function of the laser frequency. The ion is initially Doppler cooled and prepared in the $m_j = -1/2$ Zeeman sub-level of the $S_{1/2}$ state by turning off one of the blue cooling lasers. The 854 nm repump laser is turned off and a pulse of the 729 nm spectroscopy laser is then applied to try to shelve the ion to the $D_{5/2}$ state. Because of the very low decay rate of the $D_{5/2}$ state, measuring the probability of excitation cannot be done by detecting the corresponding decay photons at 729 nm. Instead, after the 729 nm pulse, both the 397 nm lasers are turned on but the 854 nm laser kept off. If the ion was driven to the $D_{5/2}$ state by the 729 nm pulse, the cooling transition $S_{1/2} \longleftrightarrow P_{1/2}$ is not excited and no fluorescence is observed. This provides a binary test (fluorescence or no fluorescence) which is repeated many times (typically 100 to 400 times) to obtain a probability of excitation

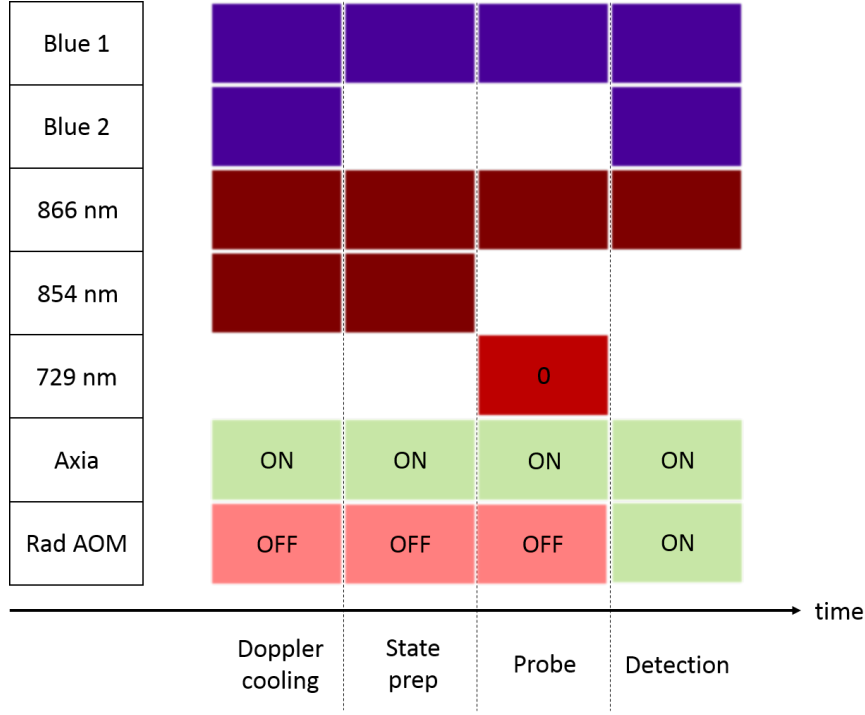


Figure V.9: Sequence for resolved sideband spectroscopy of the axial motion on a Doppler cooled ion. An empty square means the laser is off. Typical pulse durations are 8 to 12 ms for Doppler cooling and detection, 100 μ s for the state preparation. The duration of the probe pulse depends on the 729 nm laser intensity and usually ranges from 10 to 200 μ s. The number in the 729 nm box indicates the DDS profile of the 729 AOM driver in use which determines the laser frequency. For axial spectroscopy, the axialisation drive is on throughout. The radial AOM is turned on during the detection to reduce the intensity sent to the trap and therefore the background on the PMT signal. The pulse durations are not to scale on the figure.

for a given frequency. The frequency of the spectroscopy laser is stepped and the above procedure repeated to obtain a spectrum. The pulse sequence for spectroscopy can be seen in figure V.9 and figure V.10 gives an example of a spectrum obtained with this method. In this example, a single ion was Doppler cooled for 8 ms and the axial motion was probed. The spectrum exhibits the characteristic sideband structure as explained in section III.2.

Windowing

In order to save time, some spectra including the one in figure V.10 are ‘windowed’ i.e. data points are taken only around the sidebands. The regions where no excitation is expected are skipped. Since there is a known relation between the trapping voltage and the axial frequency; if the position of the carrier is known, there is no need to scan the whole spectrum to find the sidebands.

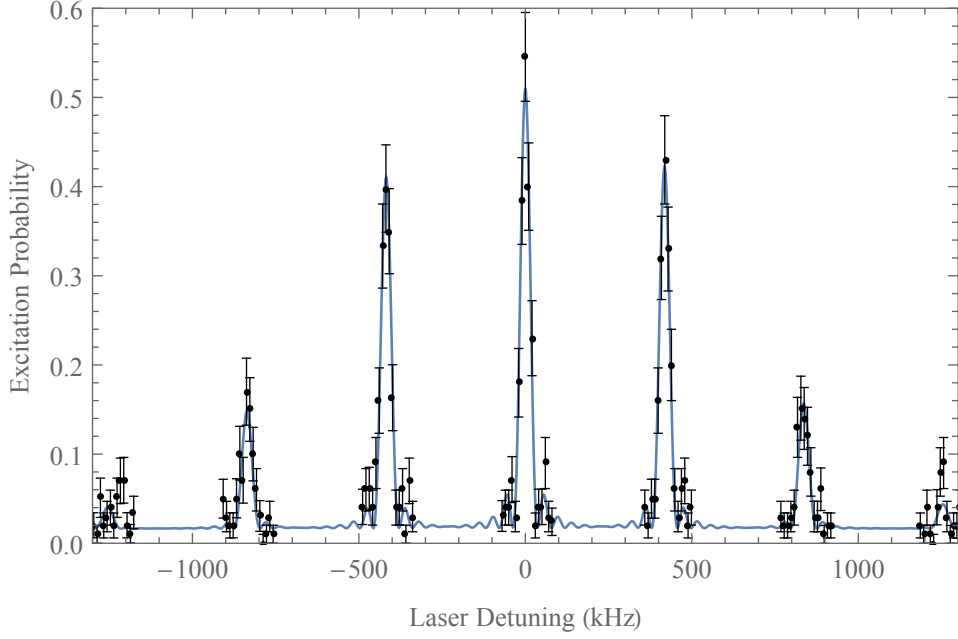


Figure V.10: Axial motional spectrum of a single ion after 8 ms of Doppler cooling. The axial frequency is $\nu_z = 418$ kHz ($\eta = 0.15$, trapping voltage 250 V). The blue line is a fit to the data points giving an average phonon number $\bar{n} = 34(3)$ and a Rabi frequency of roughly 19.4 kHz.

Interleaved spectra

Two spectra or more (up to four) can be recorded simultaneously by concatenating sequences like the one in figure V.9. The spectra are said to be interleaved. Each sub-sequence contains one detection stage to which corresponds a data point. The sequences may be different, allowing for comparisons of spectra taken with experimental conditions as identical as possible, apart from the ones deliberately made different.

State determination

To decide whether an ion is fluorescing or not, a threshold on the number of photons counted by the PMT or camera has to be set. In the absence of fluorescence, a background signal is detected by the PMT. In our system this background is mostly due to laser light scattered from the electrodes of the trap and the windows of the vacuum chamber. Only the 397 nm lasers contribute to the background as the other wavelengths are filtered out. The radial beam contributes a bigger part than the axial one. The background and the fluorescence from an ion follow a Poisson distribution: if λ is the average number of photons counted over many identical trials, the probability to observe n photons is

$$P(n) = e^{-\lambda} \frac{\lambda^n}{n!}. \quad (175)$$

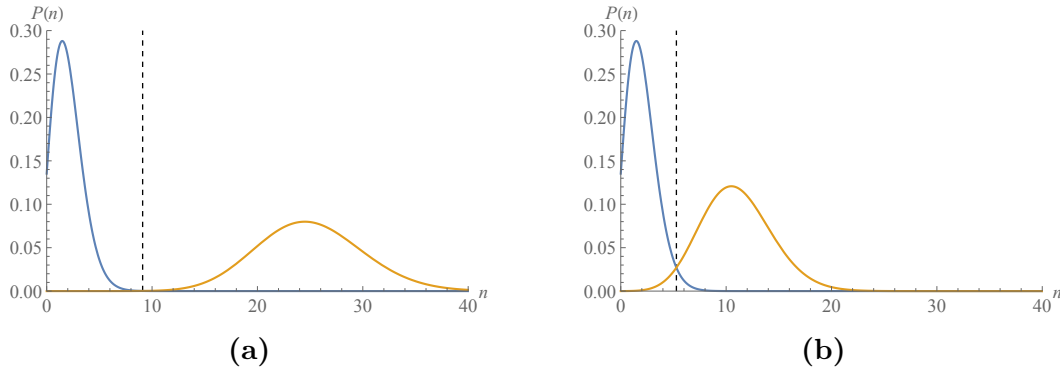


Figure V.11: (a) Dark (blue) and bright (yellow) distributions represented by Poisson distributions with respective averages of 2 and 25. The parts of the dark and bright distributions respectively above and below the threshold (black dashed line) are about 0.02 %. (b) Bright distribution with an average of 11. 5.27 % of the dark distribution is above the threshold, 3.75 % of the bright under.

We call respectively dark and bright the distributions in the absence and presence of fluorescence. The bright distribution has of course a higher average. A possible threshold is the value of the number of photons n_T for which the probabilities are equal for the bright and dark distributions:

$$\begin{aligned}
 P_{dark}(n_T) &= P_{bright}(n_T) \\
 \Rightarrow n_T &= \frac{\lambda_{bright} - \lambda_{dark}}{\ln(\lambda_{bright}) - \ln(\lambda_{dark})}.
 \end{aligned} \tag{176}$$

When performing spectroscopy, if the number of photons counted during the detection stage (see figure V.9) is lower than the set threshold, it will be registered as a dark count and so the ion will be interpreted as shelved in the $D_{5/2}$ state. This detection scheme is not infallible as it wrongly considers the ion shelved for the part of the bright distribution below the threshold and vice versa. This can be significant if the overlap between the two distributions is large (see figure V.11). The part of the bright distribution below the threshold leads to a background in the spectrum while the parts of the dark distribution above the threshold decreases the apparent probability of excitation.

The number of photons collected depends on the duration of the detection. If we call R_d the rate of photon collection from the background (d for dark) and R_f that from fluorescence, then the average numbers of photons collected over a detection period t_D simply are $\lambda_{dark} = R_d t_D$ and $\lambda_{bright} = (R_d + R_f) t_D$ respectively for a dark and bright ion. The obvious way to avoid that the distributions overlap is thus to increase the detection time. However, an issue inherent to Penning traps, that is the decay of the $P_{1/2}$ state to the $D_{5/2}$ due to j -mixing, limits the benefits of a long detection time. Because the 854nm laser is off during the detection, the cooling cycle is not perfectly closed. If the ion has not been shelved by the 729nm laser pulse, we normally expect to collect fluorescence during the detection but there is

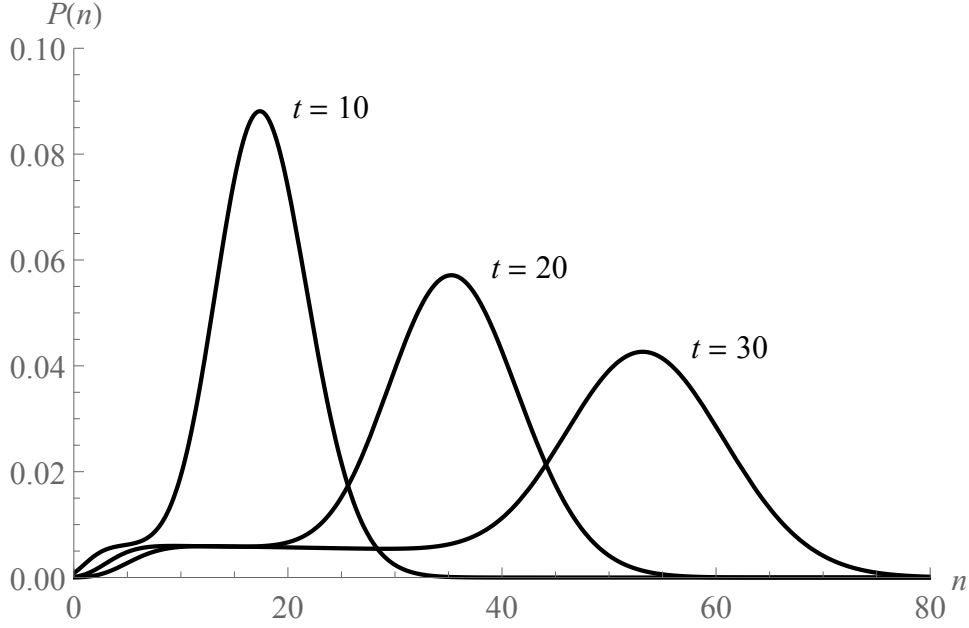


Figure V.12: Bright distribution in the presence of j-mixing for detection times $t = 10$, $t = 20$ and $t = 30$, a decay time $\tau = 100$, $R_f = 1.6$ and $R_d = 0.2$.

a chance that the ion decays to $D_{5/2}$ where it will not be re-pumped. In this case the ion appears as dark and only background light will be collected. The longer the detection time, the higher the probability of decay to $D_{5/2}$. The resulting bright distribution with a detection time t taking j -mixing into account is [90, 92]:

$$P_{\text{bright}}(n, t) = e^{-t/\tau} e^{-(R_d+R_f)t} \frac{((R_d + R_f)t)^n}{n!} + \frac{1}{R_f \tau} \int_{R_d t}^{(R_d+R_f)t} e^{-(\lambda-R_d t)/(R_f \tau)} \frac{e^{-\lambda} \lambda^n}{n!} d\lambda \quad (177)$$

where τ is the characteristic decay time to the $D_{5/2}$ state. Plots of equation 177 are shown in figure V.12. The curves show a plateau for low n which will always overlap with the dark distribution. Besides, increasing the detection period prolongs the overall duration of an experiment which is undesirable in our set-up where the blue lasers are rather old and tend to unlock frequently. We find that a detection time of 10 ms offers a good compromise between signal to noise ratio and decay to the $D_{5/2}$ state.

For single ion experiments where the fluorescence is low, one way to improve the state determination is to lower the intensity of the blue radial beam during detection to reduce the background using the AOM system described in section V.2. This also diminishes the fluorescence level but the overall effect is positive with a smaller overlap between the dark and bright distributions. Figure V.13 shows histograms of the photons counted during detection periods. They were obtained experimentally by exciting the carrier transition of a Doppler cooled ion, in one case with the radial AOM turned on during detection, in the other without. We fit

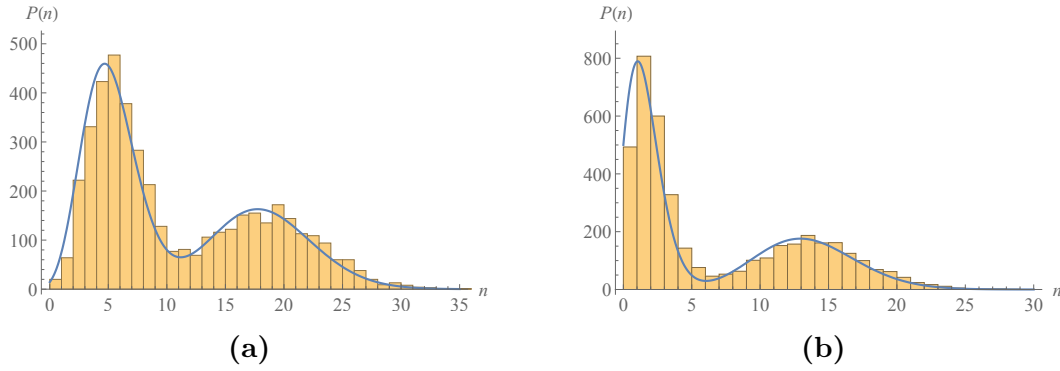


Figure V.13: (a) Histogram of the photon count after exciting the carrier transition of a Doppler cooled ion with a pulse of the 729 nm laser. The radial AOM is off throughout the experiment so that the intensity of the blue radial beam is not lowered during detection. (b) With the radial AOM on during detection, reducing the intensity of the blue radial beam by about 71 %. In both experiments, the detection time is 10 ms. The duration and intensity of the probe pulse was chosen to obtain a probability of excitation of roughly one half. The blue line is a fit to the data point (sum of two Poisson distributions). The Doppler cooling sequences (see figure V.9) were repeated about 4000 times.

the histograms with a sum of two Poisson distributions where we allow for different pre-factors for the two distributions to take into account that the probability of excitation is not exactly one half. Decay to the $D_{5/2}$ state is neglected for the fit which is reasonable given the detection time is only 10 ms. When the radial AOM is not used, the dark distribution is found to have an average of 5.2 and the bright 18.3. This corresponds to 2.67 % of the dark distribution above threshold and 3.93 % of the bright under. With the radial AOM turned on during detection, the dark distribution has an average of 1.6 and the bright 13.5. The overlap is reduced with 1.98 % of the dark distribution above threshold and 0.55 % of the bright below. The diffraction efficiency of the AOM in this example is 71 %. Note that for spectroscopy of the radial motional modes of a single ion, the radial AOM is not used for technical reasons explained in section VI.2.

Spectroscopy on Coulomb crystals

Spectroscopy on Coulomb crystals follows the same procedure as the single ion case but presents specific difficulties. The most notable differences in the sequence are a typically longer and more intense probe pulse (more ions with a lower Lamb-Dicke parameter) and that the radial AOM is always off. The reason for not using the radial AOM is that the configuration of the ions depends, among other things, on the intensity of the radial cooling beam. Therefore, to avoid instabilities of the crystal or even a change of configuration, the blue radial beam intensity should not be reduced during detection.

The main challenge of spectroscopy on ICC comes from the fact that the (N) ions may not all be excited at the same time. We have two devices at our disposal

to collect fluorescence: the PMTs and the EMCCD camera. The PMTs are not position sensitive and therefore only collect a global signal from the ions so that the histogram of photon counts is not simply made of two Poisson distributions but rather a dark distribution and N bright distributions. Although it is sometimes possible to distinguish the dark distribution from the bright ones and thus say when all ions were excited, it is often more challenging to identify the different bright distributions as they are not well separated. At best, it is possible to find how many ions are excited but never which ones. The EMCCD camera is able to form an image of the trapped ions, seen from the radial plane. When the ions are in a chain, they appear static on the camera so that they can be distinguished and individual spectrograms can be acquired for each of them by selecting appropriate regions of interest. For radial crystals however, because of the rotation about the magnetic field axis, ions are indistinguishable except for the central one if present. Another problem with radial crystals is that the intensity of the 729 nm laser is not uniform in the radial plane so that in crystals with several shells (6 ions onwards), the ions closer to the centre are subject to a stronger laser field than the outer ones. The resulting differences in Rabi frequencies makes it very difficult (if not impossible) to extract quantitative information from the spectrogram.

V.7 General Maintenance

The experimental apparatus is a complex system made of a multitude of elements, some prone to failure. To keep the system operational, various servicing operations are required. During my time in the laboratory, I took part in several of them; these included:

- refilling the superconducting magnet regularly with cryogenics: liquid nitrogen every week and liquid helium every four months,
- re-energising the said magnet after a failed liquid helium refill and finding new resonant laser frequencies (the value of the magnetic field changed a little),
- changing the flash lamp of the Nd:YAG laser,
- replacing the reference HeNe laser tube,
- replacing a piezoelectric stack in the blue scanning cavity,
- replacing the current controller of one of the blue lasers,
- fixing or replacing various electronics,
- fixing bugs in the control software.

VI

Sideband Cooling of the Axial and Radial Motional Modes of a Single Ion

In this chapter, I present results of laser cooling of a single ion, both for the axial and radial motions, with a focus on sideband cooling. Aside from the results themselves, a description of relevant experimental methods is given here. Ground state cooling of the axial motion at high frequency, in or close to the Lamb-Dicke regime, was first achieved in our group in 2014. It is discussed in ref. [46] as well as in Joe Goodwin's and Graham Stutter's PhD theses [78, 90]. Since then, we were able to achieve ground state cooling at lower axial frequencies with the ion well outside the Lamb-Dicke regime using two different techniques described below. We also succeeded in cooling simultaneously both the modified cyclotron and magnetron modes close to the ground state.

VI.1 Axial Cooling

Doppler cooling

Doppler cooling is performed by shining the blue lasers at 397 nm and repump lasers at 866 nm and 854 nm tuned close to resonance. A rough tuning of the frequencies is done by adjusting the current of the laser diode and the position of the piezo-electric actuators of the ECDL for the different lasers. Doppler cooling is necessary to detect the ions and detecting the ions is required to set the lasers' frequencies accurately. The wavemeter only gives a rough measurement of the frequencies so the resonant positions have to be found by observing fluorescence from the ions. To this end, the frequency of one of the blue lasers – say blue 2 – is scanned over about 1 GHz to increase the chance of hitting the resonant frequency. Assuming the other lasers are close to resonance, the fluorescence signal as a function of time displays a characteristic sawtooth pattern: the fluorescence increases as blue 2 approaches resonance from the red side of the spectrum and suddenly drops once it passes it

(the ions are then heated). When this signal is observed, the scan can be stopped and the frequency of the blue and NIR lasers adjusted to maximise the fluorescence. This is assuming only one ion is present in the trap, otherwise the additional ones have to be pulsed out first. The lasers are then locked but the frequencies can still be fine-tuned thanks to the scanning cavities' piezoelectric transducers, in steps of 1 MHz for the blues and 0.5 MHz for the NIR. For efficient Doppler cooling, the blue lasers are red-detuned by about 10 MHz.

This section concerns laser cooling of the axial motion but radial Doppler cooling is also active whenever the axial one is. This is necessary to keep the ion close to the centre of the trap in a tight, stable orbit. The radial motion is particularly sensitive to the beam position as explained in section IV.2 and the optimal position depends on the size of the ion cloud. For a single ion, a good position for the radial beam is found by minimising the radial spread i.e. the ion's orbit on the image from the EMCCD camera. To assist the radial cooling, a small axialisation field is applied with an amplitude usually between 100 and 500 mV. It is particularly important at high trapping potentials where keeping the ion in a small orbit just using intensity gradient cooling is challenging. The relatively small amplitude of the axialisation does not add any significant micromotion to the ion.

The intensities of the blue lasers are kept low enough in order not to saturate the transition and avoid excessive recoil heating. Typically, in the axial beam, the power of blue 1 and blue 2 is 3 to 6 μW each and between 7 and 14 μW in the radial beam. The radii at the centre of the trap are around 45 μm for the axial beam and 100 μm for the radial. The radii of the beams are assumed to be roughly equal to the waists of the beams i.e. that the centre of the trap is close to the focal point. The measurement of the waist is done by placing a mirror on the optical breadboard (below the magnet) at a known distance from the trap centre to direct the beam on a camera, also at a known distance from the mirror and measuring the spot size.

Although the fluorescence level and the ion's image on the camera allow finding good cooling parameters, spectroscopy is required to finalise the optimisation and reach the lowest possible temperature. Performing Doppler cooling and recording spectra of Doppler cooled ions are routine experiments and always precede sideband cooling experiments. Reaching the minimum temperature with Doppler cooling which requires a careful adjustment of the lasers' frequencies and powers is usually not our goal but good Doppler cooling facilitates sideband cooling. Resolved sideband spectroscopy from a Doppler cooled ion gives invaluable information about the temperature of the ion and the position of the sidebands. Fitting the data points of a spectrum using the theory developed in chapter III gives access to the average phonon number of the motion. We use a model derived from the probability of excitation of a two level system by near resonant light (equation 89), the expression of the coupling strengths of the sidebands (equation 113), and the thermal distribution

function (equation 118) with the detuning of the laser as the independent variable and the Rabi frequency Ω_0 (as defined by equation 74), the average phonon number $\bar{n}$ and the axial frequency ν_z as free parameters. The probability of excitation on the s^{th} order sideband with a detuning δ from the centre frequency of the said sideband is:

$$P_{e,s}(\delta, t) = \sum_{n=0}^{\infty} \frac{\bar{n}^n}{(\bar{n} + 1)^{n+1}} \times \frac{\Omega_{n,n+s}^2}{\Omega_{n,n+s}^2 + \delta^2} \sin^2 \left(\frac{\sqrt{\Omega_{n,n+s}^2 + \delta^2}}{2} t \right). \quad (178)$$

In practice, for obvious computational reasons, the sum is truncated to a value of n higher than the expected $\bar{n}$ (typically 200 to 400). The time t is the duration of the probe pulse and is a known parameter of the experiment. The probe time and the power of the 729 nm laser are chosen to avoid saturation so that the Gaussian envelope of the sidebands is visible.

The fitted spectrum of a Doppler cooled ion is shown on figure VI.1. Due to the rather low axial frequency (187 kHz), sidebands up to the sixth order are visible and the fit gives an average phonon number $\bar{n} = 57(3)$. This is to be compared with the spectrum in figure V.10 in the previous chapter where the axial frequency was 418 kHz, the average phonon number $\bar{n} = 34(3)$ and where sidebands only up to the third order were visible. These differences between high and low axial frequencies are expected: we know from equations 93 and 144 that the Lamb-Dicke parameter and the Doppler limit go down with increasing axial frequencies. Using the expression of the final average phonon number in the case of an isotropic emission (equation 145), the Doppler limit is $\bar{n} = 56$ at 187 kHz and $\bar{n} = 25$ at 418 kHz. Both values are close to our results which suggests we have an efficient Doppler cooling process. This also gives a foretaste of the difficulty to perform sideband cooling at lower trapping frequencies: the starting point, the average phonon number after Doppler cooling is higher, the number of sidebands is larger and their spacing smaller thus increasing the probability of off-resonant excitations that can lead to heating.

Although necessary to determine $\bar{n}$, Doppler spectra are rarely fitted; a visual evaluation of the sideband heights suffices in practice to assess the quality of the cooling. Importantly, the Doppler spectrum is used to find the positions of the sidebands (their centre frequency) which are essential parameters for sideband cooling. Poor cooling is revealed by a “flatter” spectrum where the heights of the first few sidebands are almost equal. It would be in principle possible to fit the spectra as soon as they are acquired provided a fitting tool was added to the spectroscopy software and enough computing power was available (fitting a Doppler spectrum takes a few minutes on Mathematica with a modern desktop computer) but this was not deemed necessary.

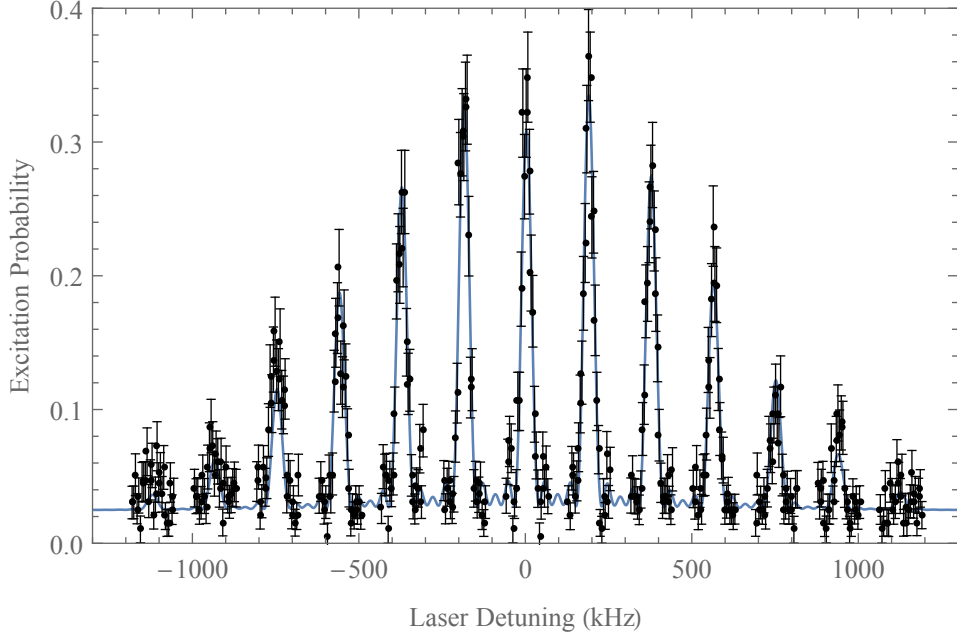
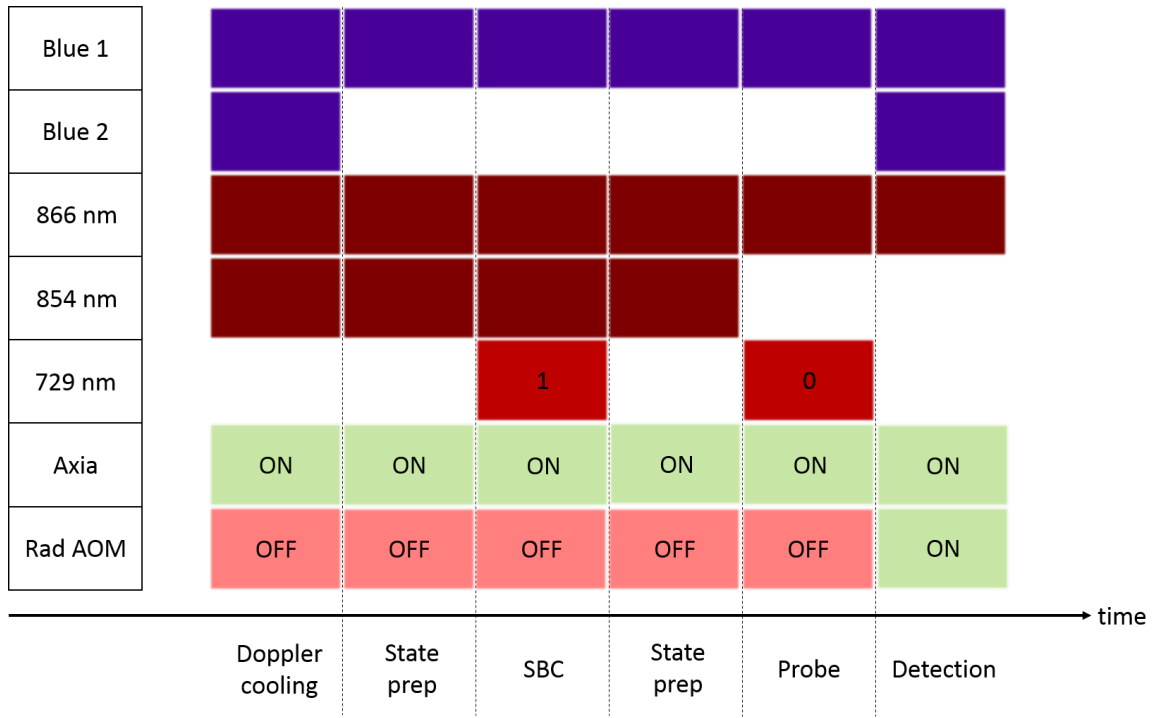


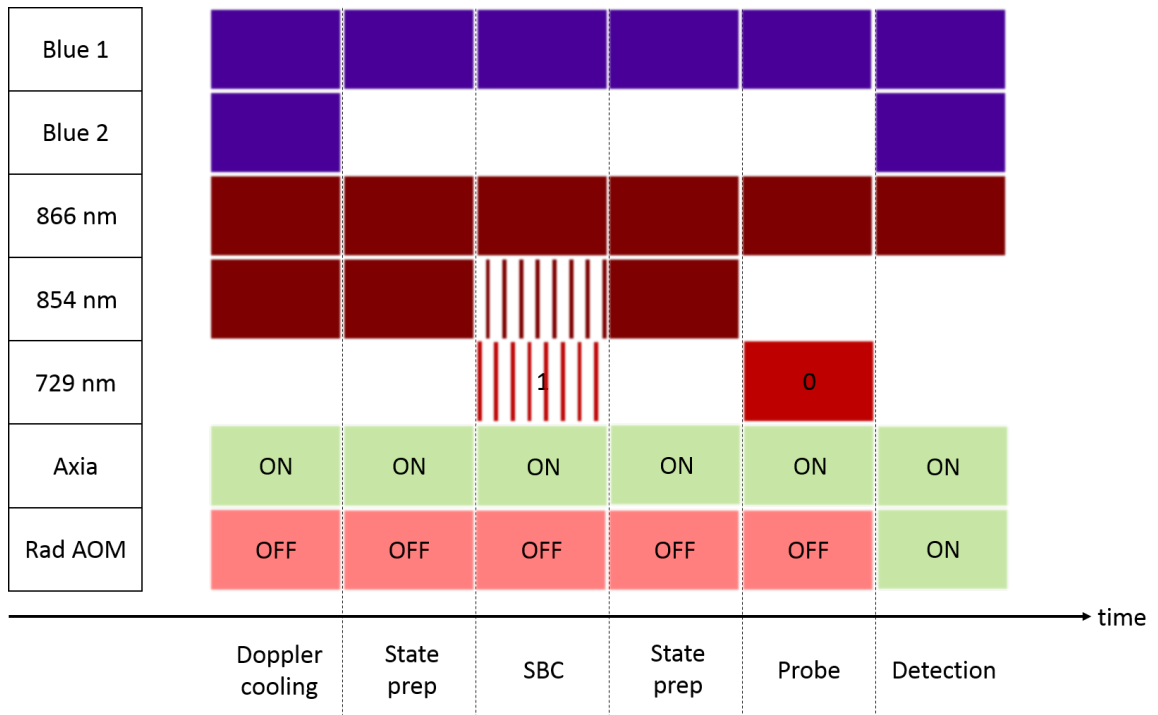
Figure VI.1: Axial motional spectrum of a single ion after 12 ms of Doppler cooling. The axial frequency is $\nu_z = 187$ kHz ($\eta = 0.224$, trapping voltage 50 V). The blue line is a fit to the data points giving an average phonon number $\bar{n} = 57(3)$ and a Rabi frequency of roughly 20 kHz.

Continuous and pulsed sideband cooling

Sideband cooling (SBC) of $^{40}\text{Ca}^+$ is done by driving the $S_{1/2} \longleftrightarrow D_{5/2}$ transition at 729 nm. The slow decay rate of $D_{5/2}$ requires the use of the 854 nm repump laser to accelerate the process. We distinguish two methods to use the repump laser called continuous sideband cooling and pulsed sideband cooling. In the former, the 729 nm and the 854 nm lasers are turned on simultaneously so whenever the ion is promoted to the $D_{5/2}$ state, it is rapidly repumped to $P_{3/2}$ and decays back to the ground state to close the cooling cycle. Both lasers are kept on for the whole duration of sideband cooling which is typically 5 to 30 ms. In pulsed sideband cooling, a short pulse of the 729 nm laser of the order of 10 to 100 μs , depending on the laser intensity, first excites the ion with a probability given by equation 89, the 729 nm is then turned off and a pulse of the 854 nm is applied. The process is repeated many times to decrease the phonon number significantly. The pulse sequences for continuous and pulsed SBC are shown in figure VI.2. Continuous SBC has the advantage of being faster than pulsed SBC where, for a given 729 nm illumination time, the extra time needed for the 854 nm pulses makes the overall cooling time longer. Besides, with pulsed SBC, care must be taken that the 729 nm pulses are short enough so that they cannot coherently drive the ion's atomic state from the ground state to the excited state and back to the ground state in which case no cooling will occur. This means that the pulse duration has to be based on the highest Rabi frequency as given by equation 113. Where the Rabi frequency is lower, the pulse may be too



(a)



(b)

Figure VI.2: Sequences for continuous (a) and pulsed (b) single stage sideband cooling followed by spectroscopy as done for the axial motion of a single ion at high trapping frequencies. During sideband cooling, profile 1 of the DDS is in use and has a constant frequency throughout the experiment. The number of individual 729 nm pulses in pulsed sideband cooling ranges from a few tens to a few hundred. On the probe pulse, profile 0 is in use; the frequency of which is of course different at each step of the spectrum.

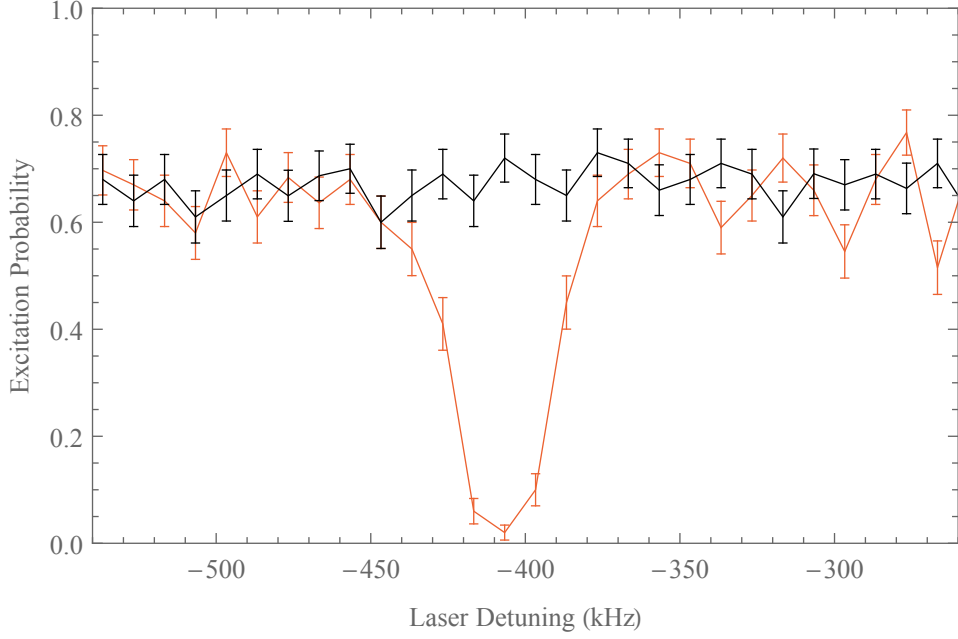


Figure VI.3: Dip spectrum (orange points) showing the probability of excitation as a function of the frequency of the 729 nm laser during the sideband cooling pulse (expressed in detuning from the carrier). The frequency of the probe is fixed at the position of the unshifted first red sideband. The black points show the height of the red sideband after Doppler cooling for comparison. The axial frequency is $\nu_z = 400$ kHz. The minimum of the dip is slightly to the left of the -400 mark indicating an AC Stark shift of about -8 kHz.

short to excite the ion with a significant probability resulting in inefficient cooling. The greater efficiency of continuous SBC and its simpler pulse sequence makes it our preferred technique in many situations. However, because of the Stark shift induced by the 854 nm laser, the position of the first red sideband (or higher order) on the Doppler spectrum cannot be used directly to set the 729 nm laser’s frequency. To find the position of the Stark shifted sideband, we perform a “dip spectrum”: the sideband cooling frequency is scanned around the position of the first red sideband as found on the Doppler spectrum and the the probe is fixed at the unshifted red sideband. The dip spectrum therefore gives the probability of excitation on the red sideband as a function of the SBC pulse frequency. An example is given in figure VI.3. If the SBC pulse frequency is resonant with the Stark shifted red sideband, cooling will occur and the height of the sideband will go down as the ion approaches the ground state. A dip in the spectrum – hence the name of the technique – indicates the position to set the laser to for efficient cooling. Note that the dip spectrum is in itself a demonstration of sideband cooling but a resolved sideband spectrum is necessary to find the final average phonon number.

In simple cases, with well resolved Doppler spectra where the sidebands are clearly identifiable, the dip spectrum method works fine but it can be impractical in more complex situations where the spectrum features many sidebands close to each other.

In contrast, pulsed sideband cooling does not require a dip spectrum to be recorded since the ion is not affected by the AC Stark shift predicted by equation 165 when the $S_{1/2} \longleftrightarrow D_{5/2}$ transition is driven. Therefore, pulsed SBC can be more convenient to use in certain situations. Unless mentioned otherwise, the results presented below were obtained with continuous sideband cooling.

Single-stage SBC

We call single-stage sideband cooling (SSSBC) a pulse sequence where the frequency and power of the 729 nm laser is constant during the SBC pulse. This is to be differentiated from multiple-stage sideband cooling (MSSBC) where there are at least two SBC pulses with different parameters (frequency, power) for the 729 nm laser. This cooling technique works well for a single ion with a high axial frequency (small Lamb-Dicke parameter) but is not suitable at lower frequencies as we will see below.

The spectrum in figure VI.4 was obtained from a sideband cooled ion with an axial frequency of 400 kHz ($\eta = 0.153$). Several features of this spectrum are noticeable: the absence of a visible red sideband and the high blue sideband are the signs of a high ground state occupation probability; the fringes on the side of the carrier show that the $S_{1/2} \longleftrightarrow D_{5/2}$ transition is coherently driven by the laser light and are consistent with equation 89. Note that on this spectrum the probe length and power are chosen to maximise the height of the blue sideband i.e. to do a π pulse. A fit to the data points, done in a similar fashion as for a Doppler spectrum, yields an average phonon number $\bar{n} = 0.015(18)$ or, under the reasonable assumption that only the motional states $|0\rangle$ and $|1\rangle$ may be occupied, a 98.5(1.8) % occupation probability of the ground state. The heating rate is rather low at this frequency, around $0.3(2) \text{ s}^{-1}$ [46].

All else being equal, when the trapping frequency is lower, the average phonon number of the ion after Doppler cooling is higher (equation 144). Besides, the coupling strength of the first red sideband goes to zero at a lower phonon number therefore the probability for the ion to be at a phonon state above this zero coupling point is significantly higher. Thus, sideband cooling on the first red sideband will cool the population under the zero coupling point normally, steadily decreasing the phonon number but for the population above the zero, the cooling will stop when this point is reached as the laser light does not couple with the internal states of the ion any more, leading to population trapping. Figure VI.5 shows a spectrum obtained in such a situation. The axial frequency is 187 kHz and the cooling time 25 ms. Notice the large asymmetry between the first blue and red sidebands and the still high second and third order sidebands. The excitation on the first blue sideband comes mainly from the part of the population in the ground state while the trapped population creates a non-zero coupling to the higher order sidebands.

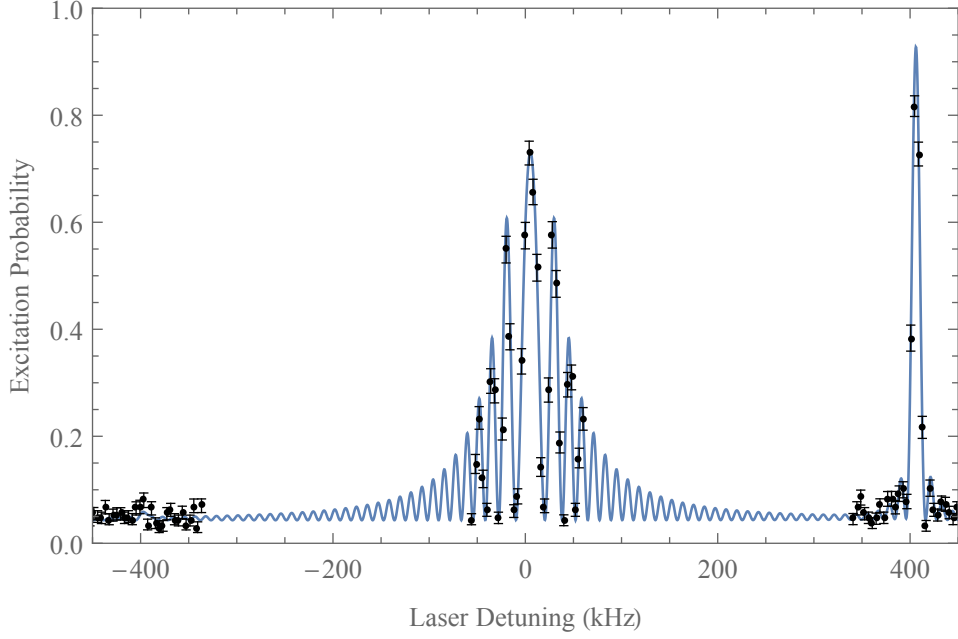


Figure VI.4: Axial spectrum after 21 ms of sideband cooling on the first red sideband. The trapping frequency is $\nu_z = 400$ kHz ($\eta = 0.153$, trapping voltage 230 V) and the axialisation amplitude 200 mV. The blue line is a fit to the data points giving an average phonon number $\bar{n} = 0.015(18)$ and a Rabi frequency of roughly 30 kHz.

From the Doppler spectrum (not shown), an average phonon number $\bar{n} = 46(8)$ is found which means that, considering a thermal distribution, between 15 % and 26 % of the population is above the first zero-coupling point of the first red sideband which is at $n = 73$ at this frequency. To fit the sideband cooled spectrum, we need to take into account the fact that a part of the population has a thermal distribution close to the ground state and another part is trapped. We assume that all the trapped population is concentrated at a single Fock state ($n = 73$). The equation 178 is thus modified to:

$$P_{e,s}(\delta, t) = (1 - x) \sum_{n=0}^{10} \frac{\bar{n}^n}{(\bar{n} + 1)^{n+1}} \times \frac{\Omega_{n,n+s}^2}{\Omega_{n,n+s}^2 + \delta^2} \sin^2 \left(\frac{\sqrt{\Omega_{n,n+s}^2 + \delta^2}}{2} t \right) + x \frac{\Omega_{73,73+s}^2}{\Omega_{73,73+s}^2 + \delta^2} \sin^2 \left(\frac{\sqrt{\Omega_{73,73+s}^2 + \delta^2}}{2} t \right) \quad (179)$$

where x is the part of the population trapped. In the first term, the sum is truncated at $n = 10$ which is enough given that $\bar{n}$ is much smaller than one. The fit yields $\bar{n} = 0.15(2)$ and $x = 0.155(6)$ which remains in the range expected from the Doppler spectrum, albeit at the lower end. It is however likely that some of the population initially above $n = 73$ still got cooled to the ground state since the coupling strength of the first red sideband at the trapping point is not strictly zero ($0.0007 \times \Omega_0$ to be precise) so a small excitation remains possible. Overall, the ground state occupation probability is quite low at about 72 % and so SSSBC is not a suitable technique to

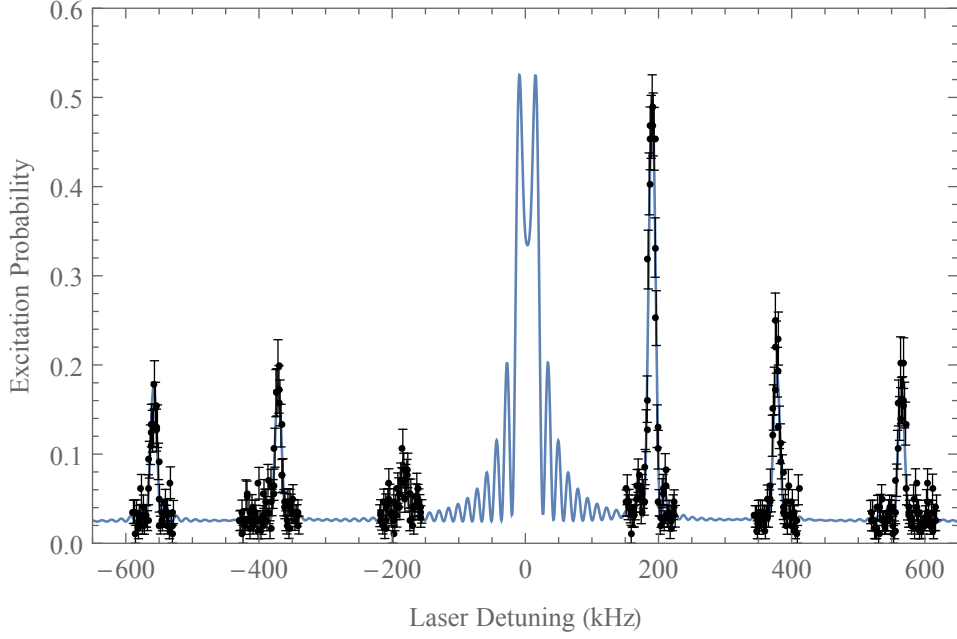


Figure VI.5: Axial spectrum after 25 ms of sideband cooling on the first red sideband. The trapping frequency is $\nu_z = 187$ kHz ($\eta = 0.224$, trapping voltage 50 V). Due to an error when acquiring the spectrum, the data points for the carrier are missing. The fit is done only on the sidebands and suggests that around 15 % of the population is trapped. The rest of the population has an average phonon number $\bar{n} = 0.15(2)$.

reliably prepare an ion in the ground state when initially well outside the Lamb-Dicke regime.

Multiple-stage SBC

We saw in section IV.4 that higher order sidebands can be addressed to avoid population trapping. The basic strategy is to apply a pulse with the 729 nm laser tuned to the second red sideband before cooling on the first red sideband. The role of the first pulse is to cool the entire population below the zero-coupling point of the first red sideband. The second pulse (on the first red sideband) can then cool the ion towards the ground state. However, population trapping can also occur when addressing higher order sidebands. For instance, in the example above where the axial frequency is 187 kHz and $\bar{n} = 46$ after Doppler cooling, there is about 8 % of the population above the first zero-coupling point of the second red sideband ($n = 132$) and 3 % above that of the third red sideband ($n = 204$) (see figure VI.6). Thus, with a simple two stage sequence targeting the second and first red sideband, these 8 % would still be trapped.

For this reason, actual sequences used at low trapping frequencies are more sophisticated and comprise a series of pulses switching between different sidebands for an optimal cooling process in terms of speed and final average phonon number. Simulation work on sideband cooling dynamics done in our group by Franziska Beck [93] and Manoj Joshi [94] gave us templates of good cooling sequences which were then

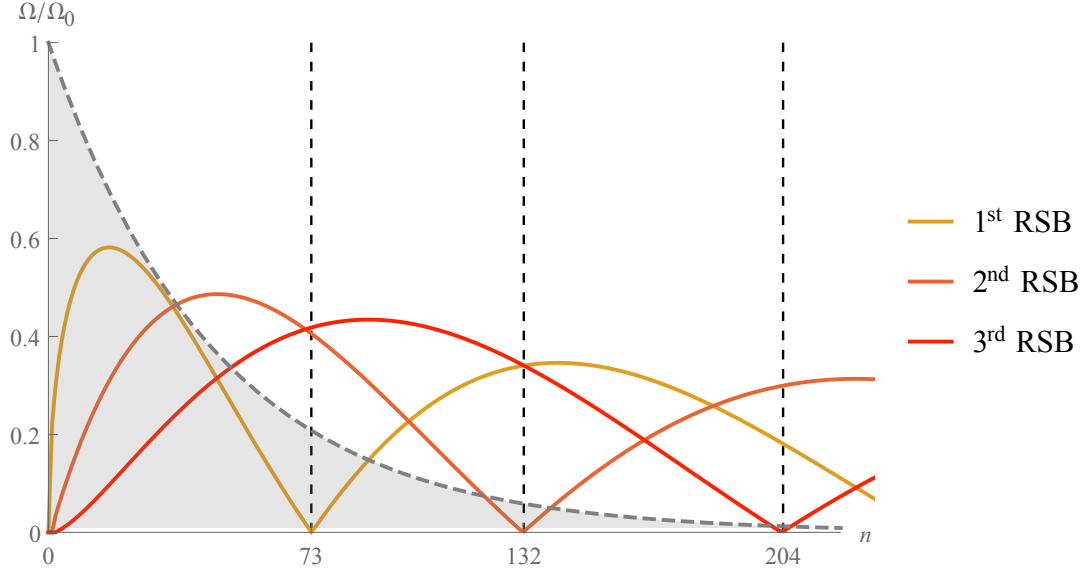


Figure VI.6: Normalised Rabi frequencies for the first, second and third red sidebands as a function of the motional state number for $\nu_z = 187$ kHz ($\eta = 0.224$). The dashed grey line represents the probability of occupation of a given motional state for a thermal distribution (normalised to 1 for $n = 0$) with an average phonon number $\bar{n} = 46$. 22 % of the population is above $n = 73$, 8 % above $n = 132$ and 3 % above $n = 204$.

Sideband	Pulse time (ms)
2 nd RSB	5
3 rd RSB	5
2 nd RSB	5
3 rd RSB	5
1 st RSB	5

Table VI.1: 729 nm pulse sequence used for multiple-stage sideband cooling at low trapping frequencies. The sidebands on the table are addressed sequentially from top to bottom. RSB: red sideband.

refined through experiments. Using the sequence shown in table VI.1 a single ion with an axial frequency of 187 kHz was cooled to the motional ground state with a high occupation probability ($\bar{n} = 0.018(11)$) as evidenced by the spectrum in figure VI.7. Unlike the spectrum of figure VI.5 the red sidebands are all very low, within the noise. The first four stages in this sequence alternate between the second and third red sidebands to avoid population trapping and increase the cooling rate as more phonons are removed per scattering event compared to cooling on the first red sideband. A final pulse on the first red sideband remains of course necessary to reach the ground state. Thanks to the eight profiles of the DDS chip in the 729 AOM driver, up to seven different sidebands (one profile is reserved for the probe pulse) can be addressed.

There is nevertheless a practical limit on the trapping frequency at which MSSBC

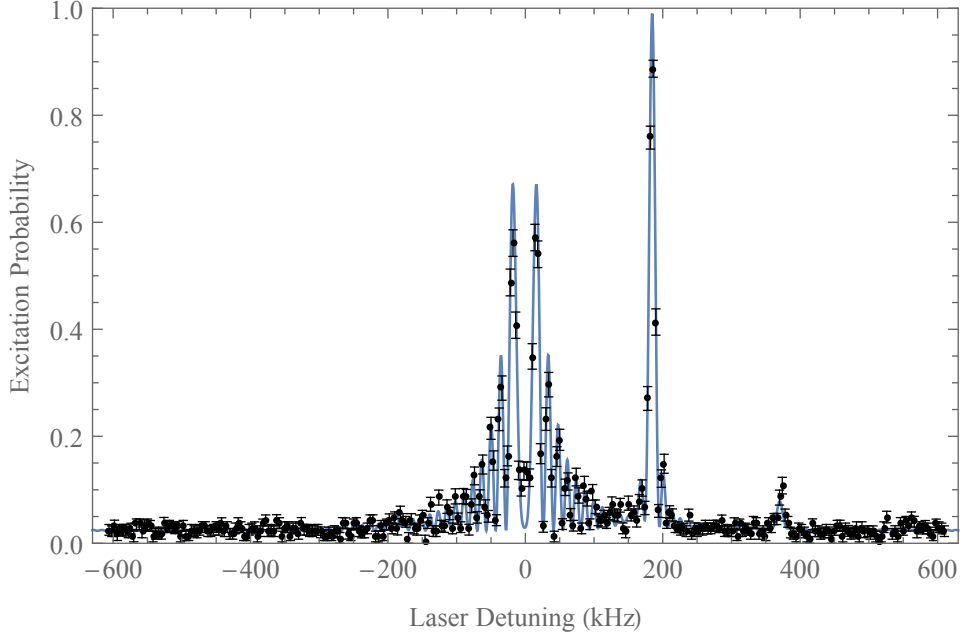


Figure VI.7: Axial spectrum after 25 ms of multiple-stage sideband cooling (sequence shown in table VI.1). The trapping frequency is $\nu_z = 187$ kHz ($\eta = 0.224$, trapping voltage 50 V). The blue line is a fit to the data points giving an average phonon number $\bar{n} = 0.018(11)$ and a Rabi frequency of roughly 25 kHz.

can be implemented. As the Doppler limit keeps getting higher and the Lamb-Dicke parameter larger at lower frequencies, the pulse sequence becomes more complicated and the cooling time very long. Figure VI.8 shows a spectrum obtained for a trapping frequency of 143 kHz, the lowest for which we managed to do MSSBC and get a relatively good ground state occupation. This one was done with pulsed sideband cooling; the pulse sequence is given in table VI.2. The spectrum has features suggesting that the ion is close to the ground state: low excitation probability on the red sidebands, high excitation on the first blue and fringes around the carrier. Nevertheless, a fit to the data points made the same way as for figure VI.5 indicates that around 7 % of the population is trapped at the zero-coupling point of the first red sideband. This is evidenced by the presence of small but non negligible second and third order red sidebands. The part of the population near the ground state has an average phonon number $\bar{n} = 0.048(25)$ which is substantially more than what was measured at $\nu_z = 187$ kHz. The cooling time is also much longer at 63.2 ms, partly because it is pulsed and not continuous SBC.

Aside from cooling an ion with a low axial frequency to the motional ground state, multiple-stage sideband cooling is indispensable for sideband cooling of the radial motion and for ion Coulomb crystals.

Laser on	Sideband	Pulse time (μs)	Repetition
729	4 th RSB	60	200
854	NA	7	
729	3 rd RSB	60	
854	NA	7	
729	2 nd RSB	60	300
854	NA	7	
729	1 st RSB	70	200
854	NA	7	

Table VI.2: Cooling sequence for multiple-stage pulsed sideband cooling used for the spectrum on figure VI.8. Total time of the sequence: 63.2 ms.

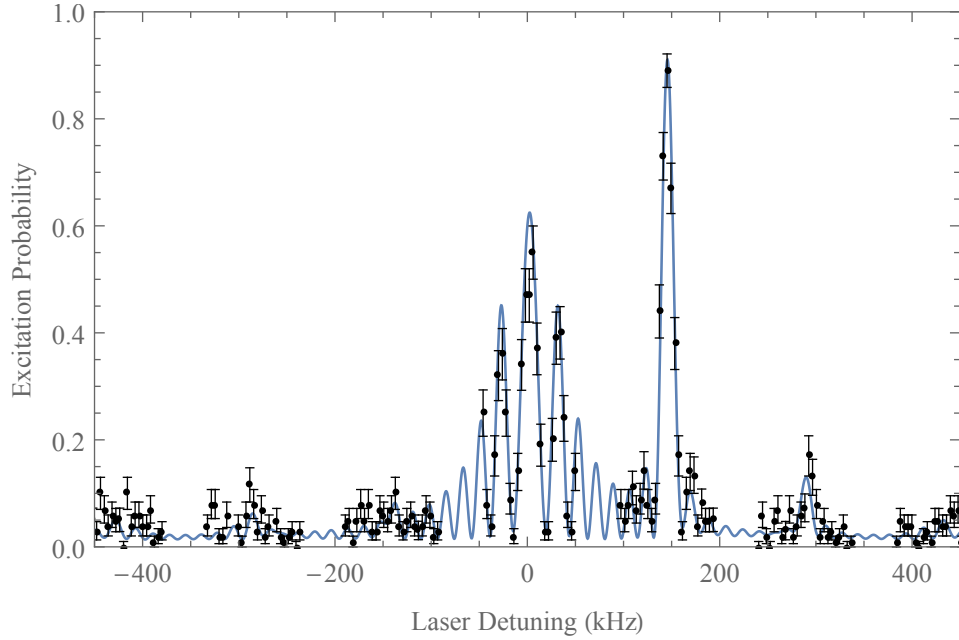


Figure VI.8: Axial spectrum after 63.2 ms of multiple-stage pulsed sideband cooling (sequence shown in table VI.2). The trapping frequency is $\nu_z = 143$ kHz ($\eta = 0.256$, trapping voltage 30 V). The fit is done with the population trapping model (equation 179) and suggests that 7(1) % of the population is trapped. The rest of the population has an average phonon number $\bar{n} = 0.048(25)$. The Rabi frequency is close to 30 kHz.

Adiabatic cooling

An alternative to multiple-stage sideband cooling to prepare an ion in the ground state at a low trapping frequency consists in doing sideband cooling at a high frequency and then lowering the trapping potential. If the relaxation is slow enough, the process is adiabatic in the quantum mechanical sense and the ion remains in the ground state. This cooling technique is simple to implement and very effective. For the trapped ion, a sufficient condition for the process to be adiabatic is that the rate of change of the trapping frequency is small compared to the trapping frequency itself. This can be translated mathematically to [95, 96]:

$$\kappa(t) = \frac{1}{\omega_z^2} \frac{d\omega_z}{dt} \ll 1. \quad (180)$$

Lacking a high voltage power supply able to ramp down the trapping voltage on command of the FPGA, this role falls to a home-built electronic circuit whose schematic is shown in figure VI.9. The circuit is inserted between the low noise high voltage supply (Delta Elektronika E0300-0.1) and the trap's electrodes. Depending on the logic level of a signal coming from the FPGA, the output of the circuit may be equal to the high voltage input or lower by an amount set by a potentiometer. When reducing the voltage, the circuit is, in essence, a passive potential divider with a switching behaviour characteristic of an RC circuit: the voltage drop follows an exponential decay. The time constant of the decay depends on the ratio between the output and input voltages which is expected from such a circuit (see figure VI.10.a). The inequality in equation 180 can be expressed in terms of the trap voltage U as:

$$\sqrt{\frac{mR^2}{4e}} \frac{1}{U^{3/2}} \frac{dU}{dt} \ll 1 \quad (181)$$

and, knowing the expression for the voltage as a function of time, we are able to verify that the condition for an adiabatic process is fulfilled as can be seen in figure VI.10.b.

Our original motivation for using adiabatic cooling was technical: we wished to observe an interference signal oscillating at the trapping frequency from a ground state cooled ion (see section VIII.2 on motional coherence) but the time resolution of the FPGA was not fine enough to do so at high trapping frequency where sideband cooling is easier. It was therefore necessary to work at lower frequencies, not easily attainable with MSSBC.

Figure VI.11 gives examples of spectra taken after adiabatic cooling. We first prepare the ion in the ground state by performing sideband cooling at $\nu_z = 420$ kHz i.e. at a trapping potential of 250 V. The potential can then be reduced by any amount to reach the desired frequency. At a final trapping frequency of $\nu_z = 152$ kHz (figure VI.11.a), close to the practical limit of MSSBC, we find an average phonon number

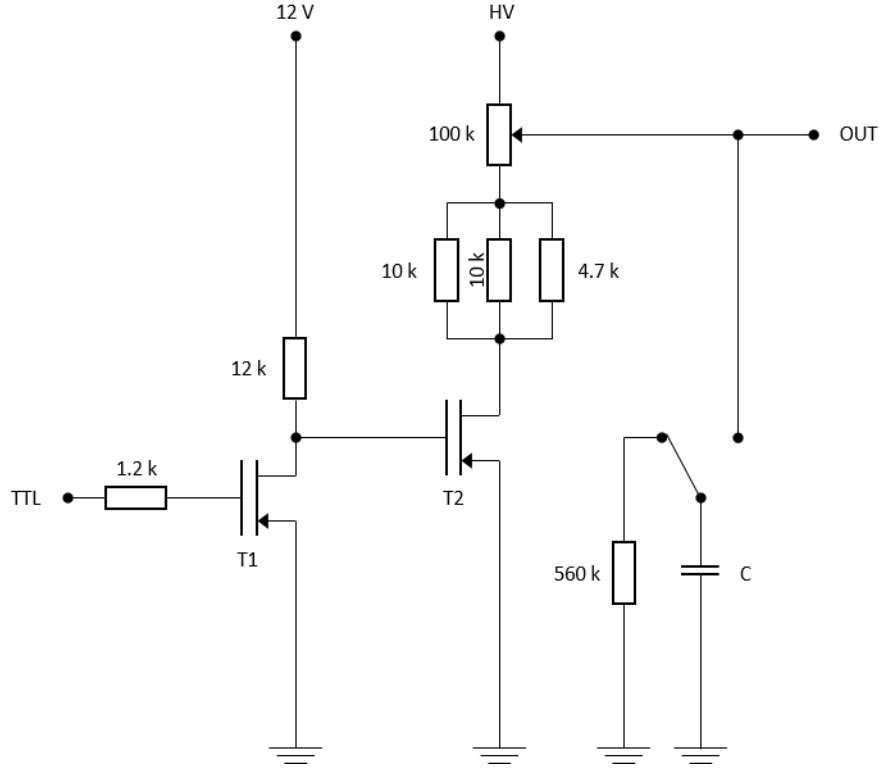


Figure VI.9: Electronic circuit used to decrease the trapping voltage. The output voltage depends on the state of the transistors T1 and T2 controlled by the digital TTL input. A logic high yields an output voltage equal to the high voltage input (HV) while a logic low activates the voltage divider producing an output lower than HV, depending on the position of the 100 k Ω potentiometer. An additional capacitor can be connected to the output of the circuit to increase its RC constant for a slower output voltage change when the TTL level is switched. The capacitor is not soldered to the electronic board and can be changed easily to vary the RC constant. If not connected to the output, the capacitor discharges in a 560 k Ω bleeder resistor.

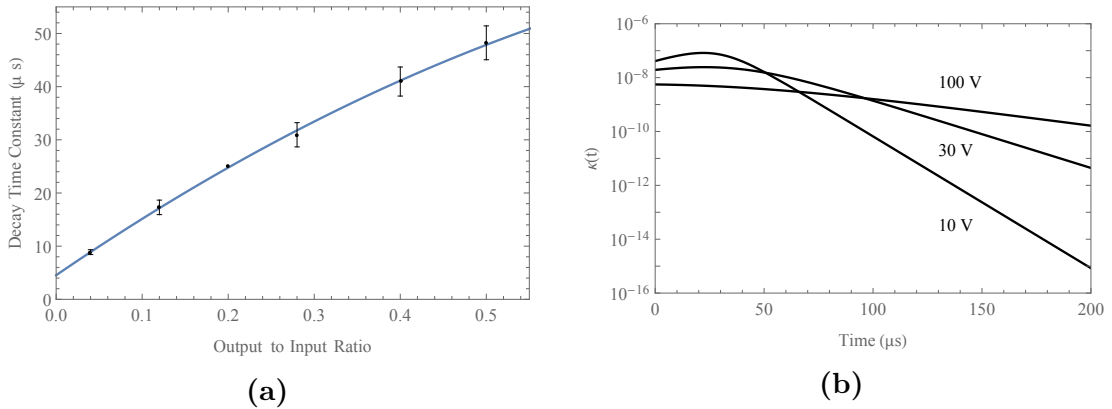
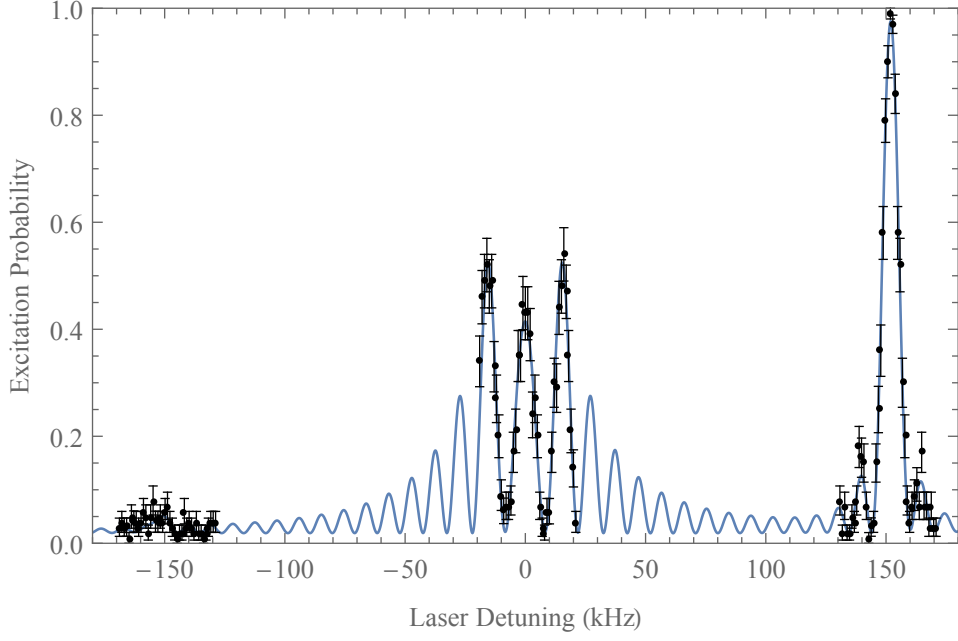


Figure VI.10: (a) Decay time constant of the voltage divider as a function of the ratio between the output and the input. Measured with the additional capacitor (see figure VI.9) disconnected and a constant input voltage of 250 V. The data is fit to a second order polynomial. (b) $\kappa(t)$ as defined by equation 180 for trap voltage relaxations from 250 V to 10, 30 and 100 V.

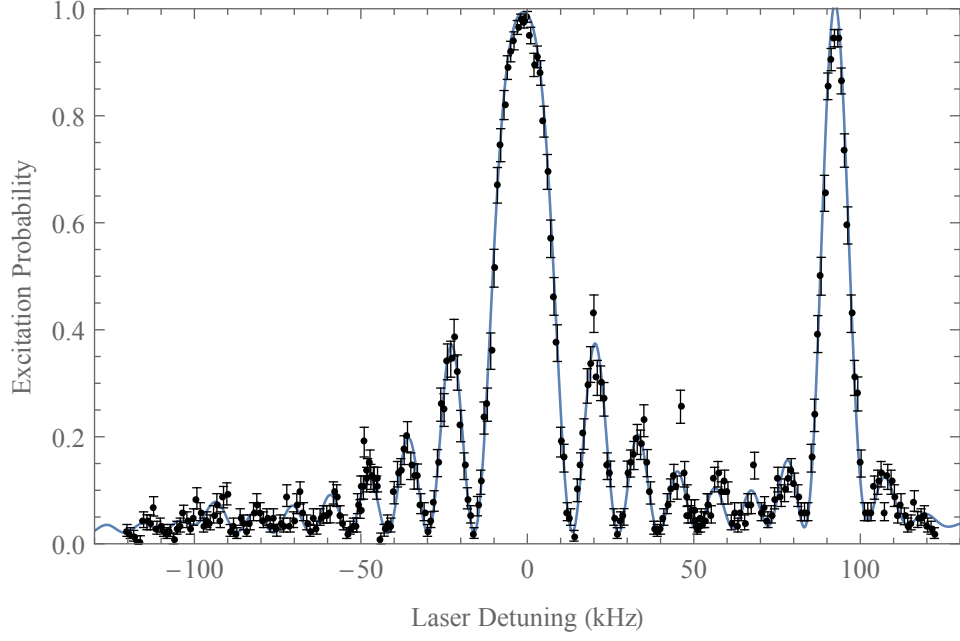


(a)

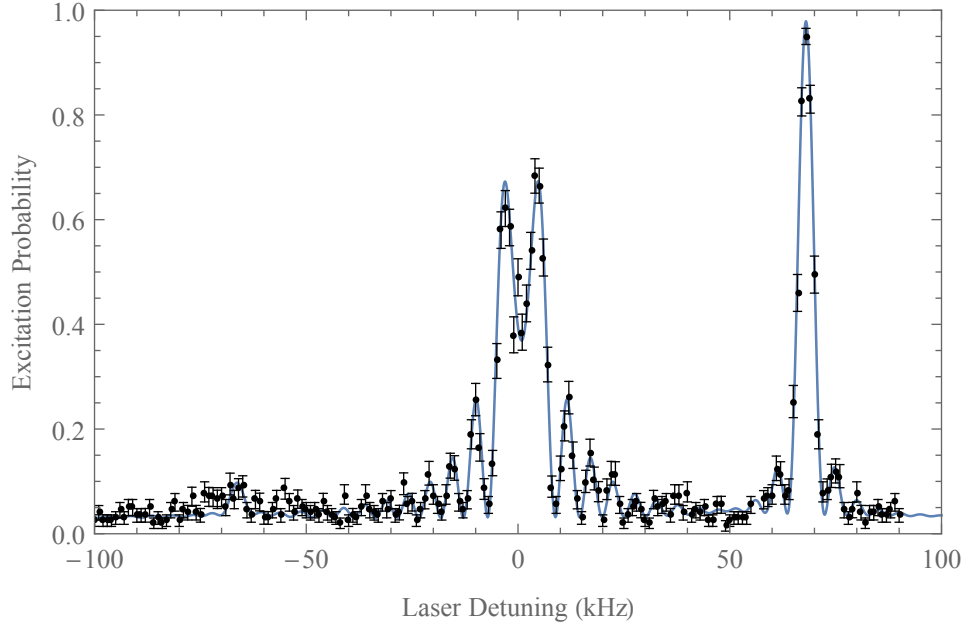
Figure VI.11

$\bar{n} = 0.028(10)$ despite cooling for only 12 ms. A good ground state occupation is maintained for even lower frequencies: $\bar{n} = 0.034(9)$ at $\nu_z = 94$ kHz and $\bar{n} = 0.07(1)$ at $\nu_z = 67$ kHz (figures VI.11.b and c). The frequency can however not be reduced to arbitrarily low values as the potential should remain large enough to confine the ion. Besides, the voltages on the compensation electrodes have to be set very carefully to avoid any displacement of the trap zero field point when reducing the trapping potential. A possible explanation for the raising average phonon number at lower frequencies could be a higher heating rate. A measurement at $\nu_z = 118$ kHz (20 V) showed a heating rate of $9.6(1) \text{ s}^{-1}$ (figure VI.12) which is substantially higher than what was measured at high voltage.

Adiabatic cooling of a single ion has been previously demonstrated in a linear RF trap [96]. This experiment also used a $^{40}\text{Ca}^+$ ion cooled to the ground state with sideband cooling and was performed at trapping frequencies close to ours. Similar results were found. The authors suggest adiabatic cooling may find applications in quantum information processing, high resolution spectroscopy and ultra cold chemistry.



(b)



(c)

Figure VI.11: Axial spectra after 12 ms of sideband cooling at a trapping frequency $\nu_z = 420$ kHz (250 V) followed by adiabatic relaxation to (a) $\nu_z = 152$ kHz, (b) $\nu_z = 94$ kHz and (c) $\nu_z = 67$ kHz. The blue lines are fits to the data points giving average phonon numbers $\bar{n} = 0.028(10)$, $\bar{n} = 0.034(9)$ and $\bar{n} = 0.07(1)$ for (a), (b) and (c) respectively.

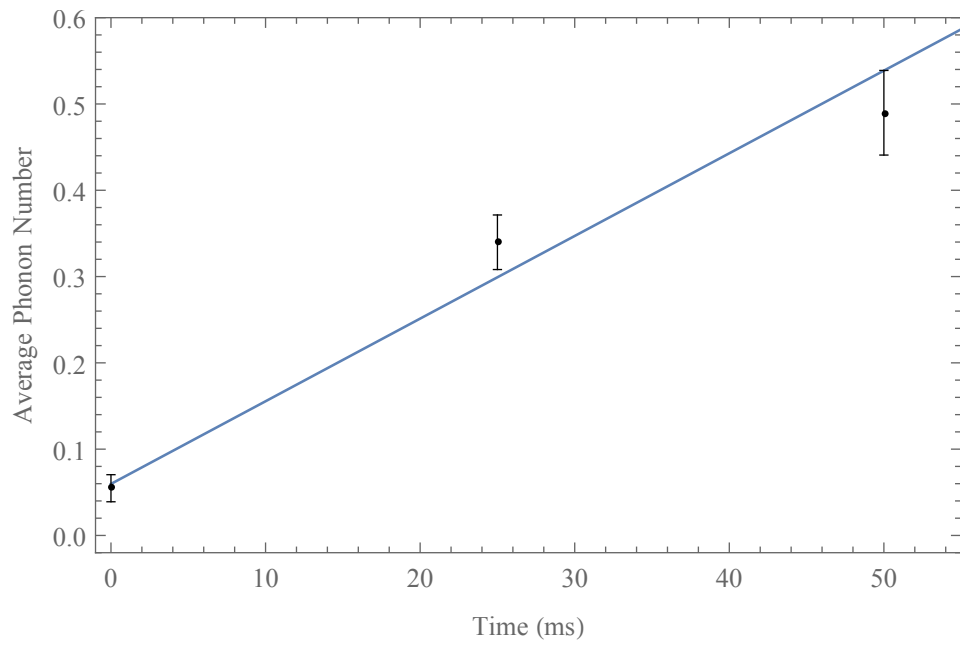


Figure VI.12: Average phonon number as a function of the wait time between the interruption of sideband cooling and the spectroscopic measurement at $\nu_z = 118$ kHz (20 V). The ion is initially cooled at 250 V ($\nu_z = 420$ kHz) before relaxing the trapping potential. A linear fit gives a heating rate of $9.6(1) \text{ s}^{-1}$.

Rabi oscillations

Cooling an ion to the ground state gives the possibility to perform various coherent manipulations. Among those, an important one is the observation of Rabi oscillations which provide useful information about the coherence of the system and the strength of the light-ion coupling. Importantly they are used to measure the duration of a π pulse, a key parameter for many experiments which will be discussed in chapter VIII.

Experimentally, Rabi oscillations can be observed by exciting the $S_{1/2} \longleftrightarrow D_{5/2}$ transition with the spectroscopy laser tuned on resonance, usually at the carrier frequency but it can also be at the first blue sideband, and measuring the probability of excitation for increasing excitation times. The method is therefore similar to resolved sideband spectroscopy except that the variable is the duration of the probe pulse rather than its frequency. Each data point is also the result of hundreds of repetitions in order to obtain meaningful probabilities. Figure VI.13 shows Rabi oscillations on the carrier transition taken after sideband cooling at $\nu_z = 420$ kHz. Notice how the amplitude of the oscillations decreases with time. This damping may have several origins such as imperfect cooling, intensity noise and frequency noise. This can be understood by looking at the expression for the probability of excitation on the carrier transition for an ion in a thermal state:

$$P_{e,s}(\Omega_0, \delta, \bar{n}, t) = \sum_{n=0}^{\infty} \frac{\bar{n}^n}{(\bar{n} + 1)^{n+1}} \times \frac{\Omega_{n,n}^2}{\Omega_{n,n}^2 + \delta^2} \sin^2 \left(\frac{\sqrt{\Omega_{n,n}^2 + \delta^2}}{2} t \right). \quad (182)$$

Clearly, if $\bar{n}$ is non zero, and it always is, the probability of excitation as a function of time will be the result of contributions from sinusoids with different frequencies. Besides in each of the hundreds of repetitions making a data point, the Rabi frequency Ω_0 and the detuning δ may be slightly different due to noise in the laser's intensity and frequency. Another possible cause of instabilities of the frequency noise is magnetic field noise. All these effects lead to a damping of the oscillations. The Rabi oscillations alone cannot distinguish between them with certainty but we can use other measurements to make an educated guess. The typical average phonon number after sideband cooling is known from spectroscopy which allows us to put a limit on the effect imperfect cooling may have on the Rabi oscillations. From the equation above, it is apparent that frequency noise will have a significant effect only if it results in a detuning comparable to the coupling strength $\Omega_{n,n}$. Independent measurements (see chapter VIII) suggest the laser linewidth is of the order of a kilohertz or below. The intensity noise is known to be below 1 % after the noise eater although it can be higher at the trap as explained in the previous chapter.

The fit to the data points in figure VI.13 is done by setting $\bar{n} = 0.05$ which corresponds to what was measured from spectroscopy. Neglecting the frequency noise and assuming that the Rabi frequency has a Gaussian distribution due to intensity

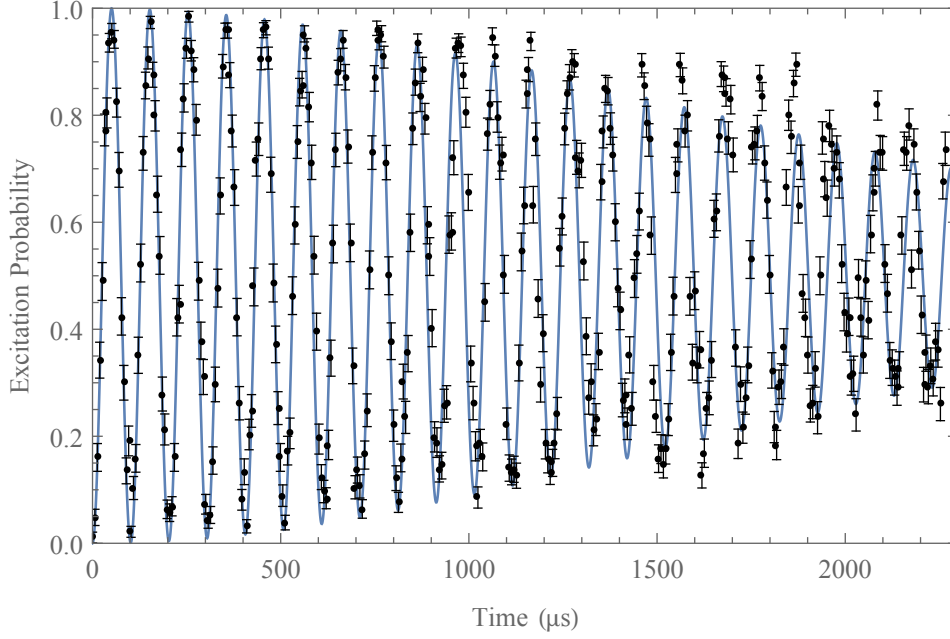


Figure VI.13: Probability of excitation as a function of the probe pulse duration showing Rabi oscillations with a frequency of 9972(4) Hz. The fit assumes a Gaussian profile for the decay of the oscillations amplitude with a characteristic time $\tau_d = 1.79(7)$ ms (see text for details).

noise, the function for the probability of excitation used to fit the data is¹

$$P_{e,s}(\Omega_0, \bar{n}, \tau_d, t) = \sum_{n=0}^{10} \frac{\bar{n}^n}{(\bar{n} + 1)^{n+1}} \times \frac{1}{2} \left(1 - \cos(\Omega_{n,n} t) e^{-t^2/(2\tau_d^2)} \right). \quad (183)$$

This yields a characteristic decay time $\tau_d = 1.79(7)$ ms and a mean Rabi frequency of 9972(4) Hz. The standard deviation of the Rabi frequency, in hertz, is related to the decay time by $\sigma = 1/(2\pi\tau_d)$ i.e. 89 Hz which represents about 0.9 % of the mean value. If we define the coherence time T_c as the point where the visibility of the oscillations is equal to $1/e$, we have $T_c = \sqrt{2}\tau_d$. With the previous results, it gives $T_c = 2.53(10)$ ms.

VI.2 Radial Cooling

Doppler cooling

Even when cooling the axial motion of the ion, a blue laser beam propagating perpendicularly to the trap axis cools the radial motion as explained in the previous section. However, whereas our concern was simply to maintain a tight radial con-

¹This equation is found by integrating the product of the cosine with the probability density function of the normal distribution (a Gaussian) over $\mathbb{R}$. The result of such a definite integral is known and can be found for example in ref. [97] (equation 7.4.6): $\int_{-\infty}^{\infty} \exp(-ax^2) \cos(2xt) dx = \sqrt{\pi/a} \exp(-t^2/a)$.

finement, we now seek to reach a regime from which it is possible to initiate sideband cooling. We thus need to make the average phonon numbers of both the modified cyclotron ($\bar{n}_+$) and magnetron ($\bar{n}_-$) modes as low as possible.

Recalling equation 144, the first step is to increase the scattering rate from the radial beam and reduce that from the axial one in order to lower the Doppler limit of the radial modes. This is of course at the expense of the axial motion. Experimentally, this is done by reducing the intensity of the axial blue beam in the following manner: we swap the axial and radial beam paths on the optical table which means that the beam going through the AOM shown in figure V.2 will now propagate in the axial direction instead. Rather than simply using a neutral density filter, we do this because it allows us to modulate the axial beam intensity with the AOM which is desirable for two reasons. First, by adjusting the power of the RF signal driving the AOM, we can fine tune the intensity of the beam to find the best compromise between keeping a good confinement in the axial direction and lowering the cooling limit of the radial modes. Second by turning off the AOM during the detection stages of spectroscopic measurements, we can increase the scattering from the axial beam to improve the signal to noise ratio. Remember that background light detected by the PMTs is mostly due to the radial beam.

The Doppler limit of the radial modes increases with the trapping voltage therefore it would seem preferable to work at low voltage. However, keeping in mind that our ultimate goal is to perform sideband cooling for which we need to resolve the sidebands and address them individually, the trapping voltage should be high enough for the magnetron sidebands to be well separated.

Radial Doppler cooling is also very sensitive to the focus and the position of the radial beam as this determines the parameter y_0 on which the cooling limits of the radial modes depend. Because $\bar{n}_+$ decreases with larger y_0 while $\bar{n}_-$ increases, a trade-off between cyclotron and magnetron cooling has to be made when choosing the geometrical parameters of the radial beam. The diameter of the beam at the trap centre is around 100 μm which is quite large and therefore more favourable for the modified cyclotron mode. Having a tighter focus would require using a lens with a shorter focal length than the one currently used and installing it closer to the trap. This is not possible due to limited access to the trap vicinity (it is inside the bore of a magnet). Intensity gradient cooling alone does not succeed in cooling simultaneously the two radial modes well enough. Although the cyclotron mode can be efficiently cooled with this method, the average phonon number of the magnetron mode remains too high (several hundred) to be a reasonable starting point for sideband cooling. We find that using a combination of intensity gradient cooling and axialisation gives the best results. The magnetron mode then benefits from the efficient cooling of the cyclotron such that its phonon number is significantly decreased. Two spectra centred on the carrier transition of the modified cyclotron

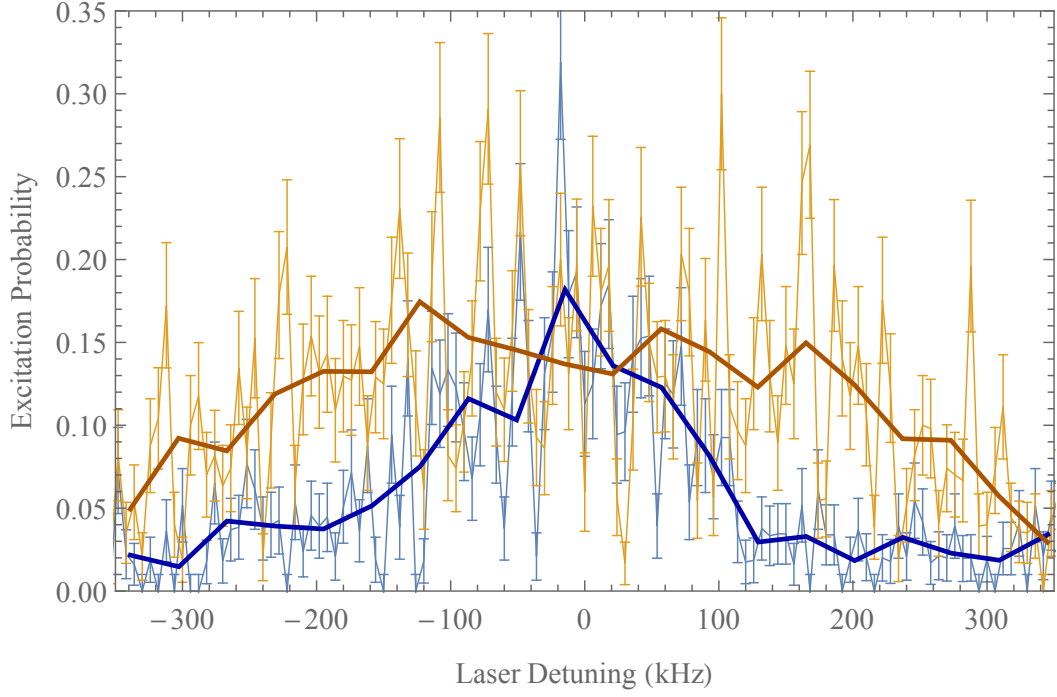
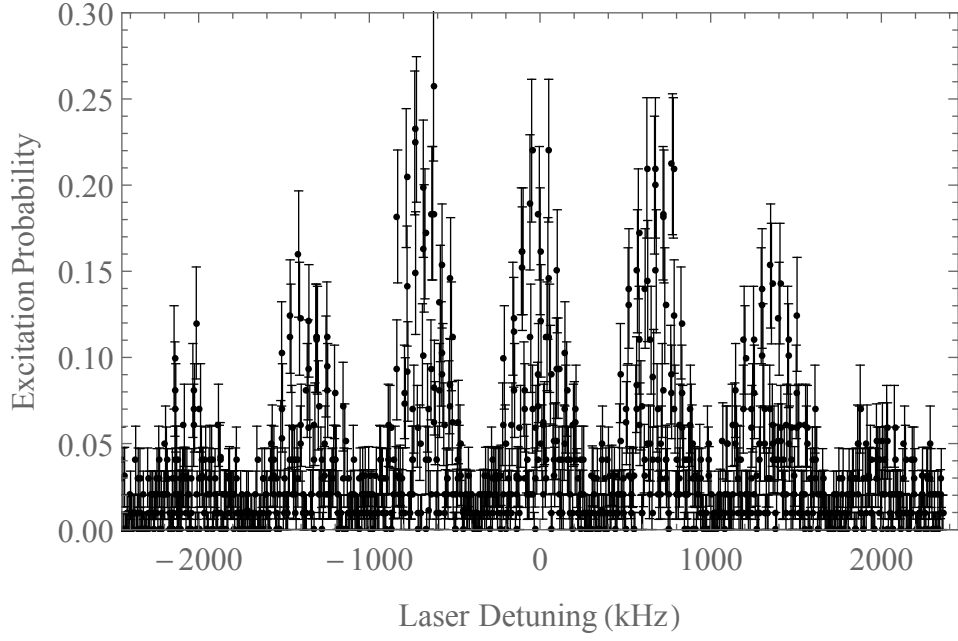
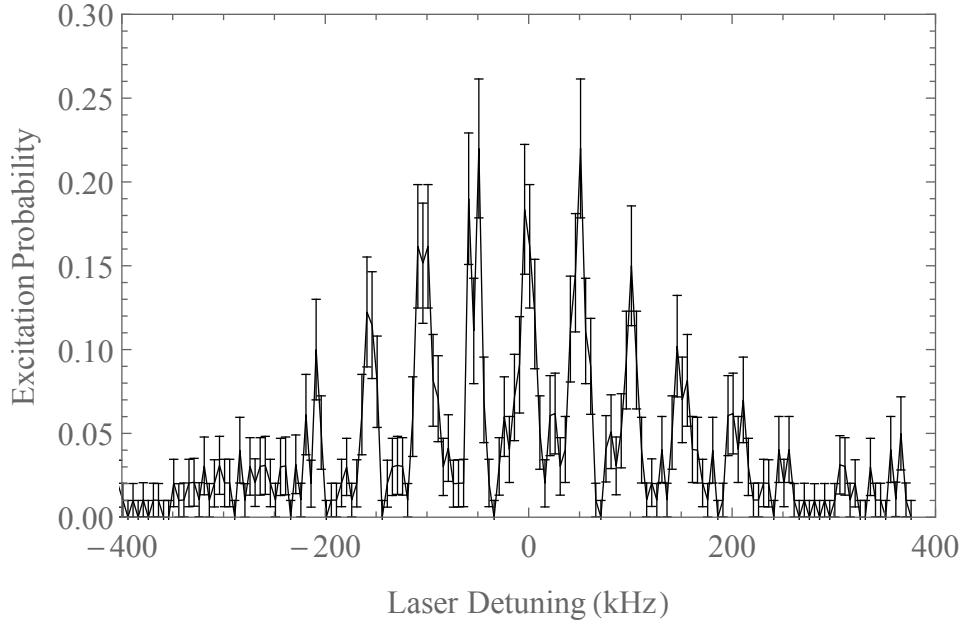


Figure VI.14: Radial spectrum of a Doppler cooled ion with axialisation (in blue) and without axialisation (in orange) around the carrier transition of the modified cyclotron. The overall width of the sideband is smaller with axialisation, indicating a lower phonon number for the magnetron mode. As a visual aid, the thicker lines represent running averages over six consecutive points. The modified cyclotron frequency is $\nu_+ = 693.5$ kHz and the magnetron $\nu_- = 14.8$ kHz.

can be seen in figure VI.14. The peaks spaced by ~ 15 kHz are the magnetron sidebands. On the spectrum taken with axialisation, fewer of them are visible and the overall width of the band is smaller, indicating a lower average phonon number for the magnetron mode. These spectra were taken at a trapping voltage of 30 V where the magnetron frequency is low so the individual sidebands are poorly resolved. Figure VI.15 shows a spectrum of a Doppler cooled ion at 100 V with 1 V of axialisation where the magnetron frequency is 52 kHz and the modified cyclotron frequency 677.4 kHz. The magnetron sidebands are well resolved and therefore can be precisely addressed with the 729 nm laser for sideband cooling.



(a)



(b)

Figure VI.15: (a) Radial spectrum of a Doppler cooled ion with $\nu_+ = 677.4$ kHz and $\nu_- = 52$ kHz (trapping voltage of 100 V) and 1 V of axialisation. (b) Expanded view of the carrier of the modified cyclotron motion showing the resolved magnetron sidebands.

Radial SBC

Sideband cooling of the radial motion requires a multiple-stage approach for several reasons. First, Doppler cooling leaves the ion outside the Lamb-Dicke regime (at least for the magnetron mode) hence it is necessary to address sidebands of different orders to avoid population trapping. Second, although there are transitions that change both the modified cyclotron and magnetron phonon numbers simultaneously, their coupling strength is weaker in the vicinity of the ground state than those that affect only one of the modes. We find that it is more efficient to address the latter but it means the two modes are cooled separately. In principle, axialisation could be used for radial sideband cooling: the laser would be tuned to, for example, a cyclotron sideband and the magnetron mode would be cooled by energy transfer with the cyclotron mode. This was tried but did not give satisfactory results. Therefore axialisation is kept off during sideband cooling. Last, because of the high heating rate of the radial motion (see below), cyclotron and magnetron sidebands should be targeted alternately to avoid that one mode heats up too much when the other is being cooled. As a consequence of these requirements, the radial sideband cooling sequences are quite long and complicated. The pulse sequence used to obtain the spectra of figure VI.16 is given in table VI.3. For the magnetron mode, the *blue* sidebands (higher frequency) are addressed as they are the ones corresponding to a *decrease* in the magnetron phonon number. Because of its proximity to the carrier transition, the laser intensity is reduced when addressing the first magnetron sideband to limit off-resonant excitation. A fit of the spectrum in figure VI.16.a gives average phonon numbers $\bar{n}_+ = 0.17(4)$ for the modified cyclotron and $\bar{n}_- = 0.8(1)$ for the magnetron mode. Note how the *red* sideband of the magnetron mode is higher than the blue one in contrast with the cyclotron sidebands.

By inserting a delay where all lasers are off in the experimental sequence between the sideband cooling and spectroscopy stages, we can measure the heating of the ion's radial motion due to background noise. Three interleaved spectra were taken: one without delay (figure VI.16.a), one with a 1.5 ms delay shown in figure VI.16.b and one with 3 ms of delay (not shown). Comparing the two spectra, the effect of heating is apparent from the reduction of the asymmetry of the blue and red sidebands for both modes. Fitting the data points give the average phonon numbers, plotted as a function of the delay time in figure VI.17. Assuming the increase in the phonon number is linear in time, the heating rates for the modified cyclotron and magnetron modes are respectively $0.5(1) \text{ ms}^{-1}$ and $1.2(3) \text{ ms}^{-1}$. These are considerably larger than what was measured for the axial motion and probably explain in part why higher ground state occupation probabilities could not be achieved. It is unclear why the heating rate is so high for the radial mode.

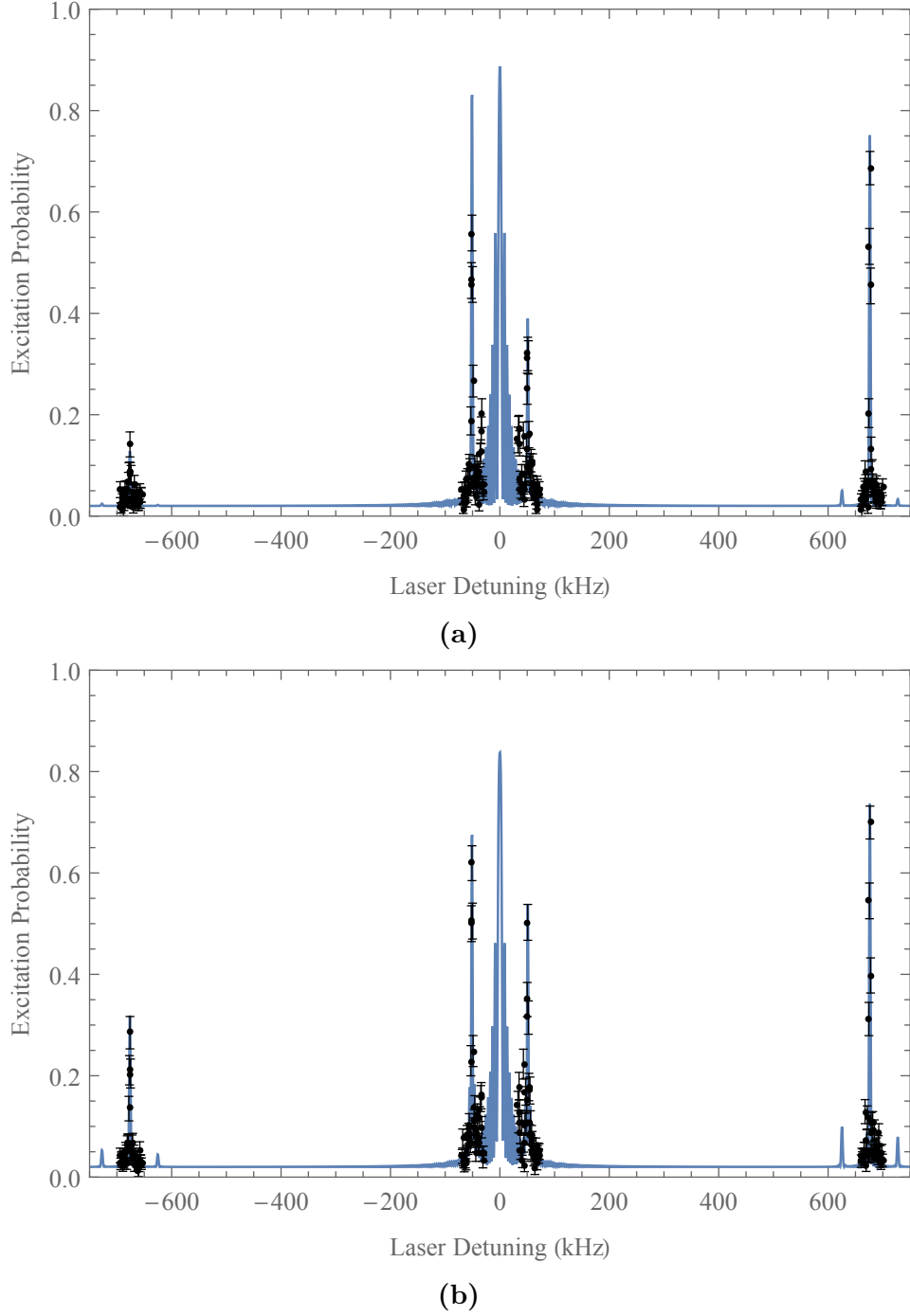


Figure VI.16: (a) Spectrum of the radial motion after 69 ms of multiple-stage sideband cooling. The spectrum is windowed around the first set of sidebands for both modes. The modified cyclotron frequency is 677 kHz and the magnetron 52 kHz. A fit to the data points, done in a similar way as for axial spectroscopy, indicates average phonon numbers $\bar{n}_c = 0.17(4)$ for the modified cyclotron and $\bar{n}_m = 0.8(1)$ for the magnetron mode. The fit seems to overestimate the asymmetry between the magnetron sidebands so the phonon number for the magnetron may actually be a bit higher. (b) Similar to (a) but with a 1.5 ms delay between the end of sideband cooling and the spectroscopic measurement. $\bar{n}_+ = 0.7(1)$, $\bar{n}_- = 3.6(5)$

Sideband	Pulse time (ms)	RF power (%)
2 nd Red Cyc	5	100
1 st Red Cyc	10	100
3 rd Blue Mag	5	100
2 nd Blue Mag	5	100
1 st Blue Mag	5	50
2 nd Red Cyc	5	100
1 st Red Cyc	5	100
3 rd Blue Mag	5	100
2 nd Blue Mag	5	100
1 st Blue Mag	5	50
1 st Red Cyc	10	100
1 st Blue Mag	0.5	50
1 st Red Cyc	0.5	100

Table VI.3: Pulse sequence used for sideband cooling of the radial motion. The total cooling time is 69 ms. “Cyc” is for modified cyclotron and “Mag” for magnetron.

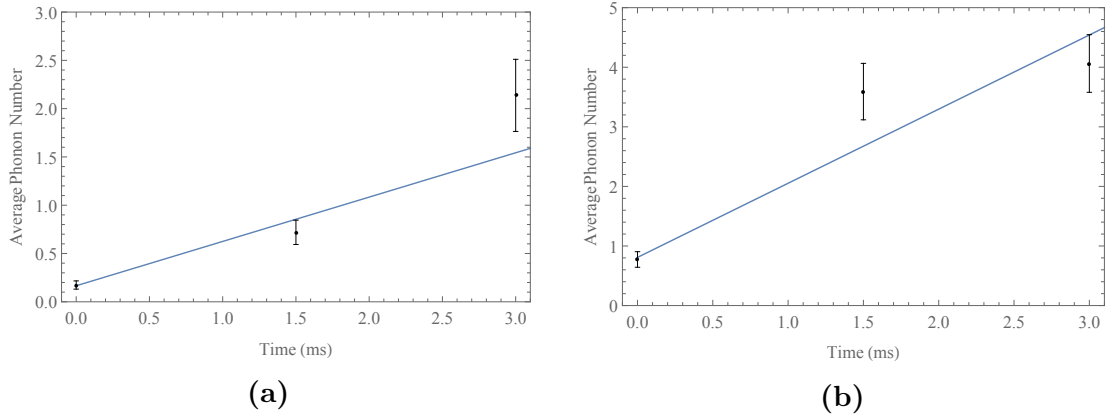


Figure VI.17: Average phonon number as a function of the wait time between the interruption of the sideband cooling and the spectroscopic measurement for the modified cyclotron (a) and magnetron mode (b). The fits give heating rates of $0.5(1) \text{ ms}^{-1}$ and $1.2(3) \text{ ms}^{-1}$ respectively for the modified cyclotron and the magnetron.

VII

Sideband Cooling of Ion Coulomb Crystals

Laser cooling of the collective motion of multiple ions to the ground state was first achieved in 1998 at NIST [98]. In this experiment, two ${}^9\text{Be}^+$ ions held in a Paul trap were cooled with Raman sideband cooling. Several other realisations of SBC of ion chains (with two or more ions) have been documented since [99, 100] but these experiments were all performed in Paul traps with one-dimensional ion Coulomb crystals. Recently, we reported sideband cooling of the axial modes of ICCs in our Penning trap, including of a two-dimensional planar crystal made of three ions [54]. The general principle of sideband cooling of ICCs is the same as for a single ion except that there are more motional modes to address so that multiple-stage sideband cooling is generally required. A fundamental difference however is that the ions in Coulomb crystals can take different configurations depending on the trapping parameters. Performing SBC on an ICC requires the ions to be in a stable configuration. For instance, with two ions, no flipping between the chain and planar configurations should occur. This puts constraints on laser cooling experiments with ICCs. In addition to the trapping voltage, the blue lasers' frequencies and intensities, the position and focus of the radial Doppler cooling beam as well as the strength of the axialisation drive all play a role in determining the crystal's configuration. In this chapter, we present results of sideband cooling of the axial modes of a two-ion chain as well as of planar crystals of different sizes.

VII.1 Sideband Cooling of a Two-Ion Chain

Chains of trapped ions have been and still are commonly used in quantum information processing (QIP) experiments. Famously, ions confined in a linear Paul trap were proposed as a medium to perform quantum gates by J.I. Cirac and P. Zoller in their landmark 1995 article [16]. This scheme uses the common motional modes of the ions to convey information between them. Early experimental realisations of

two-qubit gates were carried out on two-ion chains [27, 26]. More recently, quantum algorithms such as the Shor and Deutsch-Jozsa algorithms were implemented on larger chains of ions.[28, 29]. These one-dimensional Coulomb crystals have also been utilised to conduct quantum simulations, for instance to study frustrated Ising spins [101] or to simulate friction forces at the atomic level [4, 102].

The favoured type of trap for these experiments is the linear Paul trap where the chain configuration forms naturally along a weak trapping axis. Ion chains can also be found in Penning traps but, as discussed in section II.3, they are only stable at low trapping (axial) frequencies which greatly complicates sideband cooling.

Here we report on experiments demonstrating laser cooling of a two-ion chain in a Penning trap. The results shown in this thesis were also presented in a recent publication [54]. As for the single ion case, an initial phase of Doppler cooling is of course required before sideband cooling of the crystal can be initiated. The ions are visualised on the EMCCD camera and the experimental parameters adjusted to ensure that the crystal is stable in the chain configuration. The trapping voltage is set to 38 V which corresponds to a centre of mass frequency $\nu_z = 162$ kHz and a breathing frequency of $\nu_b = \sqrt{3}\nu_z = 281$ kHz. The axialisation amplitude is 1 V which is larger than what is usually used for a single ion and it helps to keep the ions in a chain. Figure VII.1 shows the spectrum of the two-ion chain after 20 ms of Doppler cooling which features many sidebands, barely resolved. The large number of sidebands observable is consistent with the rather large values of the Lamb-Dicke parameters. For the two-ion chain, we define the Lamb-Dicke parameters

$$\eta_c = \frac{\eta}{\sqrt{2}} \quad (184)$$

for the COM mode and

$$\eta_b = \frac{\eta}{\sqrt{2\sqrt{3}}} \quad (185)$$

for the breathing mode. η is the single ion axial Lamb-Dicke parameter as previously defined. For the breathing mode, the Lamb-Dicke parameter depends on the ion as per equation 126. Thus, for one of the ions we define $\eta_b^1 = \eta_b$ and for the other $\eta_b^2 = -\eta_b$. At $\nu_z = 162$ kHz, the Lamb-Dicke parameter for the COM mode is $\eta_c = 0.17$ and $\eta_b = 0.13$ for the breathing mode. Sideband cooling of the ion chain has to face the double difficulty of addressing two modes and, being outside the Lamb-Dicke regime, avoiding population trapping. In this regard, the situation is quite similar to sideband cooling of the radial motion of a single ion. We therefore employ a similar strategy, addressing red sidebands of different orders for both modes. There is not a unique valid sequence to perform sideband cooling of an ion chain; an example is given in table VII.1. It is essentially an extension to two ions of the type of cooling sequence which was used for a single ion at low frequency. The

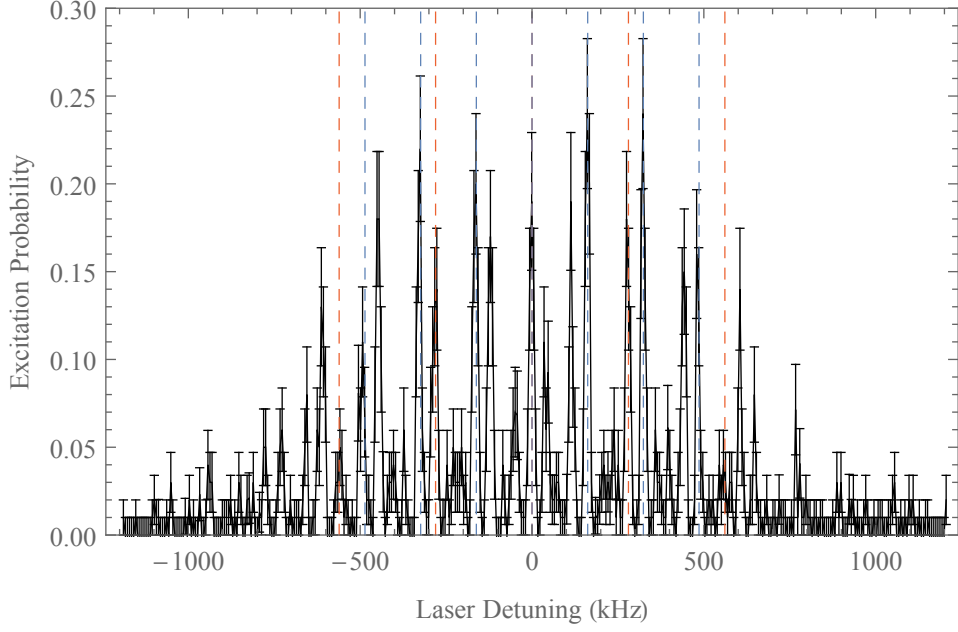


Figure VII.1: Spectrum of a two ion chain after 20 ms of Doppler cooling. The blue dashed lines mark the positions of the COM sidebands (up to the third order) spaced by $\nu_z = 162$ kHz; the orange ones, those of the breathing mode (up to the second order) spaced by $\nu_b = 281$ kHz. The other peaks visible on the spectrum are intermodulation sidebands corresponding to transitions which change both the COM and breathing phonon numbers.

presence of an intermodulation sideband which changes the phonon state of both modes is to avoid possible population trapping as explained below.

For the two-ion chain, it is possible to study the interaction of the 729 nm laser with the the electronic and motional states of the ions using a similar treatment as done in chapter III for the single ion. The unperturbed Hamiltonian is:

$$H'_0 = H_0^1 + H_0^2 + H_z$$

$$H'_0 = -\frac{\hbar\omega_0}{2}(\sigma_z^1 + \sigma_z^2) + \hbar\omega_z\left(a^\dagger a + \frac{1}{2}\right) + \hbar\omega_b\left(b^\dagger b + \frac{1}{2}\right). \quad (186)$$

The superscripts 1 and 2 on the operators distinguish the two ions. The ladder operators a and $a^\dagger$ correspond to the COM mode while b and $b^\dagger$ correspond to the breathing mode. The interaction Hamiltonian in the Schrödinger picture is:

$$H_i = \hbar\Omega_0\left(\cos(\eta_c(a + a^\dagger) + \eta_b^1(b + b^\dagger) - \omega t)\sigma_x^1 + \cos(\eta_c(a + a^\dagger) + \eta_b^2(b + b^\dagger) - \omega t)\sigma_x^2\right) \quad (187)$$

and is given in the interaction picture by:

$$H_I = e^{iH'_0 t/\hbar} H_i e^{-iH'_0 t/\hbar} \quad (188)$$

Sideband	Pulse time (μs)	Sequence repeat
2 nd COM	500	15
2 nd B	500	
3 rd COM	300	
1 st B	200	
2 nd COM 1 st B	500	
1 st B	200	2
2 nd COM	500	
1 st COM	500	
2 nd COM 1 st B	500	
2 nd COM 1 st B	500	1
1 st COM	1000	
1 st B	500	

Table VII.1: Pulse sequence used for sideband cooling of two ions. It is implied that all the sidebands are red sidebands. COM stands for centre of mass and B for breathing.

Because σ_z^1 , σ_z^2 and H_z commute, the Hamiltonian in the interaction picture can be written as:

$$H_I = \hbar\Omega_0 \sum_{j=1}^2 e^{iH_0^j t/\hbar} \sigma_x^j e^{-iH_0^j t/\hbar} e^{iH_z t/\hbar} \cos(\eta_c(a + a^\dagger) + \eta_b^j(b + b^\dagger) - \omega t) e^{-iH_z t/\hbar} \quad (189)$$

and using equation 80:

$$H_I = \hbar\Omega_0 \sum_{j=1}^2 \left(e^{i\omega_0 t} |e_j\rangle \langle g_j| + e^{-i\omega_0 t} |g_j\rangle \langle e_j| \right) e^{iH_z t/\hbar} \cos(\eta_c(a + a^\dagger) + \eta_b^j(b + b^\dagger) - \omega t) e^{-iH_z t/\hbar}. \quad (190)$$

After applying the rotating wave approximation we obtain:

$$H_I = \frac{\hbar\Omega_0}{2} \sum_{j=1}^2 |e_j\rangle \langle g_j| e^{iH_z t/\hbar} e^{i(\eta_c(a+a^\dagger) + \eta_b^j(b+b^\dagger) - \delta t)} e^{-iH_z t/\hbar} + h.c. \quad (191)$$

Let

$$\begin{aligned} \tilde{a} &= a e^{-i\omega_z t} & , & & \tilde{a}^\dagger &= a^\dagger e^{i\omega_z t} \\ \tilde{b} &= b e^{-i\omega_b t} & , & & \tilde{b}^\dagger &= b^\dagger e^{i\omega_b t} \end{aligned} \quad (192)$$

With these transformed ladder operators, the Hamiltonian becomes:

$$H_I = \frac{\hbar\Omega_0}{2} \sum_{j=1}^2 |e_j\rangle \langle g_j| e^{i(\eta_c(\tilde{a}+\tilde{a}^\dagger)+\eta_b^j(\tilde{b}+\tilde{b}^\dagger)-\delta t)} + h.c. \quad (193)$$

and with the Zassenhaus formula:

$$H_I = \frac{\hbar\Omega_0}{2} \sum_{j=1}^2 |e_j\rangle \langle g_j| e^{-i\delta t} e^{-\eta_c^2/2} e^{-(\eta_b)^2/2} e^{i\eta_c\tilde{a}^\dagger} e^{i\eta_c\tilde{a}} e^{i\eta_b^j\tilde{b}^\dagger} e^{i\eta_b^j\tilde{b}} + h.c. \quad (194)$$

Let us consider the absorption of a photon by one of the ions (ion 1) i.e. the transition $|g_1, g_2, n_c, n_b\rangle \rightarrow |e_1, g_2, m_c, m_b\rangle$ where n_c and m_c represent COM phonon numbers while n_b and m_b are phonon numbers of the breathing mode. Calculating the matrix element of the Hamiltonian yields:

$$\begin{aligned} \langle e_1, g_2, m_c, m_b | H_I | g_1, g_2, n_c, n_b \rangle &= \frac{\hbar\Omega_0}{2} e^{-i\delta t} e^{-\eta_c^2/2} e^{-(\eta_b)^2/2} \\ &\times \sum_{k_c=0}^{n_c} \sum_{l_c=0}^{m_c} \sum_{k_b=0}^{n_b} \sum_{l_b=0}^{m_b} \frac{(i\eta_c)^{k_c+l_c} (i\eta_b)^{k_b+l_b}}{k_c! l_c! k_b! l_b!} \\ &\times \sqrt{\frac{n_c! m_c! n_b! m_b!}{(n_c - k_c)! (m_c - l_c)! (n_b - k_b)! (m_b - l_b)!}} e^{i(\omega_z(l_c - k_c) + \omega_b(l_b - k_b))t} \\ &\times \langle m_c - l_c, m_b - l_b | n_c - k_c, n_b - k_b \rangle. \end{aligned} \quad (195)$$

Because of orthogonality, the terms of the above equation are non zero if $m_c - n_c = l_c - k_c = s_c$ and $m_b - n_b = l_b - k_b = s_b$. Therefore:

$$\begin{aligned} \langle e_1, g_2, m_c, m_b | H_I | g_1, g_2, n_c, n_b \rangle &= \frac{\hbar\Omega_0}{2} e^{i(s_c\omega_c + s_b\omega_b - \delta)t} e^{-\eta_c^2/2} e^{-(\eta_b)^2/2} \\ &\times \sum_{k_c=0}^{n_c^<} \sum_{k_b=0}^{n_b^<} \frac{(i\eta_c)^{2k_c+|s_c|} (i\eta_b)^{2k_b+|s_b|}}{k_c! (k_c + |s_c|)! k_b! (k_b + |s_b|)!} \times \sqrt{\frac{n_c^<! n_c^>! n_b^<! n_b^>!}{(n_c^< - k_c)! (n_b^< - k_b)!}} \\ &\times \langle n_c - k_c + s_c, n_b - k_b + s_b | n_c, n_b \rangle \end{aligned} \quad (196)$$

where $n_{c,b}^< = \min\{n_{c,b}, m_{c,b}\}$ and $n_{c,b}^> = \max\{n_{c,b}, m_{c,b}\}$. We define the coupling strength for the transition that changes the COM phonon number by s_c and the breathing phonon number by s_b :

$$\Omega_{n_c, n_c+s_c}^{n_b, n_b+s_b} = \Omega_0 e^{-\eta_c^2/2} e^{-\eta_b^2/2} \eta_c^{|s_c|} \eta_b^{|s_b|} \sqrt{\frac{n_c^<!}{n_c^>!}} \sqrt{\frac{n_b^<!}{n_b^>!}} L_{n_c^<}^{(|s_c|)}(\eta_c^2) L_{n_b^<}^{(|s_b|)}((\eta_b)^2) \quad (197)$$

such that

$$\langle e_1, g_2, n_c + s_c, n_b + s_b | H_I | g_1, g_2, n_c, n_b \rangle = \frac{\hbar}{2} e^{i(s_c\omega_z + s_b\sqrt{3}\omega_z)t} \Omega_{n_c, n_c+s_c}^{n_b, n_b+s_b} \quad (198)$$

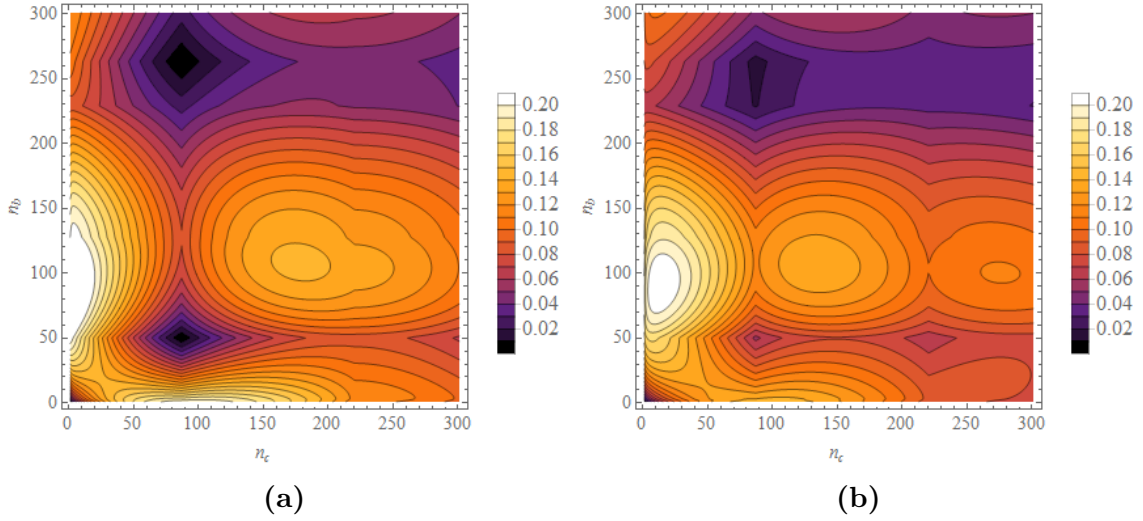


Figure VII.2: (a) Average coupling strength for the SBC sequence of a two-ion chain shown in table VII.1 with the intermodulation sideband removed. The presence of a zero-coupling region around the coordinates (87,51) can lead to population trapping. (b) With the intermodulation sideband (i.e. the full sequence of table VII.1), the coupling strength remains significant in this region.

Note the similarity with the coupling strength of a radial sideband. The coupling strength for the axial sidebands of a two-ion chain can also be represented in the form of a two-dimensional map with n_c and n_b as the variables. For a cooling sequence where several sidebands are addressed like the one in table VII.1, we can calculate an average coupling strength (Ω_{avg}) corresponding to the sum of the coupling strengths of the individual sidebands weighted by a coefficient equal to the fraction of the total cooling time when a given sideband is addressed. Mathematically:

$$\Omega_{avg}(n_c, n_b) = \sum_{s_c} \sum_{s_b} \alpha(s_c, s_b) \Omega_{n_c, n_c+s_c}^{n_b, n_b+s_b} \quad (199)$$

with

$$\alpha(s_c, s_b) = \frac{\text{Total pulse time on } s_c^{th} \text{ COM } s_b^{th} \text{ breathing}}{\text{Total cooling time}}. \quad (200)$$

Figure VII.2 shows the average coupling strength for the cooling sequence of table VII.1 without and with the intermodulation sideband. If this sideband is not present in the sequence, a region of zero-coupling can be seen on the map around the coordinates ($n_c = 87$, $n_b = 51$) where population trapping may occur. Adding the intermodulation sideband removes this problem as the coupling strength becomes significant in the incriminated region. Figure VII.3 shows the spectrum after sideband cooling at $\nu_z = 162$ kHz using the pulse sequence described above. The spectrum was acquired with the EMCCD camera which is able to distinguish the two ions. Figure VII.3 thus shows the probability of excitation of one of the ions of the chain. In contrast with the Doppler cooling spectrum, this spectrum has a

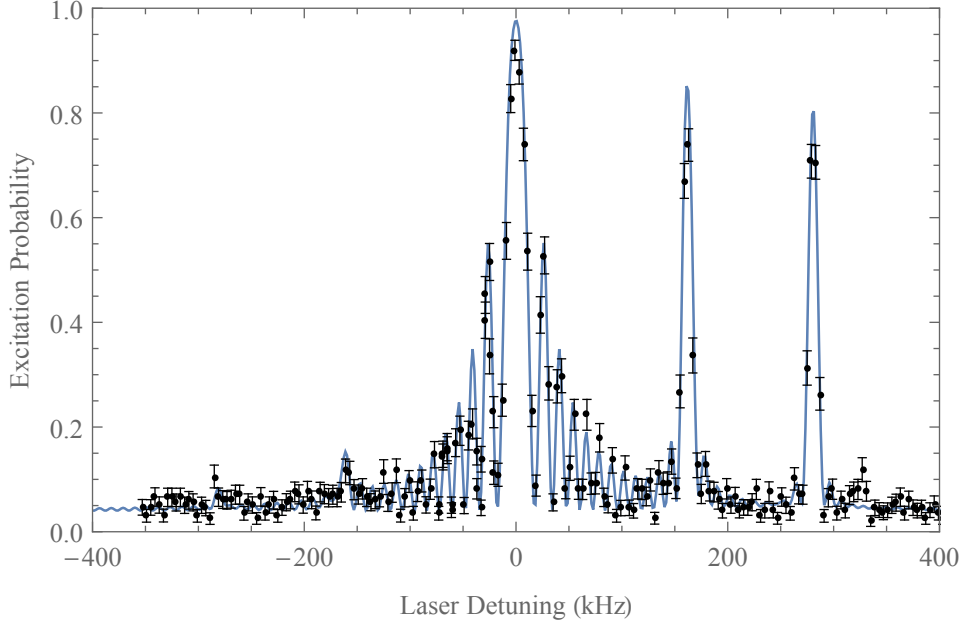


Figure VII.3: Spectrum of one ion of a sideband cooled two-ion chain at $\nu_z = 162$ kHz. The sideband cooling pulse sequence is shown in table VII.1, the axialisation amplitude is 1 V. A fit to the data points gives $\bar{n}_c = 0.25(6)$ and $\bar{n}_b = 0.07(4)$. The Rabi frequency is about 28 kHz.

simple sideband structure where only the carrier and the first blue sidebands of the COM and breathing modes are significant. The red sidebands are nevertheless not completely suppressed. To fit the spectrum and extract the average phonon numbers, we need to solve the Schrödinger equation for each sideband. This is based on a method developed in ref. [103]. Let the initial state of the ion be $|g_1, g_2, n_c, n_b\rangle$ and consider an excitation on the first blue sideband of the COM. The accessible states are $|g_1, g_2, n_c, n_b\rangle$, $|e_1, g_2, n_c + 1, n_b\rangle$, $|g_1, e_2, n_c + 1, n_b\rangle$ and $|e_1, e_2, n_c + 2, n_b\rangle$ (see figure VII.4). We can write the interaction Hamiltonian in matrix form in this basis:

$$H_I = \frac{\hbar}{2} \begin{pmatrix} 0 & -i\Omega_{n_c, n_c+1}^{n_b, n_b} e^{i(\delta-\omega_z)t} & -i\Omega_{n_c, n_c+1}^{n_b, n_b} e^{i(\delta-\omega_z)t} & 0 \\ i\Omega_{n_c, n_c+1}^{n_b, n_b} e^{-i(\delta-\omega_z)t} & 0 & 0 & -i\Omega_{n_c+1, n_c+2}^{n_b, n_b} e^{i(\delta-\omega_z)t} \\ i\Omega_{n_c, n_c+1}^{n_b, n_b} e^{-i(\delta-\omega_z)t} & 0 & 0 & -i\Omega_{n_c+1, n_c+2}^{n_b, n_b} e^{i(\delta-\omega_z)t} \\ 0 & i\Omega_{n_c+1, n_c+2}^{n_b, n_b} e^{-i(\delta-\omega_z)t} & i\Omega_{n_c+1, n_c+2}^{n_b, n_b} e^{-i(\delta-\omega_z)t} & 0 \end{pmatrix}. \quad (201)$$

Note that we do not consider two-photon processes, therefore the matrix element $\langle e_1, g_2, n_c + s_c, n_b + s_b | H_I | g_1, g_2, n_c, n_b \rangle$ is zero. The resulting system of differential equations is solved numerically. The probability of excitation observed on the spectrum of ion 1 corresponds to the sum of the probability that ion 1 is excited and ion 2 in the ground state and the probability that both ions are excited. The numerical solution for the probability is used to fit the data points, considering a thermal distribution of the population. We perform the same analysis for the second ion (the

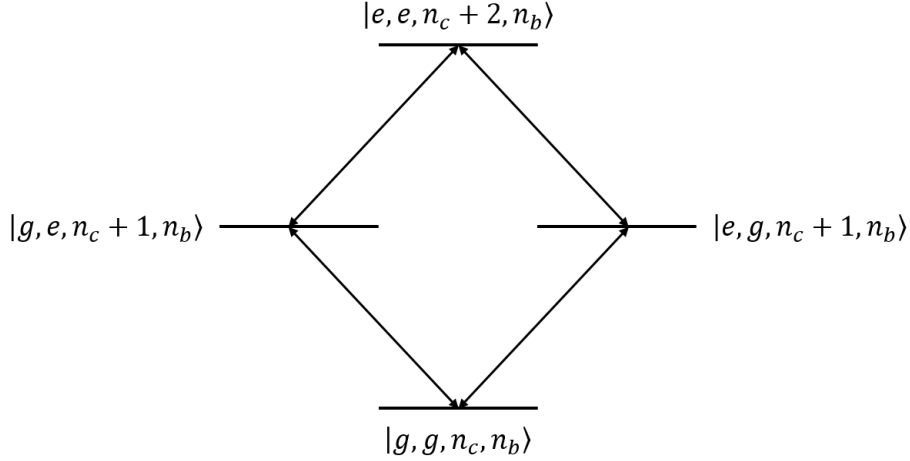


Figure VII.4: Manifold for excitation on the first blue sideband of the COM mode of a two-ion chain and the possible transitions represented by arrows. Off-resonant excitations of other sidebands are not considered.

two ions are illuminated equally) and we take the mean of the final average phonon numbers found for the two ions: $\bar{n}_c = 0.36(6)$ and $\bar{n}_b = 0.07(4)$. The heating rates are $11(2) \text{ s}^{-1}$ for the COM and $1(1) \text{ s}^{-1}$ for the breathing mode [54]. Due to magnetic field inhomogeneities, the resonant frequencies of the sidebands are slightly different for the two ions. This is visible on the spectrum when looking closely at a sideband (see figure VII.5). The difference in resonant frequency between the two ions is $2.0(5) \text{ kHz}$. This corresponds to a difference in magnetic field seen by the ions of about $0.18(5) \text{ } \mu\text{T}$. The distance between the ions at this frequency is $18.86 \text{ } \mu\text{m}$ as calculated with equation 46 and assuming a linear dependence of the magnetic field, the gradient around the centre of the trap is $9(2) \text{ mT m}^{-1}$. Despite being quite small, the frequency shift is a problem if one wants to drive coherently a sideband of the two-ion chain. Figure VII.6 shows the Rabi oscillations recorded when driving the first blue sideband of the breathing mode. The laser frequency is tuned to be close to resonance for one of the ions. The other ion experiences an additional detuning and the oscillations quickly fade away.

Although it is possible to perform sideband cooling on a two-ion chain in our trap as we have just seen, the technique is not scalable to longer chains. The ever lower axial frequencies needed to maintain the chain configuration when the number of ions increases are incompatible with sideband cooling.

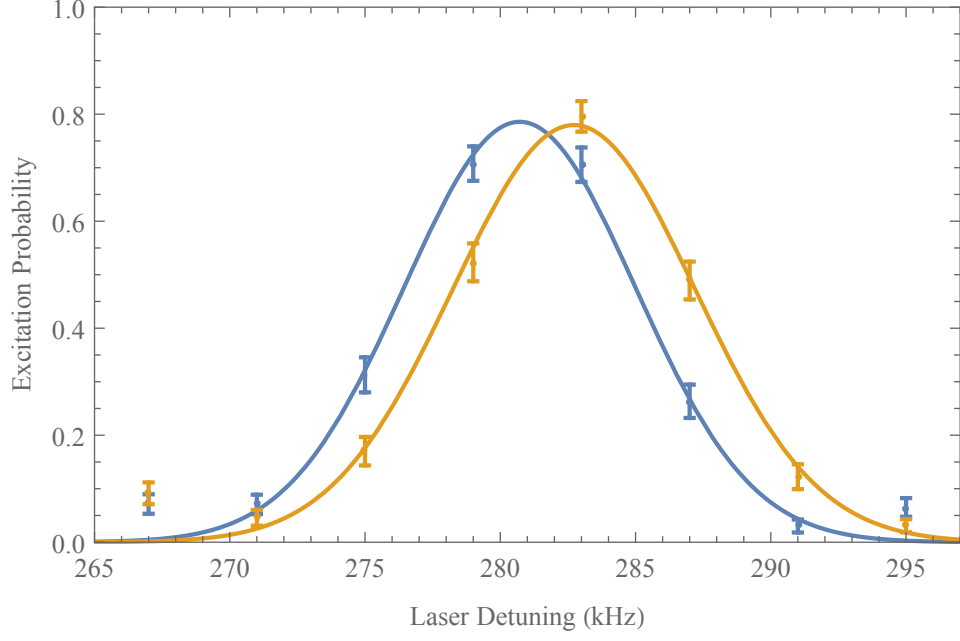


Figure VII.5: First blue sideband of the breathing mode for ion 1 (in blue) and ion 2 (in orange). The spacing between the centre frequencies of the two peaks is 2.0(5) kHz. The data points are fitted with Gaussian functions.

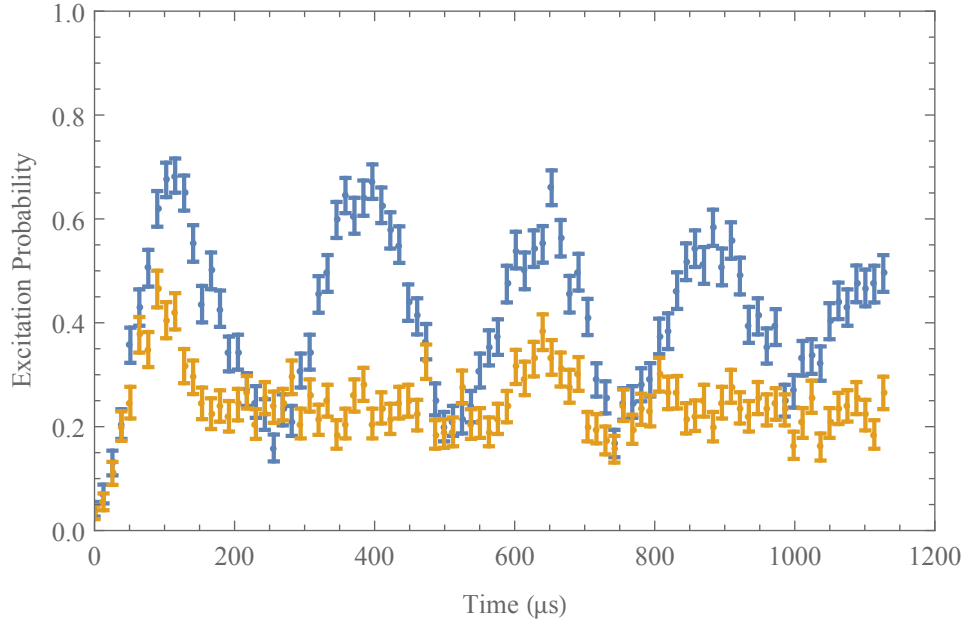


Figure VII.6: Rabi oscillations on the first blue sideband of the breathing mode (ion 1 in blue, ion 2 in orange). Because of the frequency shift due to magnetic field inhomogeneities, the laser light appears detuned for one of the ions resulting in reduced oscillations.

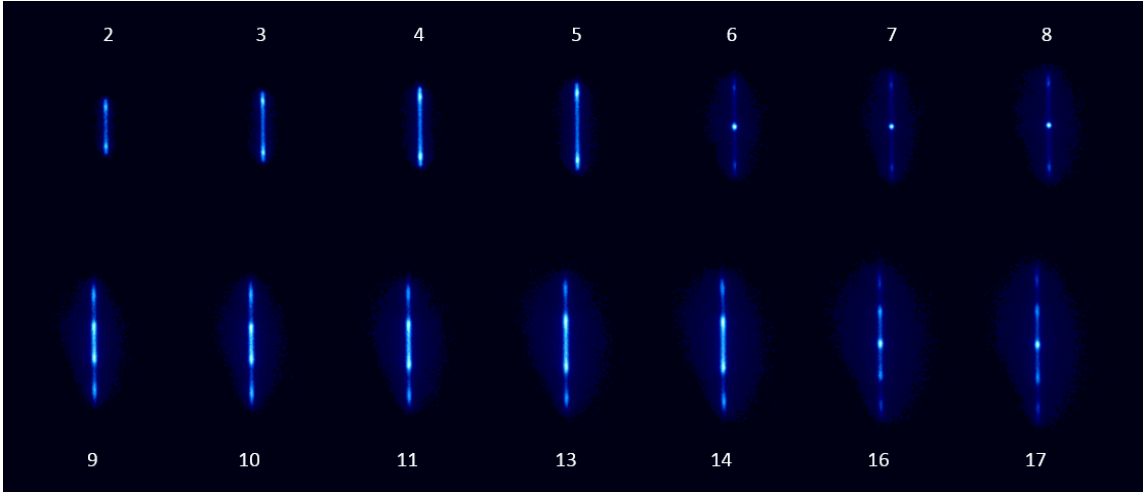


Figure VII.7: EMCCD pictures of planar crystals with different numbers of ions. All pictures were taken with the same experimental conditions (trapping potential, axialisation, laser cooling parameters, etc). The number of ions is determined by putting the crystals in a chain configuration at low voltage where the ions can be counted. The horizontal axis on the pictures corresponds to the magnetic field axis.

VII.2 Planar Crystals

When the axial trapping strength is large enough – and therefore the axial frequency high – the trapped ions form planar crystals with the ions rotating around the magnetic field axis. Depending on the number of ions, these crystals may contain one or several “shells” or “rings” where ions are (almost) equidistant from the trap centre. As we saw in chapter II, the positions of the ions can be calculated by minimising the potential energy of the crystal. Figure VII.7 shows pictures of planar crystals of various sizes taken with the EMCCD camera. Although imaging from the top (in the axial direction) would be better to see the structure, the different shells are visible on these images, which are taken in the radial direction, and match theoretical predictions.

Two-dimensional planar crystals have been proposed for applications in QIP [40, 42, 104, 105], for instance to simulate frustrated magnetic systems owing to the triangular lattice naturally formed by the ions in Penning traps. Experimental realisations have demonstrated their utility in large scale quantum simulations of the Ising model with hundreds of ions [3, 45]. Smaller ICCs have also been suggested as a medium to implement quantum error-correction protocols [44, 106] using global pulses rather than individual addressing of ions which is challenging for planar crystals [43]. These schemes all make use of the transverse (axial) motion of the crystals to some extent. Here we show results of sub-Doppler cooling of the axial modes of small planar ICC using sideband cooling. We also investigate the influence of different experimental parameters on the rotation frequency of a two-ion crystal.

Two-ion planar crystal

A two-ion planar crystal has two axial motional modes: the centre of mass (COM) mode with frequency ν_z , equal to the single ion axial frequency, and the tilt mode with frequency $\nu_t = \sqrt{\nu_z^2 - \nu_{eff}^2}$. The effective radial frequency ν_{eff} which we will simply refer to as the effective frequency is a measure of the radial trapping strength and depends on the axial and cyclotron frequencies as well as on the rotation frequency (ν_r) of the crystal (equation 56). In the absence of a direct way to control it with a rotating wall, the rotation frequency is determined by several factors such as the axialisation drive amplitude, the detuning and the position of radial Doppler cooling beam. Experimentally, we find that in most cases the rotation frequency is close to its lower limit i.e. the magnetron frequency. This signifies that the effective frequency is small and therefore the frequency of the tilt mode close to that of the COM. Due to the proximity of the two modes, their respective sidebands are not distinguishable on the Doppler spectrum in figure VII.8 taken at $\nu_z = 350$ kHz which corresponds to a Lamb-Dicke parameter $\eta_c = 0.116$ for the COM mode. This relatively small Lamb-Dicke parameter as well as the near degeneracy of the modes explain the simple sideband structure on the spectrum of the planar crystal in contrast with what was observed for the two-ion chain. Another consequence is a certain ease with which sideband cooling can be carried out. The two-ion planar crystal can be cooled in a similar way as a single ion at high frequency: two pulses on the second and first red sidebands of the COM are sufficient to cool both modes close to the ground state and avoid population trapping, the tilt mode being excited off-resonantly.

Figure VII.9 shows the spectrum obtained after 10 ms of sideband cooling at $\nu_z = 346.7$ kHz. The power of the laser on the probe pulse is reduced (Rabi frequency around 15 kHz) to resolve the sidebands of the two modes which are spaced by less than 7 kHz. Because the ions are undistinguishable on the camera, it is not possible to acquire a spectrum for each of them individually and therefore we only use the PMTs to detect fluorescence. Due to the poor light collection efficiency in our system, it is difficult to differentiate between one or two ions fluorescing: there is a large overlap of the Poisson distributions in the PMT histograms. We can more reliably distinguish between both ions being dark or at least one bright, therefore we set the threshold such that the spectrum shows the probability of both ions being in the excited state. Qualitatively, the spectrum suggest that the average phonon numbers of the two modes are very low as the red sidebands are strongly suppressed (or not visible at all). Noticeably, the tilt blue sideband is broader and not as high as the COM one. We attribute this to instabilities in the rotation frequency of the crystal.

In order to fit the data points and find numerical values for the average phonon numbers, we employ a similar method as for the chain. However because the modes

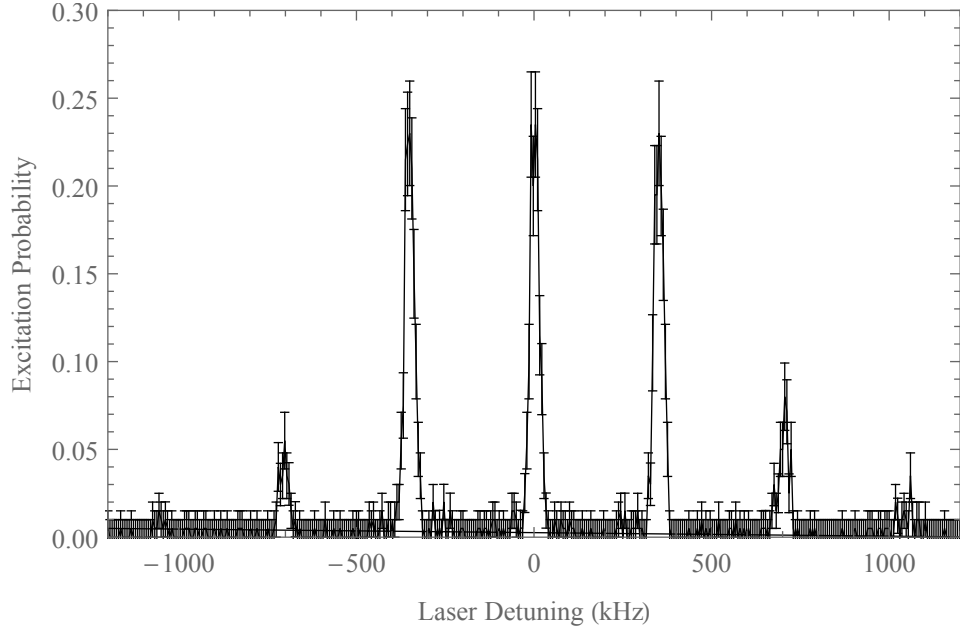


Figure VII.8: Spectrum of a two-ion planar crystal at $\nu_z = 350$ kHz after 12 ms of Doppler cooling.

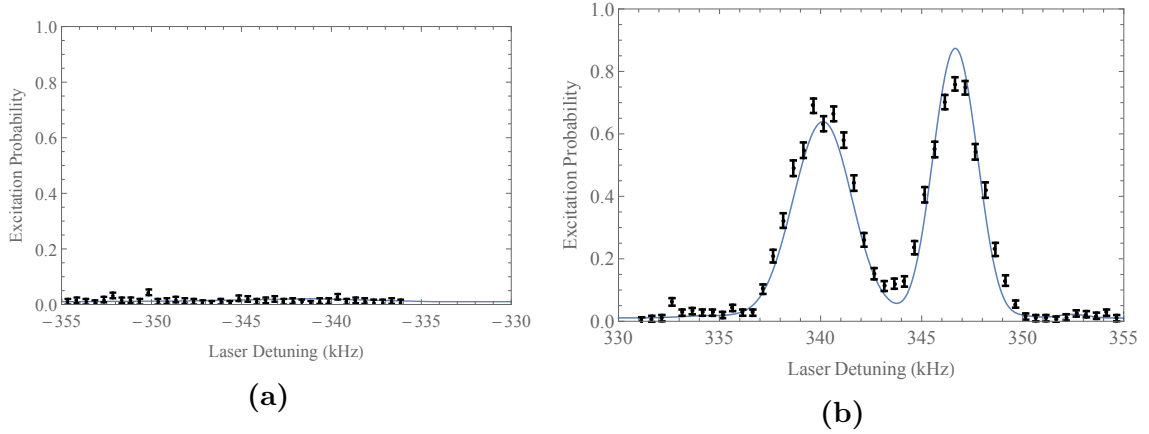


Figure VII.9: Spectrum of a two-ion planar crystal after 10 ms of sideband cooling on the first red sideband of the COM mode. (a) First red sidebands. (b) First blue sidebands. The spectrum represents the probability of both ions being excited. The frequencies of the modes are 346.7 kHz for the COM and 340.1 kHz for the tilt. The blue line is a fit to the data points and gives average phonon numbers $\bar{n}_c = 0.11(7)$ and $\bar{n}_t = 0.15(6)$ for the COM and tilt modes respectively. The standard deviation of the tilt frequency is around 1 kHz.

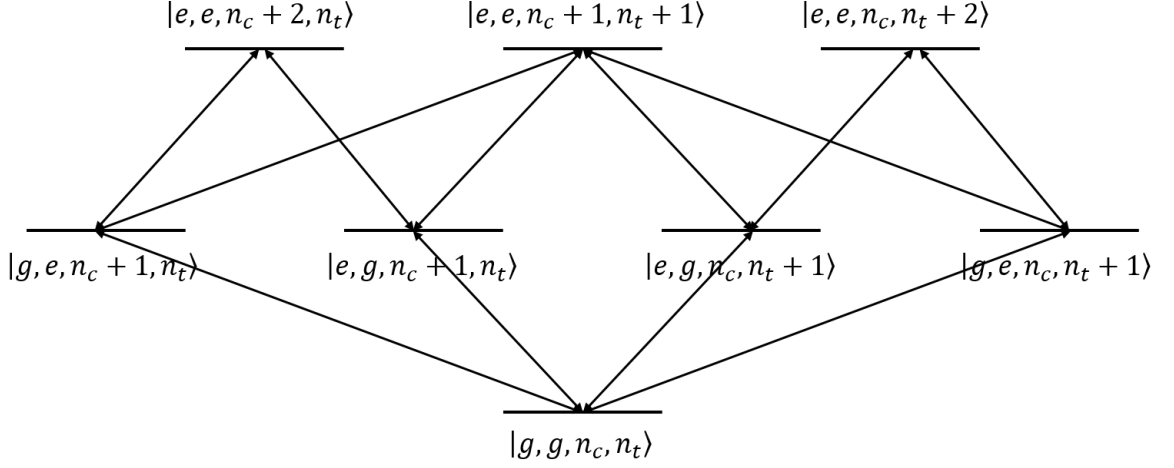


Figure VII.10: Manifold for excitation on the first blue sideband of a two-ion planar crystal and the possible transitions represented by arrows. Excitations of both modes are considered.

are close and in order to take into account off-resonant excitation, we do not solve the Schrödinger equation independently for each sideband. Instead, for the blue sidebands we consider a manifold with eight states shown in figure VII.10 and similarly for the red sidebands (the Hamiltonian is now a 8×8 matrix). The probability of both ions being excited is the sum of the probabilities to be in either $|e, e, n_c + 2, n_t\rangle$, $|e, e, n_c + 1, n_t + 1\rangle$ or $|e, e, n_c, n_t + 2\rangle$. To take into account the broadening of the tilt mode's peak, we consider the frequency of the tilt mode to be normally distributed. Fitting the data point with this method yields average phonon numbers $\bar{n}_c = 0.11(7)$ for the COM mode and $\bar{n}_t = 0.15(6)$ for the tilt mode. The standard deviation of the tilt mode's frequency is approximately 1 kHz. These numbers should nevertheless be considered with caution as the broadening of the peak and the absence of visible red sidebands make the fit less accurate. The heating rate is of the order of 1 s^{-1} for both modes.

The rotation frequency of the crystal, calculated from the positions of the sidebands is 106 kHz, roughly 9 kHz higher than the magnetron frequency. Interestingly, while the axial motion is close to the ground state, the rotation of the crystal gives the ions a tangential velocity in the radial plane of the order of 10 m s^{-1} which corresponds to a kinetic energy four orders of magnitude larger than the ground state energy of the axial motion.

In order to study the influence of experimental parameters on the rotation frequency of the crystal, we perform a series of experiments where these parameters are varied. We consider three factors: the amplitude of the axialisation, the detuning of the blue lasers and the position of the radial blue beam i.e. its offset from the trap

centre. All the experiments are performed at a constant trapping voltage of 170 V ($\nu_z = 347$ kHz). We use a 2^3 full factorial design (8 experimental runs) to study the effects of each factor individually as well as their joint effects. The levels (high and low) chosen for the different parameters are given in table VII.2 and are typical of laser cooling experiments. For each experimental run a picture is taken with the

Parameter	Label	High Level (+1)	Low Level (-1)
Axialisation	A	3.5 V	2 V
Detuning	D	-18 kHz	-7 kHz
Offset	O	Large	Small

Table VII.2: Parameters influencing the rotation frequency and their levels.

EMCCD camera. The width of the crystal on the image is used to determine the inter-ionic distance (1 pixel $\approx 1.32 \mu\text{m}$) which in turn gives access to the effective and rotation frequencies. For comparison, a sideband cooling spectrum is also acquired for each run. The effective and rotation frequencies are calculated from the measured spacing between the tilt and COM blue sidebands. This was done for several runs (17, including the 8 used in the factorial experiment) with different experimental conditions. The results are shown in figure VII.11. Although there is a good correlation between the two sets of measurements, the effective frequency calculated from spectroscopy is consistently lower than what is found through the EMCCD images (a linear regression gives a ratio of 0.87(7)). The spectra are acquired after sideband cooling when the blue lasers are off and it is possible that there is a relaxation of the effective radial frequency during the 10 ms of sideband cooling. The large errors on the data from the images are due to the limited resolution of the camera. The distance between the two ions on the image is only around 20 pixels such that the measurement error is quite large (about 10 %). The results of the factorial experiment should be understood in the light of these large errors.

The layout of the experiment and the results are summarised in table VII.3. The

Run	A	D	O	AD	AO	DO	ADO	ν_r (kHz)
1	-1	-1	-1	1	1	1	-1	120
2	1	-1	-1	-1	-1	1	1	125
3	-1	1	-1	-1	1	-1	1	103
4	1	1	-1	1	-1	-1	-1	108
5	-1	-1	1	1	-1	-1	1	103
6	1	-1	1	-1	1	-1	-1	108
7	-1	1	1	-1	-1	1	-1	102
8	1	1	1	1	1	1	1	105

Table VII.3: Experimental layout and measured rotation frequency, rounded to the nearest kHz.

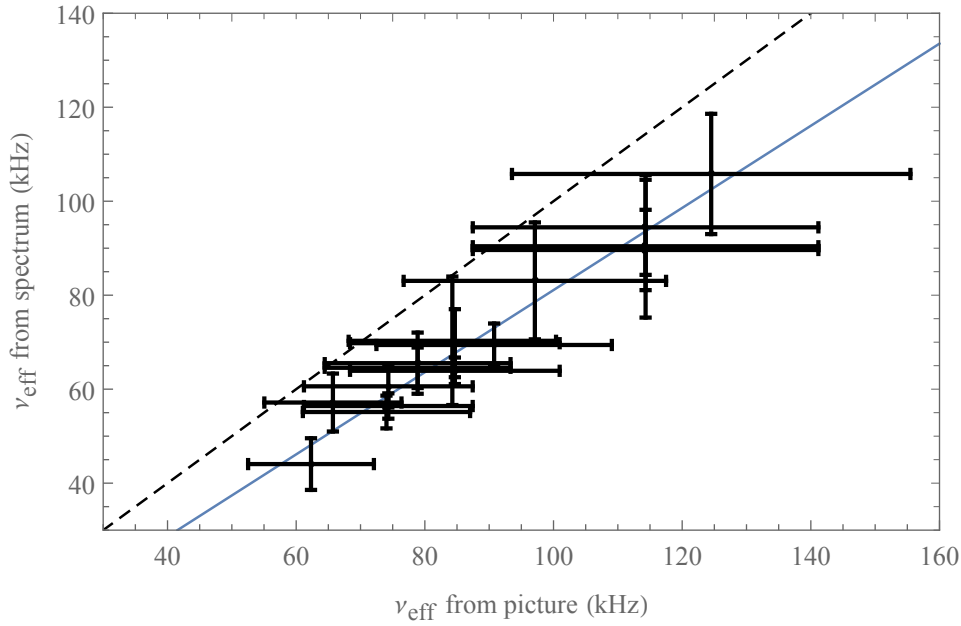


Figure VII.11: Effective radial frequency calculated from the frequency spacing of the COM and tilt modes on a sideband cooling spectrum as a function of the effective frequency calculated from the crystal's diameter on the EMCCD image. The blue line is a fit to the data points, its slope is 0.87(7). The dashed black line has a slope equal to 1.

average rotation frequency for these eight runs is 109.3 kHz and the coefficients for the three parameters and their interactions are shown in figure VII.12. The coefficient of a factor is calculated by adding the measured rotation frequencies measured on runs where the said parameter is +1 and subtracting those where it is -1. For example for A, the coefficient is $-120 + 125 - 103 + 108 - 103 + 108 - 102 + 105$. A positive value for the coefficient signifies that the rotation frequency is increased when the corresponding factor is +1. The larger (in absolute value) the coefficient, the stronger is the influence of the corresponding parameter. The magnitudes of the coefficients suggest there are four significant parameters: the three experimental variables individually and the interaction of the detuning and offset of the radial beam. As expected, a high axialisation amplitude reduces the diameter of the crystal and therefore increases the rotation frequency. The axialisation effect is not very sensitive to other factors, always changing the rotation frequency by a similar amount between the high and low levels (see figure VII.13). The detuning and offset of the radial blue beam have the strongest influence over the rotation frequency which increases for a small detuning and a small offset. There is also a strong interaction between these two parameters: the effect of the detuning depends greatly on the offset of the beam (and vice versa) and is much more significant when the offset is small (level -1) as can be seen in figure VII.14. Knowing the influence of these experimental parameters gives the experimenter the ability to exert some control on the rotation frequency of the crystal. The relative simplicity of sideband cooling of

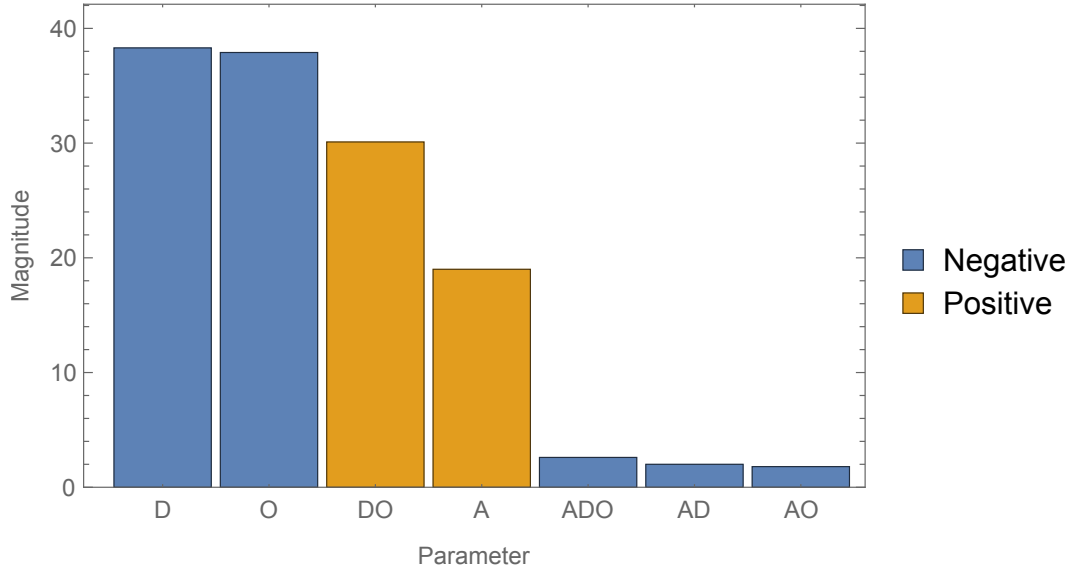


Figure VII.12: Pareto plot showing the magnitude of the coefficients for the three parameters influencing the rotation frequency and their interactions.

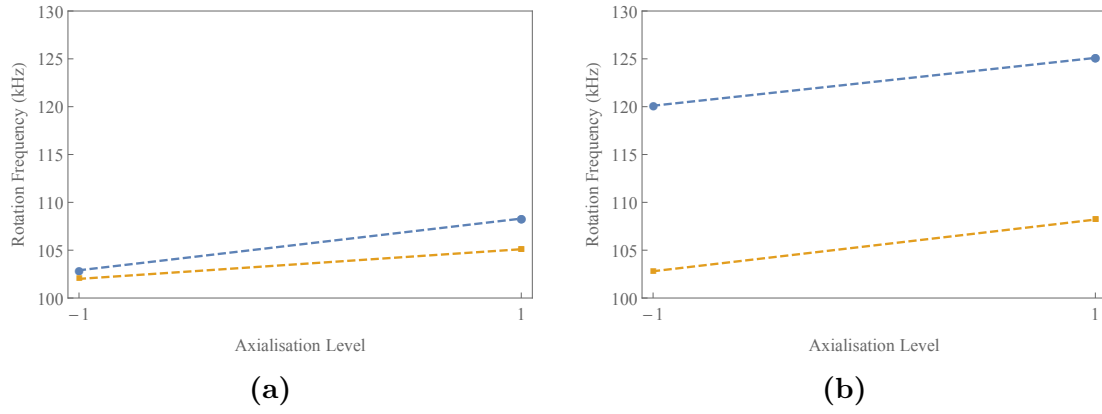


Figure VII.13: (a) Rotation frequency as a function of the axialisation level with offset at +1 and detuning at -1 (in blue) or +1 (in yellow). (b) Same with offset at -1.

the two-ion planar crystal relies in part on the possibility to excite the two modes simultaneously; a small rotation frequency is therefore preferable in this context. From the results of the factorial experiment, it appears that a large beam offset is better for the stability of the crystal as instabilities in the laser frequency will have a smaller effect.

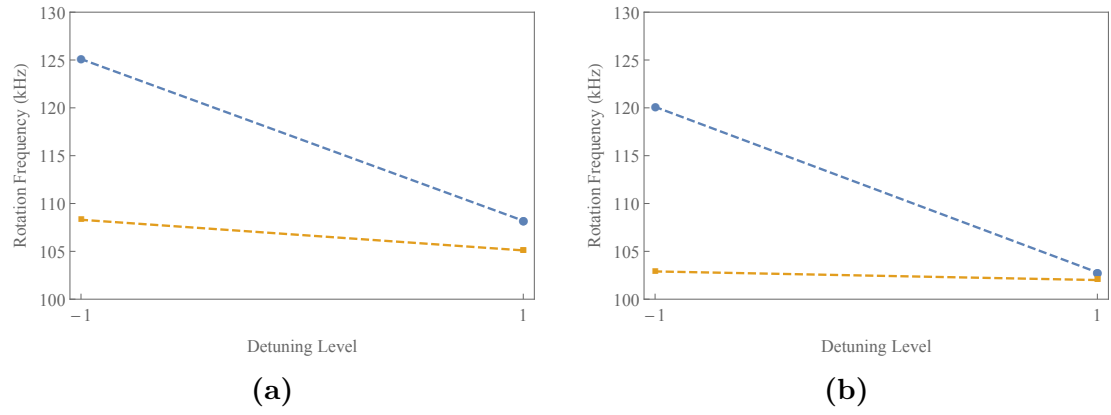


Figure VII.14: (a) Rotation frequency as a function of the detuning level with axialisation at +1 and beam offset at -1 (in blue) or +1 (in yellow). (b) Same with axialisation at -1.

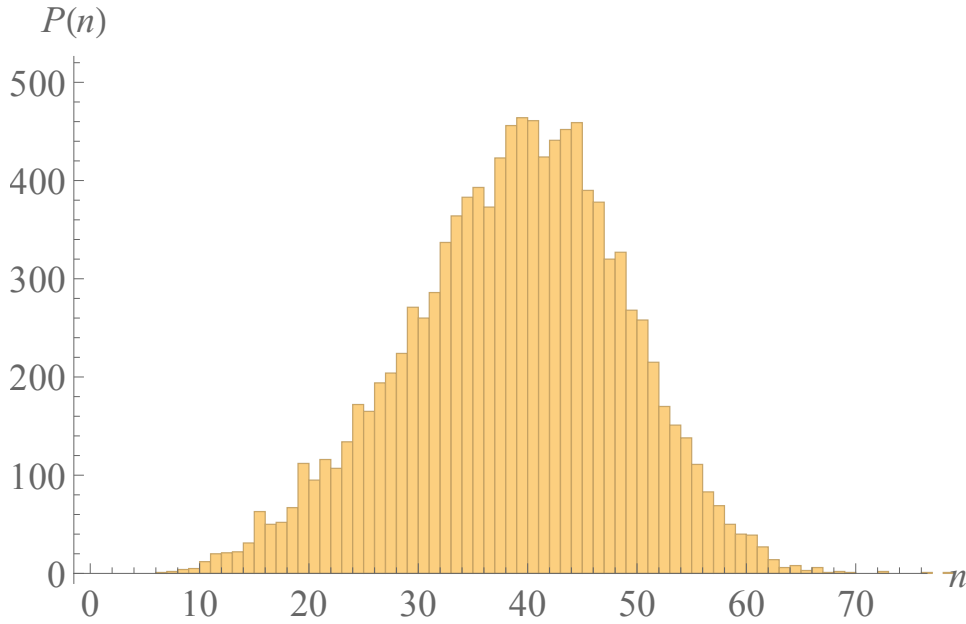


Figure VII.15: Histogram of the photon count after the probe pulse of the spectrum taken of a Doppler cooled 21-ion planar crystal.

Larger crystals

Up to five or six ions, laser cooling of planar ICCs remains relatively simple and does not differ significantly from the two-ion case. Working with larger crystals comes with additional difficulties in terms of cooling and detection. Even if the overall fluorescence increases with the number of ions, it is increasingly difficult to identify how many ions are fluorescing. On the histogram of the photon counts after the spectroscopic probe pulse, the Poisson distributions are merged into a continuum where it is not possible to set a meaningful threshold. Besides, with large crystals, because the probability to excite all the ions at once is low, the 100 to 400 hundred repetitions per data point of a typical experiment are not enough to see a significant dark distribution. As a consequence, it is sometimes not even possible to set a threshold corresponding to all the ions being in the excited state. This situation is illustrated by the histogram in figure VII.15 which was obtained from a Doppler cooled 21-ion crystal, the largest one for which we acquired a spectrum. As discussed earlier, increasing the detection time to better separate the Poisson distribution is not an option because of j -mixing. Increasing the number of repetitions substantially – say, 500 or more – to see a clearer dark distribution would lead to unreasonably long experiments. It should be kept in mind that some elements in our experimental set-up, the blue lasers in particular, are not very stable and running an experiment for more than a few hours is not recommended.

Thus, for large crystals, instead of looking at the spectra as a probability of excitation as a function of the detuning of the 729 nm laser, the total fluorescence count from the PMTs (or the camera) is used instead. The presence of a sideband

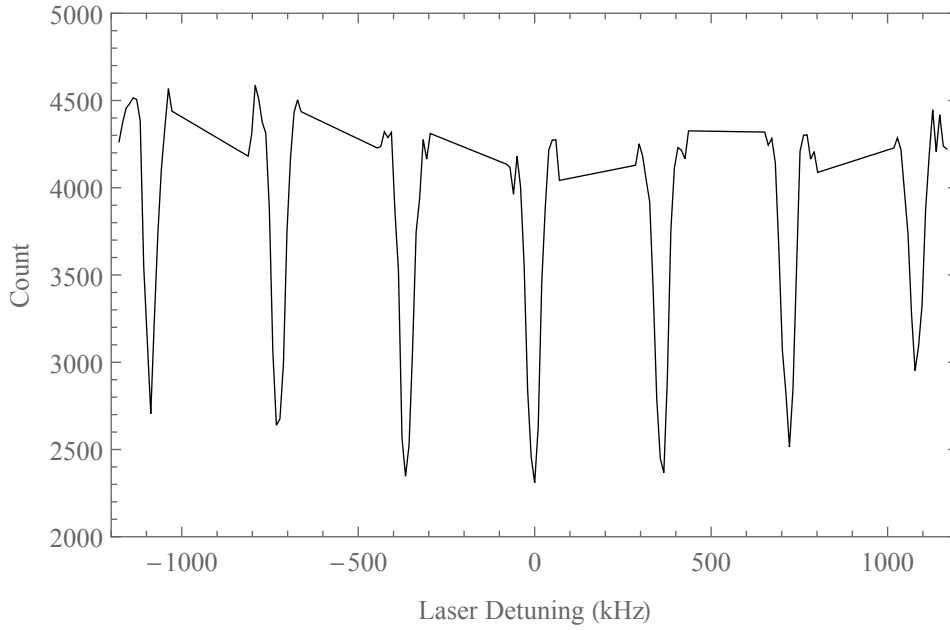


Figure VII.16: Doppler spectrum of a 21-ion planar crystal.

is marked by a dip in the fluorescence count as can be seen for the spectrum of the Doppler cooled 21-ion crystal in figure VII.16.

In the remainder of this chapter, we present results of sideband cooling of planar ICCs of different sizes as well as demonstrations of coherent driving of the ions' internal states via the observation of Rabi oscillations. Often, the spectra are windowed around the motional sidebands in order to save time. With planar crystals however it is interesting to scan the whole spectrum continuously as other features appear. Figure VII.17 shows the spectrum of a sideband cooled three-ion crystal where many peaks are visible in addition to the expected carrier and blue sidebands. These additional peaks are artefacts of the radial motion and their positions are combinations of the true cyclotron frequency ($\nu_c = 729.4$ kHz) and the rotation frequency ($\nu_r = 136$ kHz). The reason why these peaks are visible is that the 729 nm laser beam is not perfectly collinear with the z axis of the trap. The small component in the radial direction is sufficient to observe features of the radial motion. The absence of a visible red sideband for the axial modes indicates a high ground state occupation probability. The two modes (COM and tilt) are not resolved on this spectrum due to their proximity which is consistent with the low rotation frequency (the magnetron frequency is $\nu_- = 120$ kHz). The crystal was cooled using MSSBC with three 10 ms pulses on the three lowest order red sidebands (of the COM) in decreasing order.

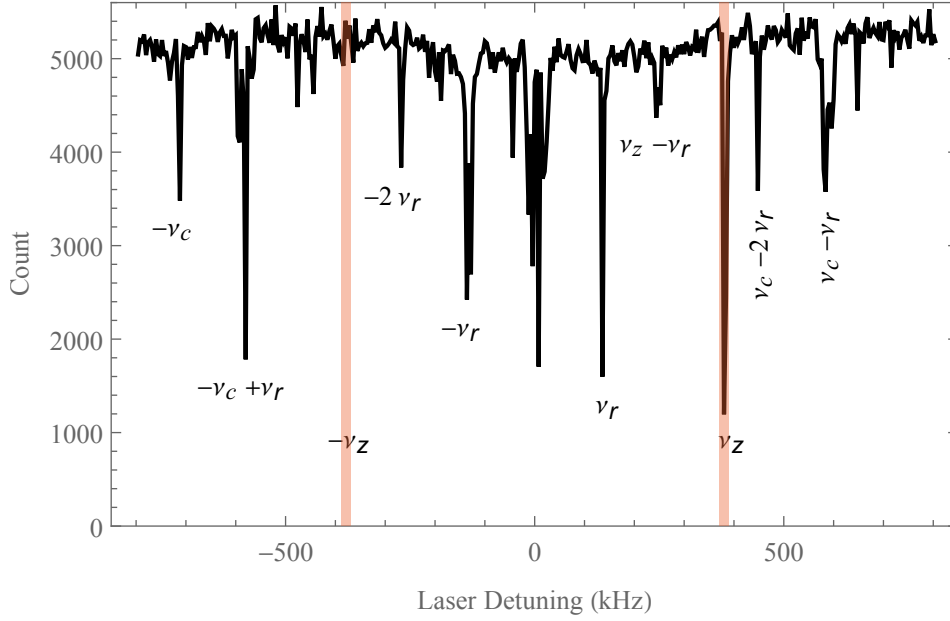


Figure VII.17: Total fluorescence count as a function of the laser frequency from a three-ion planar crystal. The crystal was sideband cooled for a total of 30 ms in a three-stage sequence with pulses on the third, second and first order red sidebands (10 ms each). The axial (COM) frequency is 382 kHz (210 V) and the rotation frequency about 136 kHz. The positions of the first blue and red axial sidebands are marked by orange stripes. The other peaks (except the carrier) are artefacts of the radial motion.

Similarly, a five-ion crystal was cooled with a two-stage sequence with 5 ms on the second red and 15 ms on the first red sideband at an axial frequency of 382 kHz, like the three-ion spectrum. Note that for a constant axial frequency, the Lamb-Dicke parameter decreases with a larger number of ions, reducing the chance of population trapping. The spectrum is shown in figure VII.18. Again, no red sideband is visible. Thanks to a finer frequency step and a larger splitting between the axial modes, their respective blue sidebands are resolved. The peaks are fitted with Gaussian functions to find their centre frequencies which are 381.7(2) kHz, 370.9(2) kHz and 363.0(2) kHz for the COM, tilt and folding modes respectively. The frequencies of the modes suggest that the rotation frequency of the crystal is about 137 kHz, close to what was found for the three-ion crystal. As an additional demonstration of the high ground state occupation probability, Rabi oscillations can be observed when driving the carrier transition with the 729 nm laser. Figure VII.19 shows the probability for all five ions to be in the excited state as a function of the probe pulse duration. The fluorescence detection threshold was set to separate the dark distribution from any fluorescence from the ions; it was possible in this case thanks to a significant dark distribution on the histogram. The probability for all ions to be excited on the carrier is simply the product of the individual probabilities as the absorptions of a photon by the ions of the crystal are independent events (not true

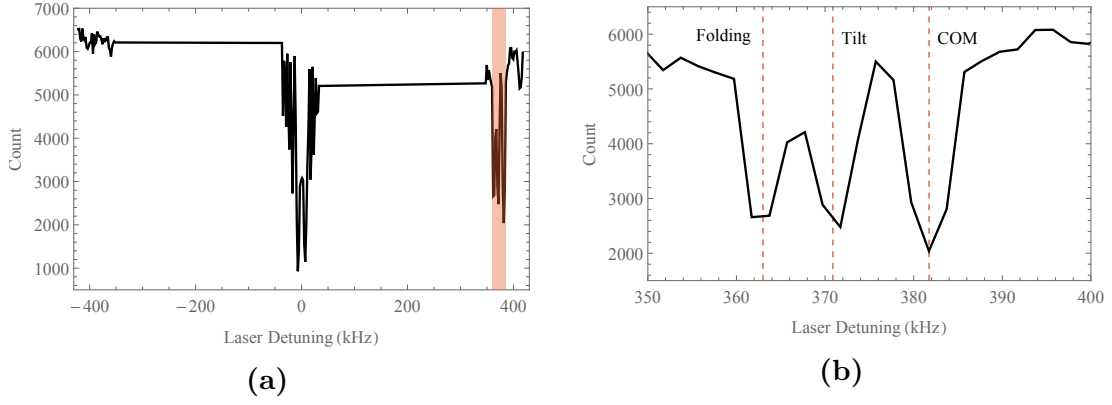


Figure VII.18: (a) Total fluorescence count as a function of the laser frequency from a five-ion planar crystal. The crystal was sideband cooled for a total of 20 ms in two stages. The trapping frequency is $\nu_z = 382$ kHz (trapping voltage 210 V) and the axialisation amplitude 3.5 V. (b) Expanded view of the blue sidebands. The sidebands from the different modes are well resolved and can be identified (see appendix B.2 for a spatial representation of the axial modes).

for a sideband excitation). Hence, on resonance we measure:

$$\begin{aligned}
 P_e(\Omega_0, t) = & \sum_{n_1=0}^{\infty} \sum_{n_2=0}^{\infty} \sum_{n_3=0}^{\infty} \sum_{n_4=0}^{\infty} \sum_{n_5=0}^{\infty} \frac{\bar{n}_1^{n_1}}{(\bar{n}_1 + 1)^{n_1+1}} \times \frac{\bar{n}_2^{n_2}}{(\bar{n}_2 + 1)^{n_2+1}} \times \frac{\bar{n}_3^{n_3}}{(\bar{n}_3 + 1)^{n_3+1}} \\
 & \times \frac{\bar{n}_4^{n_4}}{(\bar{n}_4 + 1)^{n_4+1}} \times \frac{\bar{n}_5^{n_5}}{(\bar{n}_5 + 1)^{n_5+1}} \times \prod_{k=1}^5 \sin^2 \left(\frac{\Omega(k, n_1, n_2, n_3, n_4, n_5)t}{2} \right).
 \end{aligned} \tag{202}$$

The product of squared sines explains the unusual shape of the Rabi oscillations in figure VII.19. The numbers n_i , $i \in \{1, 2, 3, 4, 5\}$ are the phonon numbers for the five motional modes. Ω is the coupling strength on the carrier transition and depends on the the phonon numbers and the Lamb-Dicke parameters which differ for each mode and ion (index k in the equation). However, in the Lamb-Dicke regime and close to the ground state, $\Omega \simeq \Omega_0$. We use this approximation to fit the data points which means the phonon number dependency is lost (and therefore we do not use a thermal model) and, in order to model the damping of the oscillations, an exponential decay is added. No particular physical origin is assumed for the exponential decay but probable contributions are non-zero average phonon numbers and decoherence. The fit gives a Rabi frequency of about 30 kHz. In a five-ion planar crystal, the ions are arranged in a regular pentagon, equidistant from the centre of the trap. As a consequence, all the ions are illuminated with the same intensity by the 729 nm laser beam. This is not the case for a six-ion ion crystal (or any larger crystal) which has one central ion and five on a pentagon (ring). Because of the Gaussian intensity profile of the beam, the central ion experiences a stronger intensity than the outer ones on the ring which means that they are driven with a different Rabi frequency than the central one. Figure VII.20 shows Rabi oscillations obtained by measuring

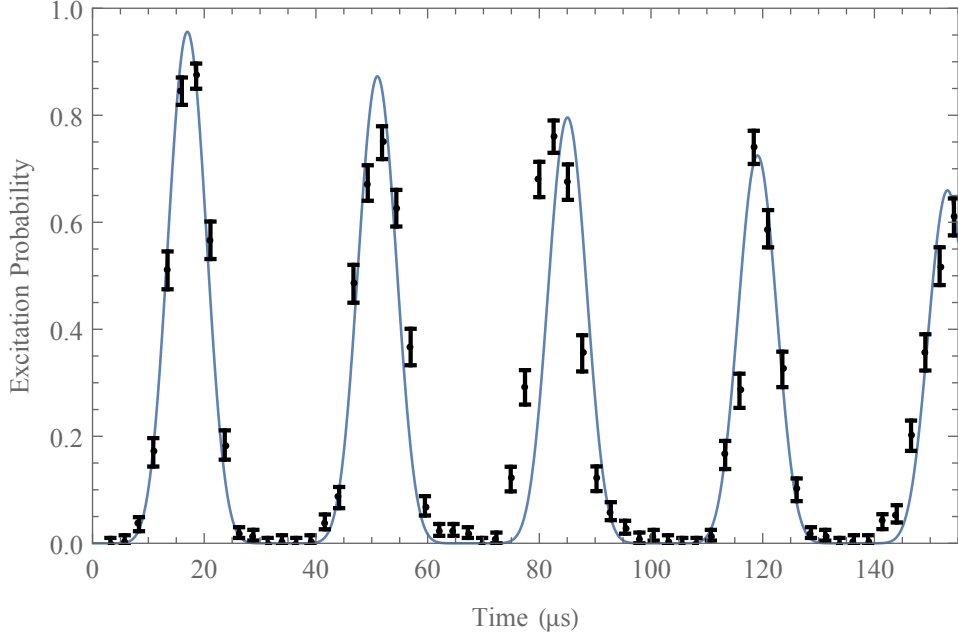
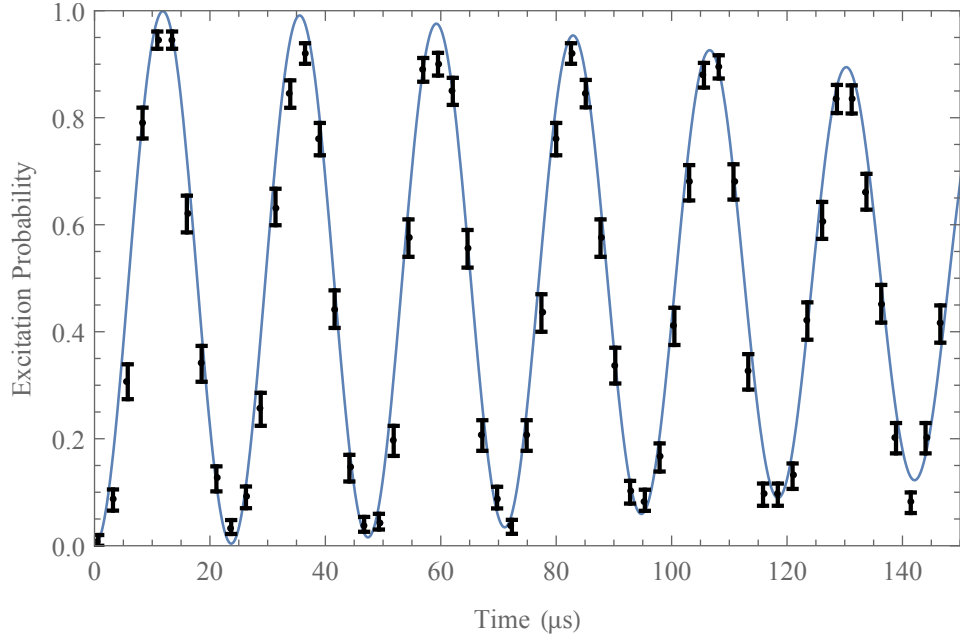


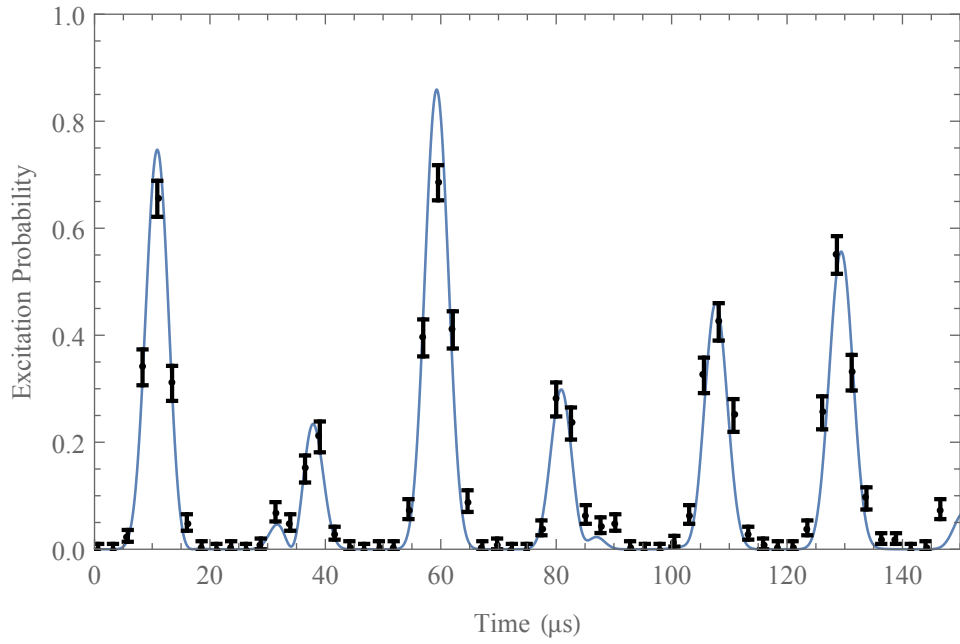
Figure VII.19: Probability of simultaneous excitation of the carrier transition of five ions in a planar crystals showing Rabi oscillations. The crystal was initially sideband cooled for 20 ms. The trapping frequency is $\nu_z = 385$ kHz (trapping voltage 210 V) and the axialisation amplitude 3.5 V. The blue line is a fit to the data points giving a Rabi frequency around 30 kHz and a decay time (assumed exponential) of 0.35(6) ms.

the probability of excitation of one ring ion (a) and the probability for all six ions to be excited (b). The Rabi oscillations on the ring ion was obtained thanks to the EMCDD camera by selecting a region of interest at the edge of the planar crystal and collecting fluorescence only from that particular region that is only from the ring ions (plus some background). This allows us to measure the Rabi frequency for the ring ions which comes to be about 43 kHz. The data on figure VII.20.b come from the fluorescence count of the PMT which is not region specific. The oscillations observed are the result of the combination of two signals with different frequencies. If Ω_1 designates the Rabi frequency for the central ion and Ω_2 that for the ring ions, the probability of excitation of all six ions has a sinusoidal factor of the form: $\sin^2(\Omega_1 t/2) \times \sin^{10}(\Omega_2 t/2)$ where the power 10 is here to take into account the five ring ions. Fitting the data under the same assumptions as for the five-ion crystal gives a Rabi frequency for the central ion of 60 kHz, rounded to the closest kHz, and 43 kHz for the ring ions. This suggests that the intensity of the laser, which is proportional to the square of the Rabi frequency, is almost twice as high at the centre than at the ring. Driving all the ions with the same Rabi frequency would require a much more loosely focused beam to have a (almost) uniform intensity over the crystal. The power of the laser should however be substantially increased to maintain a high Rabi frequency.

The SBC results presented so far were obtained by exciting the red sidebands of all the modes simultaneously with a single laser frequency. This was possible



(a)



(b)

Figure VII.20: (a) Probability of excitation of the carrier transition of at least one ring ion of a six-ion planar crystal. The crystal was initially sideband cooled for 20 ms. The trapping frequency is $\nu_z = 356$ kHz (trapping voltage 180 V) and the axialisation amplitude 3 V. The blue line is a fit to the data points giving a Rabi frequency of 42.9(1) kHz. The same method as for fitting Rabi oscillations of a single ion is used (with a Gaussian decay). (b) Probability of simultaneous excitation of the six ions. The fit assumes five ions with an identical Rabi frequency (the ones on the ring) and one with a different one (the central ion) and yields respectively 43.0(1) kHz and 59.7(1) kHz. The errors given for the frequencies are the fitting errors; they do not take into account the approximations of the model.

Detuning (kHz)	Pulse time (ms)
-359	5
-332	15
-317	10
-359	1

Table VII.4: Pulse sequence used for sideband cooling of a ten-ion planar crystal.

thanks to a limited number of modes (four non-degenerate for six ions) and low rotation frequencies which kept the axial modes close together. As the number of ions increases, so does the number of modes and their spacing so that exciting all the sidebands off-resonantly is not possible any more. The largest crystal for which we are able to repeatedly achieve near ground state cooling for all modes contains ten ions¹. Such a crystal has an inner disc with two ions and an outer one with eight ions, arranged in a slightly distorted octagon and nine non-degenerate axial modes (shown in appendix B.2). Figure VII.21 shows the spectrum after sideband cooling; the axial trapping frequency (COM) is $\nu_z = 356$ kHz and the axialisation amplitude 3 V. A four-stage sequence, shown in table VII.4, was used for a total cooling time of 31 ms. It is not necessary to address all the sidebands individually, three different frequencies are used to excite off-resonantly the nine modes. From the radial artefacts on the spectrum, the rotation frequency is measured to be 117 kHz. Numerical calculations for $\nu_z = 356$ kHz, $\nu_c = 729.4$ kHz and $\nu_r = 117$ kHz indicate that the frequencies of the axial modes should be contained between 310 kHz and 356 kHz (figure VII.22). This range (orange stripe on figure VII.21) matches well what is observed on the spectrum. There is no significant dip in the fluorescence count at the position of the red sidebands which demonstrates near ground state cooling for all modes.

Attempts to cool even larger crystals proved challenging and it is uncertain whether sideband cooling of planar crystals is scalable beyond ~ 15 ions. A loosely focused beam with a significant intensity over the extent of the crystal would certainly help the cooling process. Note that for crystals with a central ion, it is not possible to cool all the ions sympathetically by addressing only the central one which is stationary for certain motional modes (e.g. modes 2, 3, 4 and 5 of a seven-ion crystal). A better control of the crystal's rotation could also be beneficial to SBC: using a rotating wall, the rotation frequency could be set to a value close to the magnetron frequency (its lower limit) where the different axial modes are almost degenerate. This would facilitate off-resonant excitation of the sidebands and relax the necessity to use several cooling frequencies. Electromagnetically-induced-transparency cooling may be a good alternative to reach sub-Doppler temperatures with large crystals. Ground

¹There is some evidence of sideband cooling on 12 and 15-ion crystals from a limited number of spectra.

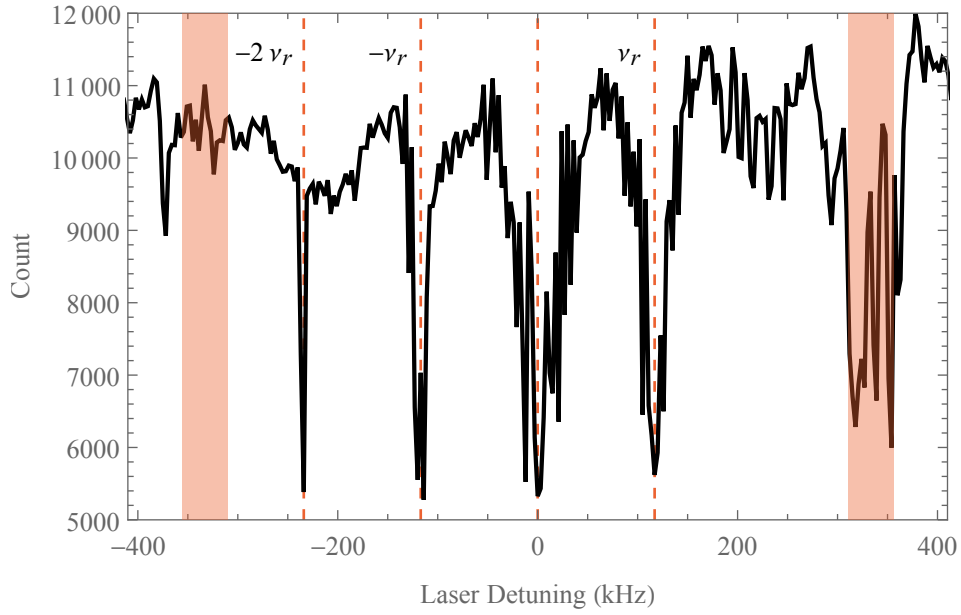


Figure VII.21: Total fluorescence count as a function of the laser frequency from a ten-ion planar crystal. The ions were sideband cooled for 31 ms in four stages (see text for details). The rotation frequency measured from the spectrum is 117 kHz. The orange bands corresponds to the range of frequencies spanned by the different axial modes (red and blue sidebands) of a ten-ion planar crystal as calculated for $\nu_z = 356$ kHz.

state cooling was demonstrated with this technique on an eighteen-ion chain held in a linear Paul trap [107].

Experiments with three-dimensional crystals were unsuccessful. On spectra taken after Doppler cooling, no clear feature could be seen even at the position of the carrier transition. This is probably because of instabilities in the crystal configuration which may change when the Doppler cooling lasers are turned off to probe the $S_{1/2} \longleftrightarrow D_{5/2}$ transition. Again, fixing the rotation frequency of the crystal with means independent from the lasers may help to maintain the crystal structure.

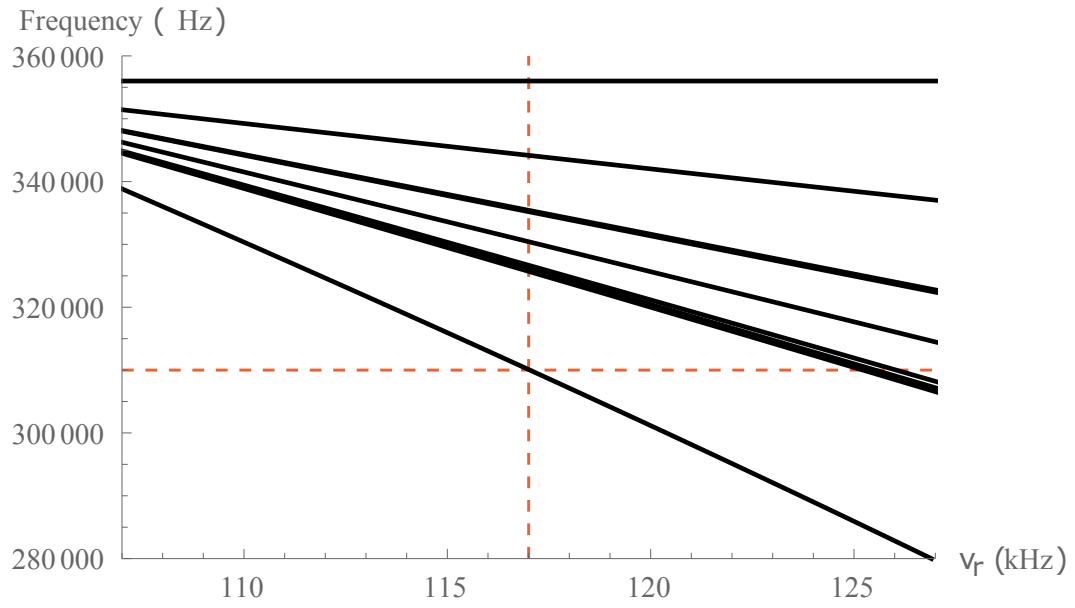


Figure VII.22: Frequency of the axial modes of a ten-ion planar crystal as a function of the rotation frequency. Calculated for $\nu_z = 356$ kHz and $\nu_c = 729.4$ kHz. For $\nu_r = 117$ kHz, the frequencies of the axial modes are between 310 kHz and 356 kHz. Spatial representations of the modes of a ten-ion planar crystal are shown in appendix B.2.

VIII

Coherence Studies on a Single Ion

This last chapter concerns the coherent manipulations of the electronic and axial vibrational states of a single ion. In these experiments, the quantum state of the ion is modified in a controlled way thanks to appropriate pulses of the 729 nm laser. We demonstrate the creation of coherent superpositions of the electronic states (section VIII.1) as well as of the vibrational states (section VIII.2 and VIII.3) and perform Ramsey experiments to measure coherence times which we use to characterise our system. In section VIII.4, we show the creation of coherent states using a bichromatic excitation.

In general, the quantum state $|\psi(t)\rangle$ of the ion can be described as a superposition of the electronic and vibrational states:

$$|\psi(t)\rangle = \sum_{n=0}^{\infty} c_{g,n}(t) |g, n\rangle + c_{e,n}(t) |e, n\rangle. \quad (203)$$

For the experiments we will describe below, it is useful to calculate the propagator governing the evolution of the quantum state when a sideband is excited. Consider the transition $|g, n\rangle \longleftrightarrow |e, n+s\rangle$. The coefficients associated with these two states, at a time t of the interaction, are given by:

$$\begin{pmatrix} c_{e,n+s}(t) \\ c_{g,n}(t) \end{pmatrix} = T_n^s \begin{pmatrix} c_{e,n+s}(0) \\ c_{g,n}(0) \end{pmatrix} \quad (204)$$

where the matrix T_n^s is the propagator which is explicitly, in the $\{|e, n+s\rangle, |g, n\rangle\}$ basis [64]:

$$T_n^s = \begin{pmatrix} e^{-i\delta t/2} \left(\cos(\Omega t/2) + i\frac{\delta}{\Omega} \sin(\Omega t/2) \right) & -i\frac{\Omega_{n,n+s}}{\Omega} e^{i(\phi+|s|\pi/2-\delta t/2)} \sin(\Omega t/2) \\ -i\frac{\Omega_{n,n+s}}{\Omega} e^{-i(\phi+|s|\pi/2-\delta t/2)} \sin(\Omega t/2) & e^{i\delta t/2} \left(\cos(\Omega t/2) - i\frac{\delta}{\Omega} \sin(\Omega t/2) \right) \end{pmatrix} \quad (205)$$

with

$$\Omega = \sqrt{\delta^2 + \Omega_{n,n+s}^2}. \quad (206)$$

The detuning δ is measured relatively to the resonant frequency of the sideband targeted i.e. $\delta = \omega - (\omega_0 + s\omega_z)$ with ω the frequency of the laser. ϕ designates the initial phase of the laser.

VIII.1 Optical Coherence

In this section, we seek to measure the coherence time of the optical qubit formed by the states $S_{1/2,-1/2}$ and $D_{5/2,-3/2}$ of $^{40}\text{Ca}^+$ later denoted $|g\rangle$ and $|e\rangle$ respectively. Interactions between the environment and the ion, experimental errors as well as imperfections in the set-up lead to an uncontrollable and unwanted evolution of the system which eventually loses its coherent behaviour. This decoherence represents a major obstacle for QIP where quantum coherence needs to be maintained. Two types of decoherence may occur: spin flip and dephasing. In our case, spin flip occurs in case of spontaneous emission of a photon leading to the decay of the ion from $D_{5/2}$ to $S_{1/2}$. Owing to the relatively long lifetime of the $D_{5/2}$ state, spin flip is a negligible source of decoherence in the time scale of our experiments.

Ramsey experiments

Ramsey interferometry is a well established technique and broadly used in atomic physics. It consists in applying two $\pi/2$ pulses separated by a wait time where the laser is turned off so that the ion evolves freely. Here, we manipulate the quantum state of the ion using pulses on the carrier transition and any off-resonant excitation of the sidebands is neglected.

Because the Rabi frequency depends on the motional state, the ion should have a well defined phonon number i.e. it should be prepared in a motional Fock state so that we can apply a $\pi/2$ pulse. The most reliable way to do so is to sideband cool the ion to the ground state. In order to ensure a maximum occupation probability of the ground state, the experiments are carried out at a high trapping frequency. At this point, the ion can be reduced to a two-level system driven by a near-resonant electromagnetic wave and therefore, at any point of the Ramsey experiment. The internal state of the ion is described by a superposition of $|e\rangle$ and $|g\rangle$:

$$|\psi(t)\rangle = c_e(t) |e\rangle + c_g(t) |g\rangle. \quad (207)$$

There are three possible ways to record an interference pattern: by changing the detuning of the laser, by changing the relative phase of the laser between the two $\pi/2$ pulses or by changing the wait time between the two pulses. For the last

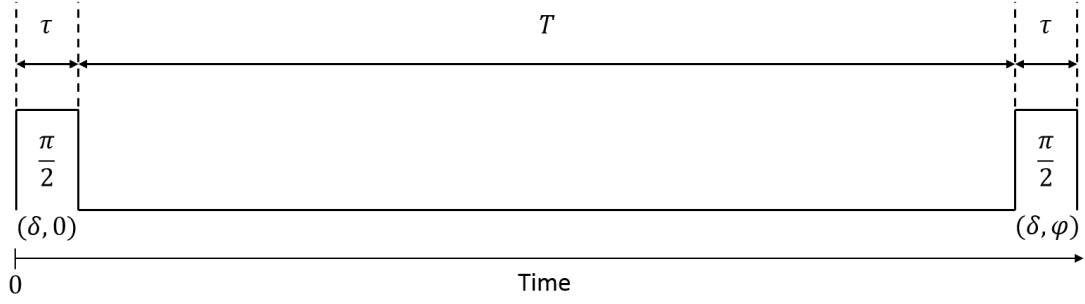


Figure VIII.1: Pulse sequence of a Ramsey experiment.

method to work, the detuning should be constant and non-zero. Let us consider an ion initially in the ground state $|g\rangle$. A near-resonant $\pi/2$ pulse of duration¹ $\tau = \pi/(2\Omega_{0,0})$ is applied. We assume δ is small enough in comparison to $\Omega_{0,0}$ such that $\cos(\Omega\tau/2) \simeq \sin(\Omega\tau/2) \simeq 1/\sqrt{2}$, $\delta/\Omega \ll 1$ and $\Omega \simeq \Omega_{0,0}$. The phase of the laser on the first pulse is used as a reference and set to zero. After the first pulse, the state of the ion is:

$$|\psi\rangle = -\frac{i}{\sqrt{2}}e^{-i\delta\tau/2}|e\rangle + \frac{1}{\sqrt{2}}e^{i\delta\tau/2}|g\rangle. \quad (208)$$

At this stage, the laser is turned off for a duration T . The ion evolves freely and the eigenstates $|e\rangle$ and $|g\rangle$ acquire a phase $e^{\pm i\omega_0 T/2}$. If the detuning of the laser is non-zero, the phase of the laser will be mismatched with that of the eigenstates by $e^{-i\delta T/2}$ for $|e\rangle$ and $e^{i\delta T/2}$ for $|g\rangle$. The new state of the ion is:

$$|\psi\rangle = -\frac{i}{\sqrt{2}}e^{-i\delta(\tau+T)/2}|e\rangle + \frac{1}{\sqrt{2}}e^{i\delta(\tau+T)/2}|g\rangle. \quad (209)$$

The second $\pi/2$ pulse is then applied but the initial phase ϕ of the laser may be different on this pulse than on the first one. A schematic of the pulse sequence is given in figure VIII.1. The final state is:

$$|\psi\rangle = -\frac{i}{2}(e^{-i\delta\tau}e^{-i\delta T/2} + e^{i\phi}e^{i\delta T/2})|e\rangle + \frac{1}{2}(e^{i\delta\tau}e^{i\delta T/2} - e^{-i\phi}e^{-i\delta T/2})|g\rangle. \quad (210)$$

The probability of excitation at the end of the Ramsey pulse sequence therefore is:

$$P_e(\delta, T, \phi) = \frac{1}{2}\left(1 + \cos(\delta(T + \tau) + \phi)\right). \quad (211)$$

It is apparent from this formula that oscillations in the probability of excitation will appear if δ , T or ϕ varies. Assuming $\phi = 0$, the probability of excitation will be maximum if $\delta(T + \tau)$ is a multiple of 2π and minimum if it is a multiple of π . In the latter case, the population which was promoted to the excited state by the first $\pi/2$

¹We first record Rabi oscillations by driving the carrier transition as explained in section VI.13 to measure the duration of a $\pi/2$ pulse.

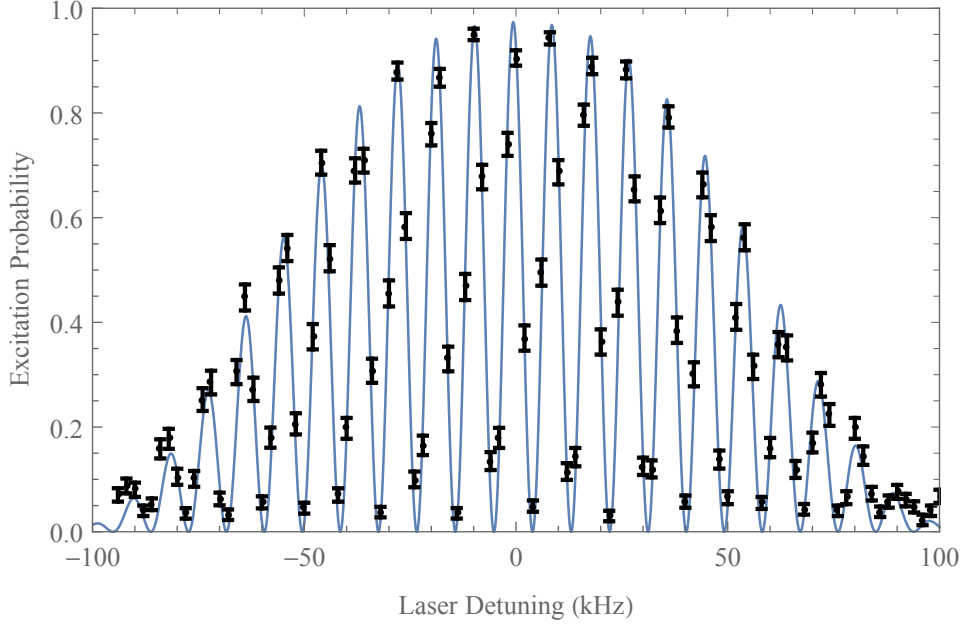


Figure VIII.2: Probability of excitation after the second $\pi/2$ pulse of a Ramsey experiment. The duration of the pulses is $9\text{ }\mu\text{s}$ and the delay between them is $100\text{ }\mu\text{s}$. The initial phase of the laser is the same for both pulses. The blue line is fitted to the data points using the full expression of the excitation probability (equation 212). The ion is initially cooled to the ground state at a high trapping frequency ($\nu_z = 414\text{ kHz}$).

pulse is transferred back to the ground state. For a fixed interval T , scanning the detuning of the laser gives rise to Ramsey fringes spaced by $2\pi/(T + \tau)$. Of course, if the detuning becomes large, equation 211 is not valid as the approximation $\delta/\Omega \ll 1$ does not hold any more. Without any assumption on the detuning, the probability of excitation is given by:

$$\begin{aligned}
 P_e(\delta, T, \phi) = & 2 \frac{\Omega_{0,0}^2}{\Omega^2} \sin^2 \left(\frac{\Omega\tau}{2} \right) \left[\cos^2 \left(\frac{\Omega\tau}{2} \right) + \frac{\delta^2}{\Omega^2} \sin^2 \left(\frac{\Omega\tau}{2} \right) \right. \\
 & + \left(\cos^2 \left(\frac{\Omega\tau}{2} \right) - \frac{\delta^2}{\Omega^2} \sin^2 \left(\frac{\Omega\tau}{2} \right) \right) \cos(\delta(T + \tau) + \phi) \\
 & \left. + 2 \frac{\delta}{\Omega} \cos \left(\frac{\Omega\tau}{2} \right) \sin \left(\frac{\Omega\tau}{2} \right) \sin(\delta(T + \tau) + \phi) \right].
 \end{aligned} \quad (212)$$

Figure VIII.2 shows the probability of excitation as a function of the detuning of the laser, describing the characteristic Ramsey fringes. The interval between the pulses, which are $9\text{ }\mu\text{s}$ long, is $100\text{ }\mu\text{s}$.

Let us now consider an experiment where the phase of the laser on the second pulse is scanned instead of the frequency. The phase on the first pulse is used as a reference and set to zero. The apparent detuning of the laser, which we assume small so that we can use equation 211, may change randomly between each repetition of a data point due to the linewidth of the laser and magnetic field noise in the trap. The detuning may also drift during an individual run of the Ramsey sequence. For a

Gaussian distribution of the detuning, centred on zero, the probability of excitation becomes:

$$P_e(\tau_d, T, \phi) = \frac{1}{2} \left(1 + \cos(\phi) \exp \left(-\frac{(T + \tau)^2}{2\tau_d^2} \right) \right) \quad (213)$$

where τ_d is the inverse of the standard deviation of the detuning distribution. For increasing intervals T , the amplitude of the sinusoidal term, which is equivalent to the visibility of the oscillations, decreases. The visibility is defined as:

$$V = \frac{\max(P_e) - \min(P_e)}{\max(P_e) + \min(P_e)}. \quad (214)$$

By measuring the visibility of the oscillations on phase scans with different intervals between the pulses, we can calculate τ_d which is related to the coherence time by $T_c = \sqrt{2}\tau_d$. Phase scans are a convenient technique to diagnose the coherence of the system as they are fast to acquire and their analysis is quite simple. In figure VIII.3, phase scans with different intervals ranging from 100 μ s to 2000 μ s show the loss in visibility over time. Note that the variable in the abscissa is the phase of the RF signal sent to the AOM, not that of the laser. Because the AOM is in double-pass configuration for this experiment, the phase shift undergone by the laser is twice that of the RF signal. Figure VIII.6 shows the visibility of the oscillations as a function of the interval. The data are fitted with a Gaussian function:

$$V(T) = V_0 \exp \left(-\frac{(T + \tau)^2}{2\tau_d^2} \right) \quad (215)$$

where the pre-factor V_0 corresponds to the visibility without delay between the pulses. V_0 may be smaller than one because of imperfect state preparation (imperfect cooling) and $\pi/2$ pulses (too short or too long, frequency offset) and fluorescence detection errors. Taking into account this pre-factor, the coherence time is given by:

$$T_c = \tau_d \sqrt{2(1 + \ln(V_0))}. \quad (216)$$

From the fit of the data points, we obtain $V_0 = 0.942(8)$ and $\tau_d = 1.28(5)$ ms which yields $T_c = 1.76(7)$ ms. This corresponds to a standard deviation of the detuning of 124(5) Hz. This figure can be used to put an upper limit on the linewidth of the laser². The coherence time due to dephasing measured through Ramsey experiments is three orders of magnitude smaller than the lifetime of the $D_{5/2}$ state which confirms that spin flip can be neglected.

²The values given here correspond to measurements made in November 2015. Measurements performed later on revealed a decrease in the coherence time to about 1 ms. It is unknown why. An increase in the laser linewidth is possible.

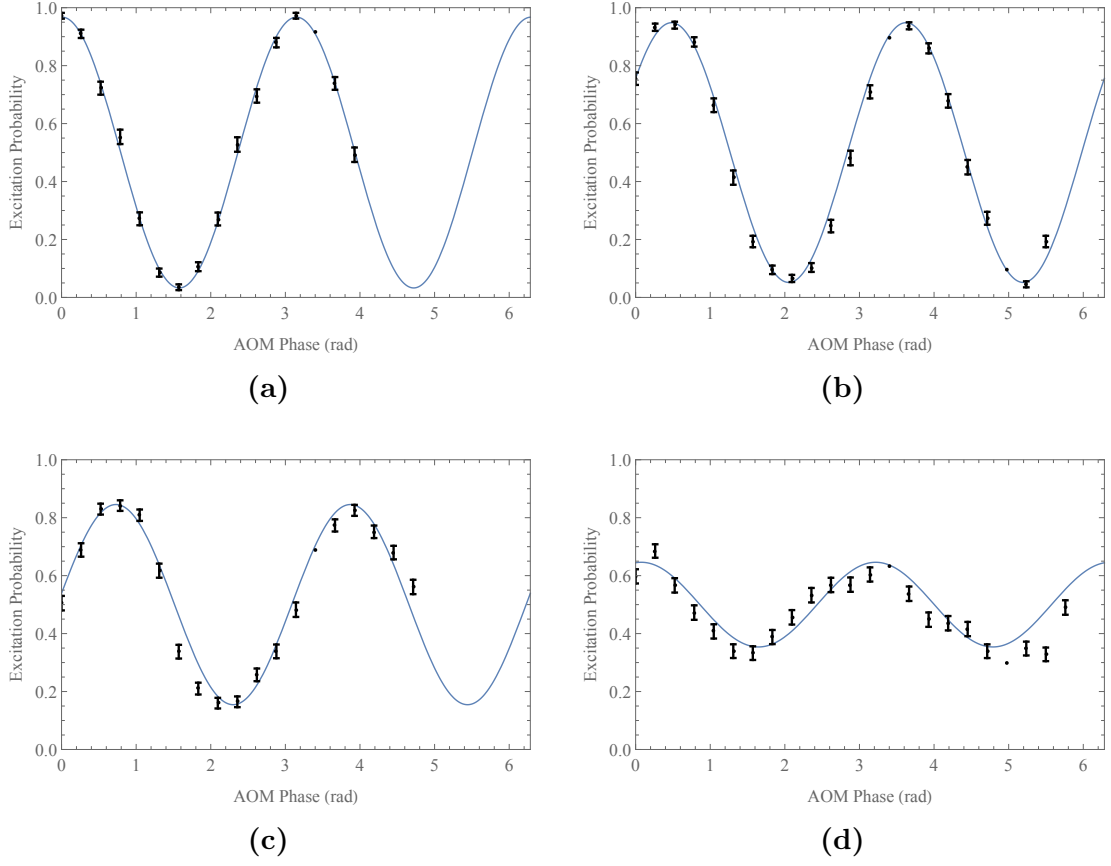


Figure VIII.3: Phase scans with delays of (a) 100 μs , (b) 500 μs , (c) 1000 μs and (d) 2000 μs between the $\pi/2$ pulses. The AOM is here in double pass configuration: the phase of the laser is twice that of the RF signal.

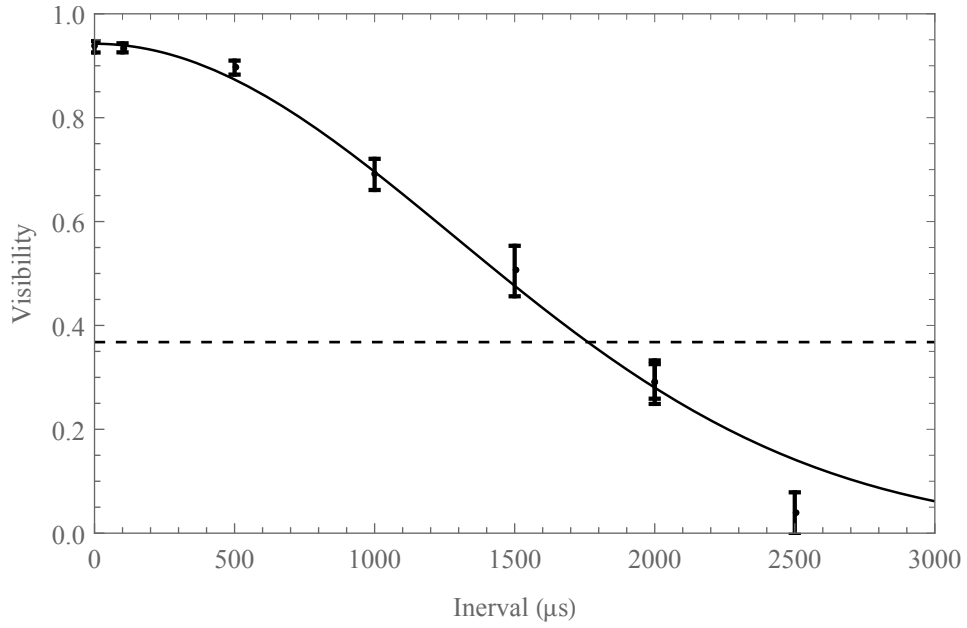


Figure VIII.4: Visibility of the phase scans as a function of the duration of the interval between the $\pi/2$ pulses. The data is fitted to a Gaussian decay, the coherence time is $T_c = 1.76(7)$ ms.

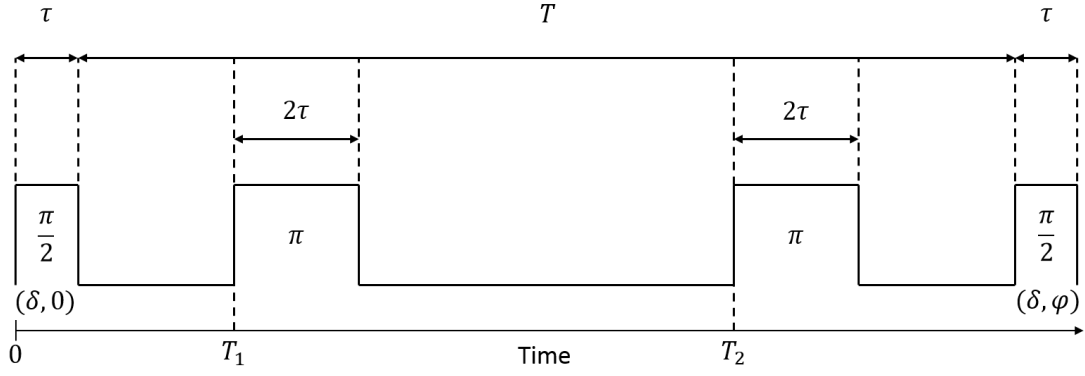


Figure VIII.5: Pulse sequence of a UDD sequence with two π pulses. The duration of a π pulse is twice that of a $\pi/2$ i.e. 2τ .

Dynamical decoupling

Dynamical decoupling is a technique to limit decoherence by decoupling the quantum system from the environmental noise [108, 109]. In the context of trapped ions, dynamical decoupling is implemented by applying a sequence of electromagnetic control pulses (typically in the microwave or optical domains) which averages out the effects of the environment. The simplest form of dynamical decoupling is the spin echo originally developed in the context of nuclear magnetic resonance (NMR) [110] where a single π pulse compensates for constant dephasing. More complex sequences involving several flip pulses include the Carr-Purcell-Meiboom-Gill (CPMG) [111, 112] and the Uhrig dynamical decoupling (UDD) scheme [113, 114] reviewed in ref. [115].

Dynamical decoupling was previously demonstrated in Penning and RF traps on microwave transitions [38, 116] and significant increases of the coherence time were observed. CPMG uses a sequence of periodic π pulses and is a direct extension of the spin echo technique while UDD employs an optimised sequence of non equidistant pulses able to suppress dephasing noise varying with time up to the $N - 1$ order using N pulses applied at times:

$$T_i = (T + \tau) \sin^2 \left(\frac{i\pi}{2(N+1)} \right) - \frac{\tau}{2} \quad , \quad i \in [1..N] \quad (217)$$

where T and τ have the same definitions as for the Ramsey experiment. The origin of time is taken at the beginning of the first $\pi/2$ pulse. As an example, the pulse sequence of a UDD experiment with two π pulses is shown in figure VIII.5. The phase of the laser is scanned on the second $\pi/2$ pulse to observe oscillations. The effect of the π pulses becomes clear when calculating the coefficients of $|e\rangle$ and $|g\rangle$ throughout the experiment. Let us consider a one π pulse sequence (spin echo); for simplicity, we assume that $\tau \ll T$ and $\delta \ll \Omega_0$ such that the pulses can be considered instantaneous and on resonance. Suppose that during the interval between the pulses, there is a

constant detuning δ between the laser and the atomic transition. After the first $\pi/2$ pulse, the quantum state of the ion is:

$$|\psi\rangle = -\frac{i}{\sqrt{2}}|e\rangle + \frac{1}{\sqrt{2}}|g\rangle \quad (218)$$

and after a delay of $T/2$, just before the π pulse, it is:

$$|\psi\rangle = -\frac{i}{\sqrt{2}}e^{-i\delta T/4}|e\rangle + \frac{1}{\sqrt{2}}e^{i\delta T/4}|g\rangle. \quad (219)$$

The π pulse swaps the population in $|e\rangle$ and $|g\rangle$:

$$|\psi\rangle = -\frac{i}{\sqrt{2}}e^{i\delta T/4}|e\rangle - \frac{1}{\sqrt{2}}e^{-i\delta T/4}|g\rangle. \quad (220)$$

Note that the signs in the exponential terms have changed. The second wait period of $T/2$ introduces an additional phase $\pm\delta T/4$ which cancels the one acquired during the first half of the experiment. After the second $\pi/2$ pulse, the probability of excitation therefore is:

$$P_e(\phi) = \frac{1}{2}(1 - \cos(\phi)). \quad (221)$$

There is no dependency on the detuning any more. Even if the detuning is different (but constant) for each repetition of a data point, the visibility of $P_e(\phi)$ will be one. Similarly, a linearly varying detuning $\delta = \alpha t$ can be compensated by a two π pulse sequence. The first π pulse is applied at $T_1 = T/4$ at which point the phase acquired is:

$$\pm \frac{1}{2} \int_0^{T_1} \alpha t dt = \pm \frac{\alpha T^2}{64}. \quad (222)$$

Between the first pulse at T_1 and the second pulse at $T_2 = 3T/4$ and between T_2 and T , the respective additional phases are $\pm\alpha T^2/8$ and $\pm 7\alpha T^2/64$. In total, for the $|e\rangle$ eigenstate, the phase due to the intervals between the pulses is:

$$-\frac{\alpha T^2}{64} + \frac{\alpha T^2}{8} - \frac{7\alpha T^2}{64} = 0. \quad (223)$$

The probability of excitation after the second $\pi/2$ pulse thus remains independent of the detuning. This reasoning can be extended to sequences with any number of π pulses. In practice, the time evolution of the detuning is not described by a simple polynomial. Nevertheless, for a given time T , we observe an increase in the visibility of the oscillations on a phase scan when UDD is used compared to a simple Ramsey experiment. This can be seen in figure VIII.6 which shows the measured visibilities for different intervals for UDD sequences with one, two and three π pulses. The

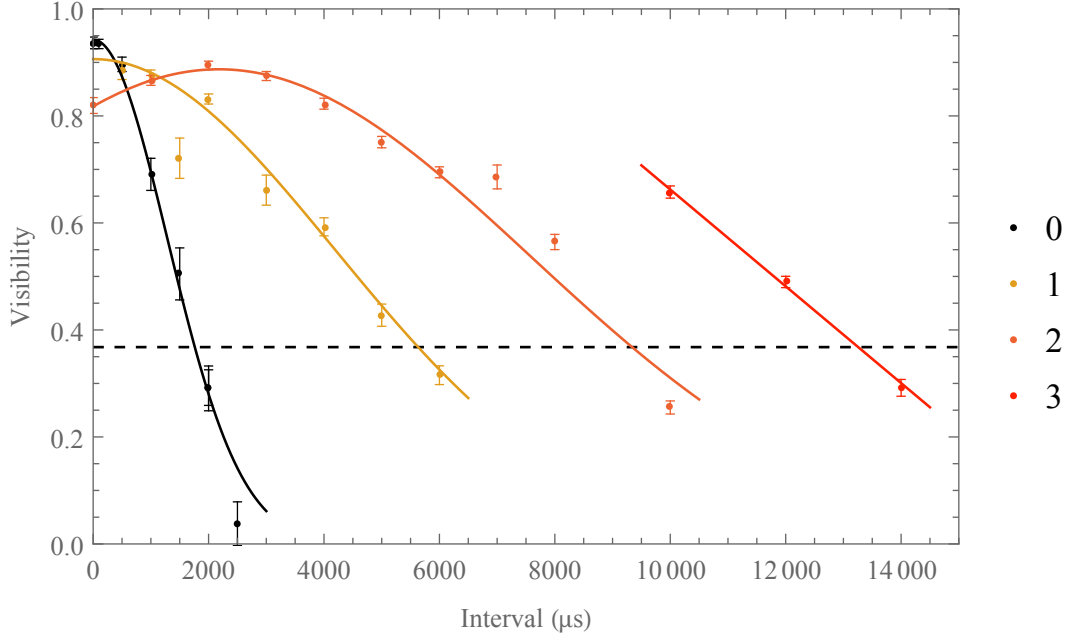


Figure VIII.6: Visibility of the phase scans as a function of the duration of the interval between the $\pi/2$ pulses and the number of π pulses in the UDD sequence.

data points from Ramsey experiments are also included as a reference. We notice that for very short intervals, the visibility is lower when UDD is employed than for the Ramsey experiment. This is because the detrimental effect of imperfect π pulses exceeds the benefits of dynamical decoupling. As for the Ramsey experiment, the data points are fitted with Gaussian functions to measure the coherence time except for the 3 π pulse sequence where a linear fit is used because it has only three data points. The coherence time as a function of the number of pulses is plotted in figure VIII.7 and a linear increase of 3.83(2) ms per pulse is found. This shows that dynamical decoupling is a powerful way to extend the lifetime of a qubit.

A coherence time exceeding ten minutes was recently reported for the microwave qubit of a single $^{171}\text{Yb}^+$ sympathetically cooled by a $^{138}\text{Ba}^+$ ion in a Paul trap thanks to dynamical decoupling [117].

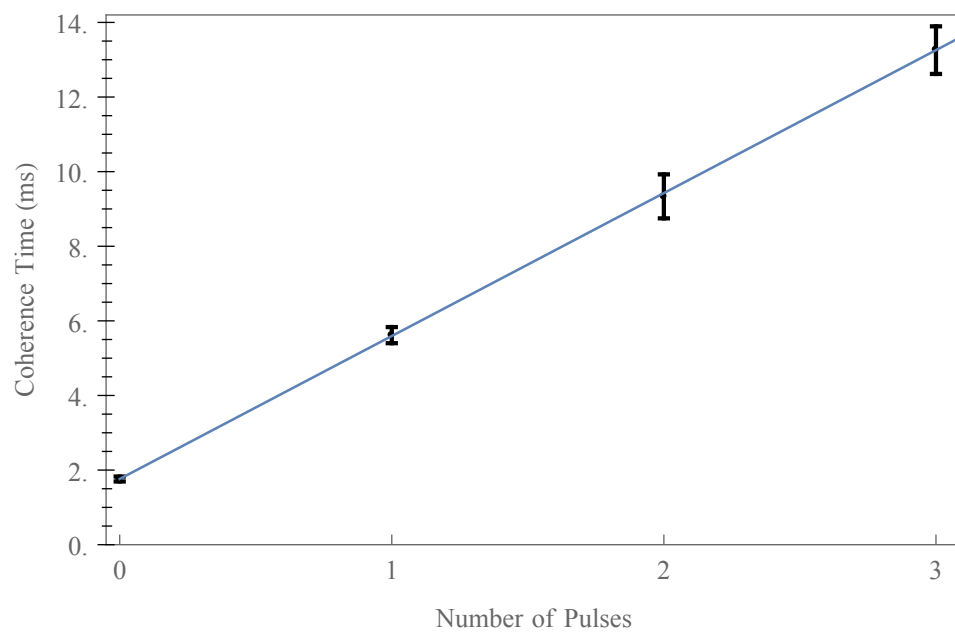


Figure VIII.7: Coherence time ($1/e$) as a function of the number of pulses in the UDD sequence. A linear fit indicates an increase in the coherence time of $3.83(2)$ ms per pulse.

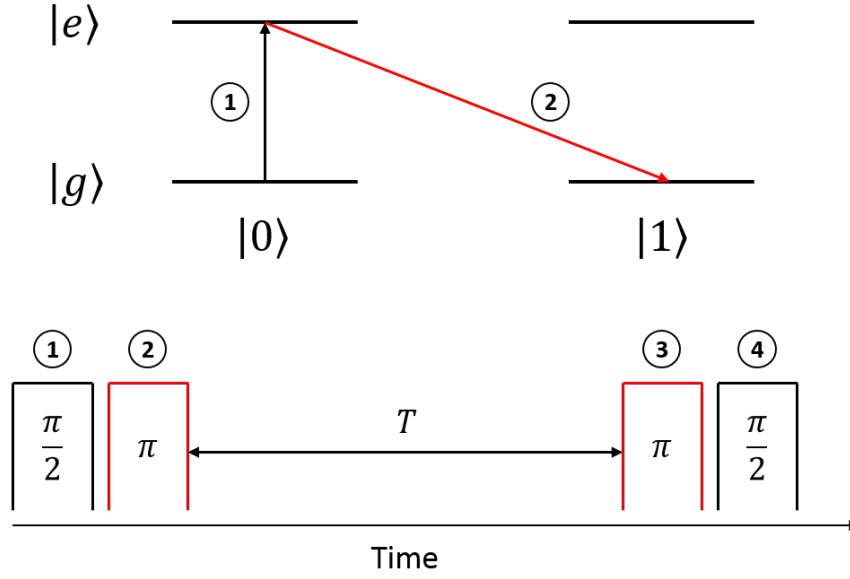


Figure VIII.8: Pulse sequence to create a superposition of $|g, 0\rangle$ and $|g, 1\rangle$ and perform a Ramsey experiment. Black rectangles indicate carrier pulses (1 and 4), red is for the first red sideband (2 and 3).

VIII.2 Motional Coherence

A coherent superposition of motional states can be created by applying resonant laser pulses on different sidebands. In this section, we explain the methods to create these superpositions and use them to investigate the coherence time of the axial motion of a single ion. Earlier work on the coherence of the motional states of trapped ions carried out at NIST can be found in ref. [118].

Superpositions near the ground state

Starting with an ion cooled to the ground state, we create a superposition of $|g, 0\rangle$ and $|g, 1\rangle$ by performing a $\pi/2$ pulse on the carrier followed by a π pulse on the first red sideband. The superposition is undone with a mirrored pulse sequence: π pulse on the first red sideband followed by $\pi/2$ pulse on the carrier. A delay of duration T is inserted between the first and last two pulses (see figure VIII.8). This is equivalent to a Ramsey experiment for the motional states.

Let us look at the evolution of the system described by the wave function $|\psi\rangle$, starting with $|\psi\rangle = |g, 0\rangle$. Given the parameters of this experiment, three states may be populated: $|e, 0\rangle$, $|g, 0\rangle$ and $|g, 1\rangle$. $|\psi(t)\rangle$ can thus be written as a superposition of these states:

$$|\psi(t)\rangle = c_{e,0}(t) |e, 0\rangle + c_{g,0}(t) |g, 0\rangle + c_{g,1}(t) |g, 1\rangle. \quad (224)$$

Assuming the pulses are resonant, that is the laser has no detuning with respect to the targeted sideband, the propagator describing the coupling between $|e, n+s\rangle$ and $|g, n\rangle$ is

$$T_n^s = \begin{pmatrix} \cos(\Omega_{n,n+s}t/2) & -ie^{i(\phi+|s|\pi/2)} \sin(\Omega_{n,n+s}t/2) \\ -ie^{-i(\phi+|s|\pi/2)} \sin(\Omega_{n,n+s}t/2) & \cos(\Omega_{n,n+s}t/2) \end{pmatrix}. \quad (225)$$

The first $\pi/2$ pulse on the carrier, with a duration $\tau = \pi/(2\Omega_{0,0})$ transfers half of the population from $|g, 0\rangle$ to $|e, 0\rangle$. Setting the phase of the laser to zero on this pulse, the propagator is

$$T_0^0 = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & -i \\ -i & 1 \end{pmatrix} \quad (226)$$

and the wave function after the pulse is:

$$|\psi\rangle = -\frac{i}{\sqrt{2}} |e, 0\rangle + \frac{1}{\sqrt{2}} |g, 0\rangle. \quad (227)$$

The following π pulse on the red sideband transfers all the population in $|e, 0\rangle$ to $|g, 1\rangle$. The population in $|g, 0\rangle$ is not affected by that pulse. The propagator governing this transition is:

$$T_1^{-1} = \begin{pmatrix} 0 & e^{i\phi_2} \\ -e^{-i\phi_2} & 0 \end{pmatrix} \quad (228)$$

where ϕ_2 is the initial phase of the laser on that pulse. The wave function becomes:

$$|\psi\rangle = \frac{1}{\sqrt{2}} |g, 0\rangle + \frac{i}{\sqrt{2}} e^{-i\phi_2} |g, 1\rangle. \quad (229)$$

A superposition of two states of the quantum harmonic oscillator has been created. The lasers are then blocked so that the ion evolves freely. Meanwhile the 729 nm laser is set to the carrier frequency during the wait time. After a time T , the population in $|g, 1\rangle$ has acquired an additional phase $\omega_z T$ and the wave function becomes:

$$|\psi\rangle = \frac{1}{\sqrt{2}} |g, 0\rangle + \frac{i}{\sqrt{2}} e^{-i(\phi_2+\omega_z T)} |g, 1\rangle. \quad (230)$$

A second pulse on the red sideband, with initial phase ϕ_3 is applied giving:

$$|\psi\rangle = \frac{i}{\sqrt{2}} e^{i(\phi_3-\phi_2-\omega_z T)} |e, 0\rangle + \frac{1}{\sqrt{2}} |g, 0\rangle \quad (231)$$

and after a last $\pi/2$ pulse on the carrier (phase ϕ_4), the final wave function is:

$$|\psi\rangle = \frac{i}{2} \left(e^{i(\phi_3 - \phi_2 - \omega_z T)} - e^{i\phi_4} \right) |e, 0\rangle + \frac{1}{2} \left(1 + e^{i(\phi_3 - \phi_2 - \phi_4 - \omega_z T)} \right) |g, 0\rangle. \quad (232)$$

The probability of the ion to be in the excited state, which is the quantity measured experimentally, therefore is:

$$P_e(T) = \frac{1}{2} (1 - \cos(\omega_z T + \Phi)) \quad (233)$$

where Φ is a phase term given by $\Phi = \phi_2 + \phi_4 - \phi_3$. The values of the initial phases depend on the duration of the pulses³. Because of the limited coherence time of the laser, for long intervals, there is no fixed relation between ϕ_2 and ϕ_3 but $\phi_2 + \phi_4 - \phi_3$ is constant⁴. Oscillations in the probability of excitation can be obtained either by scanning the phase of the laser on the last pulse (ϕ_4) or the delay T .

If the carrier pulses are replaced with pulses on the first blue sideband, a superposition of $|g, 0\rangle$ and $|g, 2\rangle$ is formed instead. In this case, the probability of excitation becomes:

$$P_e(T) = \frac{1}{2} (1 - \cos(2\omega_z T + \Phi)). \quad (234)$$

In the following, we shall refer to a superposition of $|g, n\rangle$ and $|g, m\rangle$ as a n - m superposition. Two possible sources of decoherence can be identified. First, electrical noise in the trap's electrodes leads to instabilities in ω_z which in turn provokes a randomisation of the argument in the cosine function in the expression for P_e . This causes the visibility of the oscillations to decrease with time the same way the unstable detuning of the laser did for the optical qubit. Assuming the axial frequency has a normal distribution centred around some value $\omega_{z,0}$ with a standard deviation σ , the probability of excitation at the end of the Ramsey sequence for a n - m superposition is:

$$P_e(T) = \frac{1}{2} (1 - \cos(\Delta n \omega_{z,0} T + \Phi) \exp(-(\Delta n)^2 \sigma^2 T^2 / 2)); \quad (235)$$

with $\Delta n = |n - m|$. It suggests the coherence time should decrease for larger separations of the Fock states in the superposition. The electric noise cannot be directly measured on the electrodes of the trap as they are inaccessible. We can however measure the output of the trap supply. We do so with a high precision voltmeter and convert the voltages into frequencies with equation 10. Figure VIII.9 shows the normalised histogram of the axial angular frequency from 352 measurements taken

³The output of a direct digital synthesiser is phase continuous [119] i.e. there is no discontinuity in the signal when switching between profiles with different frequencies unless a phase offset is specified on the second profile.

⁴ ϕ_3 and ϕ_4 are functions of T but in their difference, the term depending on T drops.

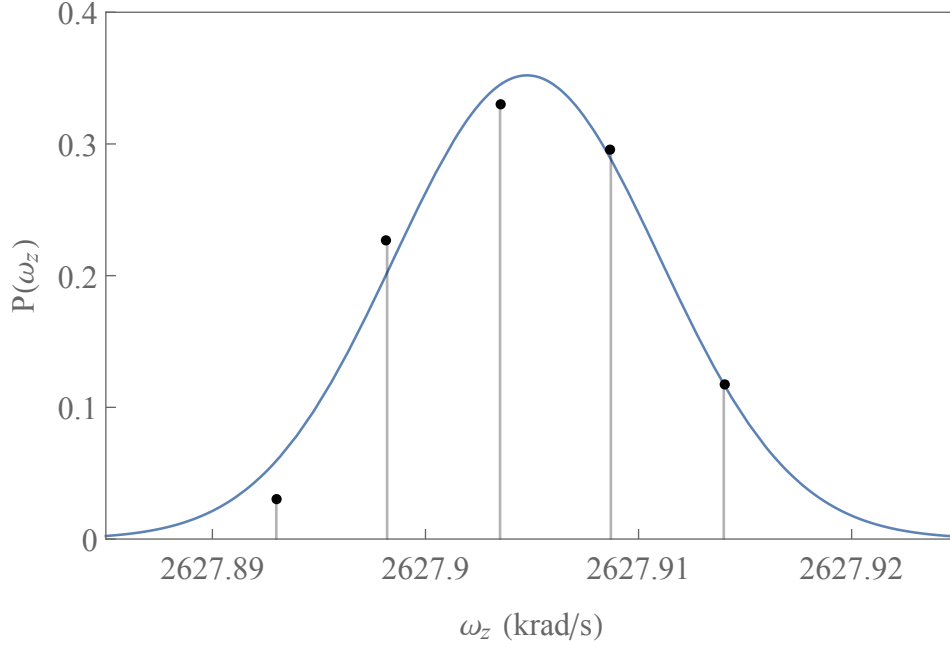


Figure VIII.9: Normalised histogram of the axial angular frequency calculated the trap supply output voltage. The Gaussian fit (blue line) gives a standard deviation $\sigma = 6.2(5) \text{ rad s}^{-1}$.

over one hour. The data points are fitted to a Gaussian, the standard deviation of the angular frequency is found to be $\sigma = 6.2(5) \text{ rad s}^{-1}$ ($0.99(8) \text{ Hz}$); the corresponding $1/e$ coherence times are $228(18) \text{ ms}$ for $\Delta n = 1$ and $114(9) \text{ ms}$ for $\Delta n = 2$. Using the nomenclature of ref. [118], decoherence due to the unstable trapping frequency corresponds to the interaction of the ion with a *phase* reservoir. The other source of decoherence is heating. If the delay between the pulses is long enough, it is possible for the ion to experience a significant heating which is an interaction with the environment and therefore leads to decoherence. The evolution of the visibility V with time under anomalous heating can be modelled by [118]:

$$V(t) = \frac{1}{(1 + \bar{n}(t))^{1+\Delta n}}. \quad (236)$$

We assume a linear increase in the average phonon number with time: $\bar{n}(t) = n_0 + \gamma t$. The coherence time is given by:

$$T_c = \frac{1}{\gamma} \left(\exp \left(\frac{1}{1 + \Delta n} \right) - 1 - n_0 \right), \quad (237)$$

it decreases with larger heating rates and Fock state separation Δn .

We perform two sets of experiments: one at low trapping voltage (22 V , $\nu_z = 125 \text{ kHz}$) and one at high voltage (250 V , $\nu_z = 420 \text{ kHz}$). To prepare the ion in the ground state at low voltage, adiabatic cooling is used. In both cases Ramsey experiments are carried out with 0-1 and 0-2 superpositions. For a 0-2 superposition at

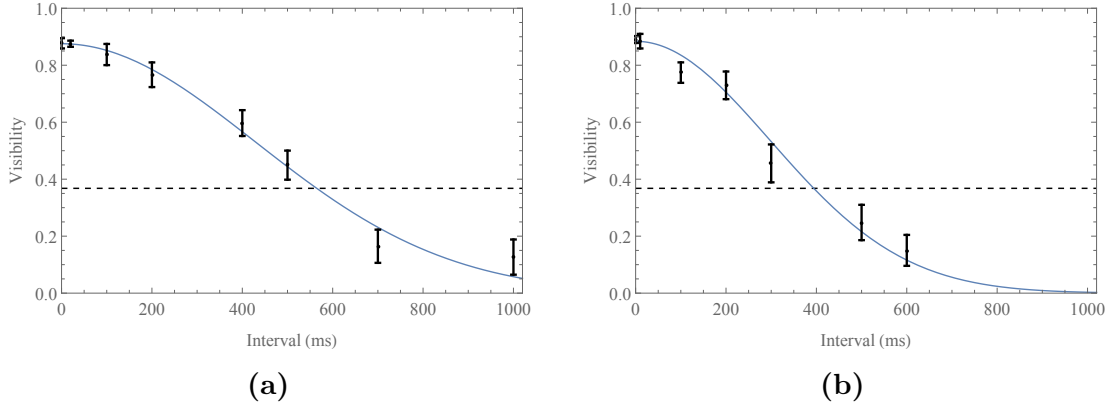


Figure VIII.10: Visibility of phase scans (at $\nu_z = 420$ kHz) as a function of the delay T in the Ramsey sequence for **(a)** a superposition of $|g, 0\rangle$ and $|g, 1\rangle$ and **(b)** a superposition of $|g, 0\rangle$ and $|g, 2\rangle$. The dashed lines mark the $1/e$ point where the coherence time is measured. The data points are fitted to a Gaussian following equation 235, the coherence times are $T_c^1 = 565(21)$ ms for $\Delta n = 1$ and $T_c^2 = 394(25)$ ms for $\Delta n = 2$.

420 kHz, a sampling frequency of 1.68 MHz is necessary to observe the oscillations of $P_e(T)$ without aliasing. At the time the experiment was realised, the maximum sampling frequency of the FPGA was 1.56 MHz. Consequently, we perform phase scans with fixed delays instead. The visibility for different intervals T is plotted in figure VIII.10. The data points are well approximated by a Gaussian function of time, consistent with equation 235 and thus suggesting that decoherence is due to frequency instabilities. The fits yield coherence times $T_c^1 = 565(21)$ ms for the superposition of $|g, 0\rangle$ and $|g, 1\rangle$ and $T_c^2 = 394(25)$ ms for $|g, 0\rangle$ and $|g, 2\rangle$. Both these values are larger than what was expected from the measurement of the noise on the trap supply and there is a deviation from the theoretical model which predicts that T_c^1 should be twice as large as T_c^2 whereas we find experimentally $T_c^1/T_c^2 \simeq 1.43$. This discrepancy can be due to the fact that decoherence from heating is not taken into account here. At 250 V the heating rate is quite low (around 0.3 s^{-1}) and therefore should not be the most important source of decoherence but may still be significant. The noise level on the electrodes may also differ between the two experiments (which were carried out on different days). The coherence times correspond to a standard deviation of the axial frequency of $0.37(1)$ Hz for the 0-1 superposition and $0.27(2)$ Hz for the 0-2 superposition.

For the experiments at low voltage, we record the probability of excitation as a function of the duration of the interval T . We measure the visibility of the oscillations on intervals between T and $T + \Delta T$ where T takes values between 0 and 200 ms and ΔT is just long enough to observe a few oscillations of P_e . Examples are given in figure VIII.11; as expected, the frequency of the oscillations is twice as large ($2\omega_z$) for a 0-2 superposition. A decrease in visibility can be seen between the scans taken with no initial delay (figure VIII.11.a and b) and those where T starts at 50 ms (figure VIII.11.c and d). Unlike the high voltage case, the visibility

as a function of time, shown in figure VIII.12, does not follow a Gaussian decay but rather the model of equation 236 which can be understood by the fact that the heating rate is higher at low frequency. We measured $9.6(1) \text{ s}^{-1}$ at 20 V (see section VI.1) which would correspond to a coherence time of $68(1) \text{ ms}$ (assuming $n_0 = 0$) for a 0-1 superposition which is significantly smaller than the coherence time limited by dephasing measured at high voltage. The visibilities in figure VIII.12 are normalised: for each scan with an initial delay $T > 0$, a scan with no initial delay is recorded simultaneously and the ratio of the visibility of the former over the latter is used as a data point. The normalised visibility at $T = 0$ is therefore one and we can fit the data with:

$$V(T) = \frac{1}{(1 + \gamma t)^{1+\Delta n}}. \quad (238)$$

We obtain coherence times $T_c^1 = 77(2) \text{ ms}$ for the 0-1 superposition and $T_c^2 = 59(3) \text{ ms}$ for the 0-2 superposition which respectively correspond to heating rates of $8.5(3) \text{ s}^{-1}$ and $6.7(3) \text{ s}^{-1}$. These figures are in fairly good agreement with the heating rate measured at 20 V. As for the data at high voltage, only one source of decoherence is considered here which can explain discrepancies and it is also possible that the heating rate varies from day to day.

The Ramsey experiments reveal two regimes: at high voltage, the motional coherence is limited by dephasing, most probably due to instabilities in the trapping frequency originating from electrical noise on the electrodes of the trap whereas decoherence at low voltage can be attributed chiefly to a high heating rate. Overall, both at high and low voltage, we find that the coherence time does not scale as strongly with the separation of the Fock states Δn as was predicted by the theoretical models but this may be because the different sources of decoherence are not correctly taken into account. To further study the motional coherence of a trapped ion, a better knowledge of these sources would be useful. Noise can be artificially added on the electrodes in a controlled manner to engineer the dephasing. The influence of heating on the coherence time could then be better characterised by performing Ramsey experiments at different trapping frequencies and systematically measuring the heating rates.

Phonon echo

In a manner similar to the spin echo technique for the optical qubit, the dephasing in a superposition of motional states can be partially compensated by applying a “ π ” pulse midway through the wait time of a Ramsey experiment. In this context, by π pulse we mean swapping the populations in the two Fock states of the superposition which is done with three laser pulses. By analogy with the spin echo, we refer to this technique as “phonon echo”. For a 0-2 superposition, the population swapping is

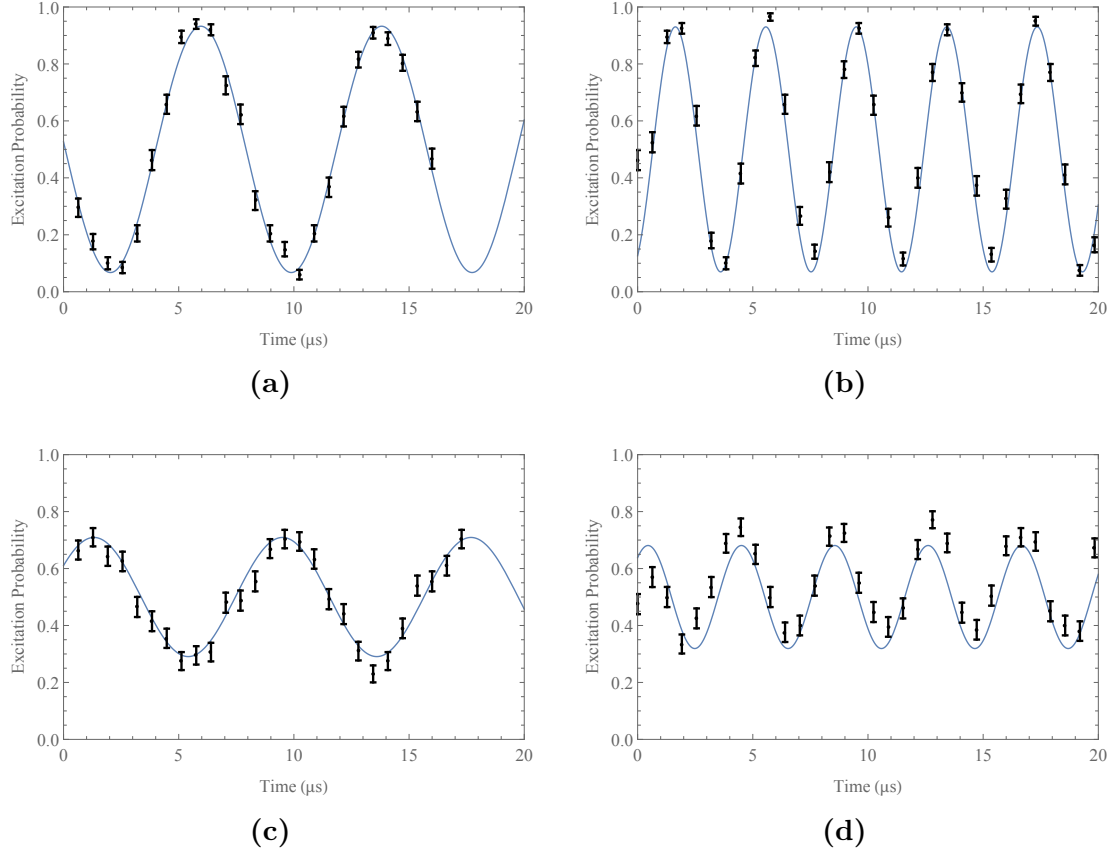


Figure VIII.11: Probability of excitation as a function of the delay between the first two and last two pulses for (a) a 0-1 superposition of and (b) a 0-2 superposition of. With an initial fixed delay of 50 ms for (c) a 0-1 superposition and (d) a 0-2 superposition.

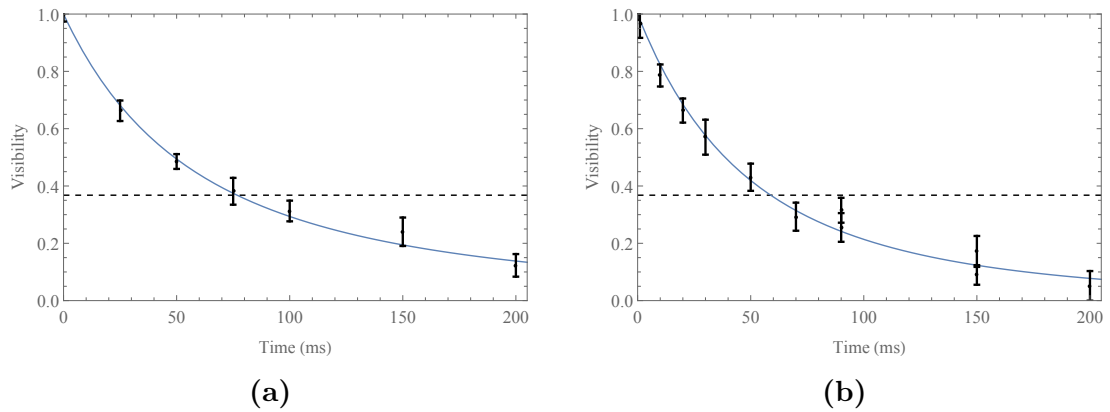


Figure VIII.12: Normalised visibility as a function of time (at $\nu_z = 125$ kHz) for (a) a 0-1 superposition and (b) a 0-2 superposition. The visibility is measured over small ranges (around 20 ms) considered punctual. The data points are fitted with equation 236; the coherence times are $T_c^1 = 77(2)$ ms for $\Delta n = 1$ and $T_c^2 = 59(3)$ ms for $\Delta n = 2$.

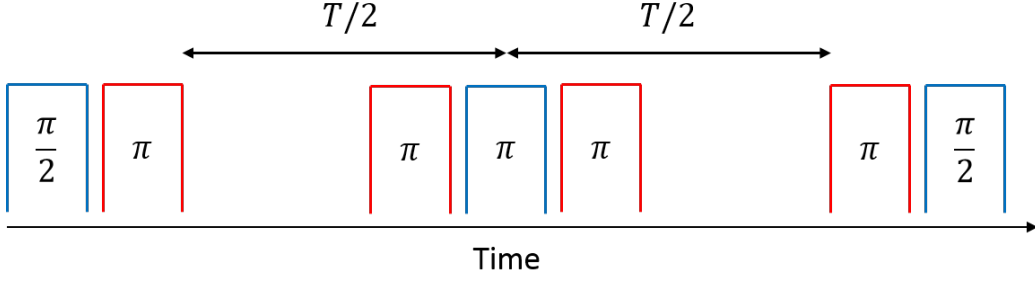


Figure VIII.13: Pulse sequence for a phonon echo with a superposition of $|g, 0\rangle$ and $|g, 2\rangle$. Pulses are applied on the first blue sideband (blue) and first red sideband (red).

performed by applying a π pulse on the first blue sideband flanked by two π pulses on the first red sideband. Let us consider the pulse sequence shown in figure VIII.13. The initial phase of the first pulse is set to zero and we denote ϕ_2 the phase of the second pulse, ϕ_3 the phase of the third pulse, etc. We assume that the trapping frequency ω_z is constant throughout the sequence. The first two pulses create a superposition of $|g, 0\rangle$ and $|g, 2\rangle$:

$$|\psi\rangle = \frac{1}{\sqrt{2}} |g, 0\rangle - \frac{1}{\sqrt{2}} e^{-i\phi_2} |g, 2\rangle \quad (239)$$

and after a delay of $T/2$ when the laser is blocked, the state becomes:

$$|\psi\rangle = \frac{1}{\sqrt{2}} |g, 0\rangle - \frac{1}{\sqrt{2}} e^{-i(\phi_2 + \omega_z T)} |g, 2\rangle. \quad (240)$$

A π pulse on the red sideband then transfers the population in $|g, 2\rangle$ to $|e, 1\rangle$ and a π pulse on the blue sideband swaps the population of $|g, 0\rangle$ and $|e, 1\rangle$. A second π pulse on the red sideband transfers the population in $|e, 1\rangle$ back to $|g, 2\rangle$ so that the state of the ion is:

$$|\psi\rangle = \frac{1}{\sqrt{2}} e^{i(\phi_3 - \phi_2 - \phi_4 - \omega_z T)} |g, 0\rangle - \frac{1}{\sqrt{2}} e^{i(\phi_4 - \phi_5)} |g, 2\rangle. \quad (241)$$

After the second wait period, we have:

$$|\psi\rangle = \frac{1}{\sqrt{2}} e^{i(\phi_3 - \phi_2 - \phi_4 - \omega_z T)} |g, 0\rangle - \frac{1}{\sqrt{2}} e^{i(\phi_4 - \phi_5 - \omega_z T)} |g, 2\rangle \quad (242)$$

and the probability of excitation after the last two pulses is:

$$P_e = \frac{1}{2} (1 - \cos(\phi_7 - \phi_6 + \phi_5 - 2\phi_4 + \phi_3 - \phi_2)). \quad (243)$$

Note that this expression does not depend on the time T nor the frequency ω_z . This means that even if the frequency ω_z is slightly different for each repetition of a data point, the visibility on a phase scan will not be affected whereas it would result in

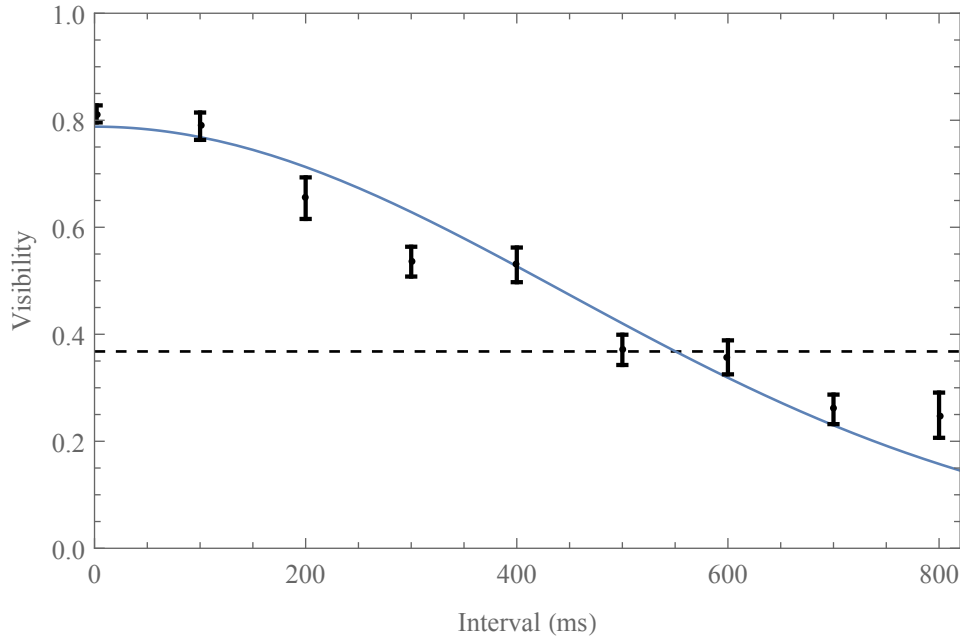


Figure VIII.14: Visibility of phase scans as a function of the total delay T of a phonon echo on a 0-2 superposition. The coherence time is 550(38) ms.

a loss of contrast in a standard Ramsey experiment. Again, this assumes that ω_z is constant for the duration of a repetition which is not the case in reality. Figure VIII.14 shows the visibility of phase scans in phonon echo sequences with different wait times, carried out at high voltage (250 V) on a 0-2 superposition. The data is fitted following the same procedure as for the Ramsey experiment, considering only dephasing. The $1/e$ coherence time is 550(38) ms which is substantially higher than what was found without the phonon echo (394(25) ms). We note however a lower initial visibility, probably because of imperfections in the additional pulses.

Superposition of three states

The coherent superposition of motional states is not restricted to two states. We can prepare a superposition of the three lowest motional states with the following method: starting from $|g, 0\rangle$, a pulse on the carrier transfers two thirds of the population to $|e, 0\rangle$. The duration of such a pulse is $\tau = 2 \arcsin(\sqrt{2/3})/\Omega_{0,0}$. A $\pi/2$ pulse on the first red sideband then takes half the population in $|e, 0\rangle$ to $|g, 1\rangle$. The population remaining in $|e, 0\rangle$ is transferred to $|g, 2\rangle$ with a π pulse on the second red sideband without affecting $|g, 0\rangle$ and $|g, 1\rangle$. A superposition of $|g, 0\rangle$, $|g, 1\rangle$ and $|g, 2\rangle$ with one third of the population in each state is created. As for the superposition of two states, a pulse sequence in the reverse order preceded by a wait time allows interferences to be observed by realising a Ramsey experiment (see table VIII.1). The probability of excitation as a function of the wait time is given

Sideband	Pulse time
Carrier	$\frac{1}{\Omega_{0,0}} \arcsin(\sqrt{2/3})$
1 st RSB	$\frac{\pi}{2\Omega_{1,0}}$
2 nd RSB	$\frac{\pi}{\Omega_{2,-1}}$
None (delay)	T
2 nd RSB	$\frac{\pi}{\Omega_{2,-1}}$
1 st RSB	$\frac{\pi}{2\Omega_{1,0}}$
Carrier	$\frac{1}{\Omega_{0,0}} \arcsin(\sqrt{2/3})$

Table VIII.1: Pulse sequence for a Ramsey experiment with a superposition of $|g, 0\rangle$, $|g, 1\rangle$ and $|g, 2\rangle$.

by (derivation in appendix A.4):

$$P_e(T) \simeq \frac{1}{9} \left(5 - \cos(\omega_z T + \Phi_2 - \Phi_1) - 2 \cos(\omega_z T + \Phi_1) - 2 \cos(2\omega_z T + \Phi_2) \right). \quad (244)$$

where Φ_1 and Φ_2 are constants depending on the phase of the laser on the different pulses. This probability is the sum of the probabilities of the ion being in $|e, 0\rangle$ and $|e, 1\rangle$ and is calculated under the approximation that the coupling strength for the carrier transition is the same for the phonon states $|0\rangle$ and $|1\rangle$ ($\Omega_{0,0} \simeq \Omega_{1,1}$). Because this scheme uses the second red sideband, it is necessary to operate at a low trapping frequency in order to have an appreciable coupling strength. We perform the experiments at $\nu_z = 125$ Hz and the ion is initially cooled to the ground state at high frequency before adiabatically reducing the trapping voltage. Figure VIII.15 shows the measured probability of excitation as a function of the wait time. The two frequency components in equation 244 are at the origin of an interference pattern reminiscent of the three-slit diffraction of a wave. Like for the superposition of two states, the visibility of the oscillations can be used to measure decoherence. When fitting the scans, we allow the components at ω_z and $2\omega_z$ to have different amplitudes so that we obtain a visibility for each component. The evolution of the visibility with the duration of the interval T is shown in figure VIII.16. There is no clear difference in the loss of visibility between the two frequency components and the overall coherence time is quite short, around 40 ms. At this trapping voltage, we expect decoherence to be dominated by heating.

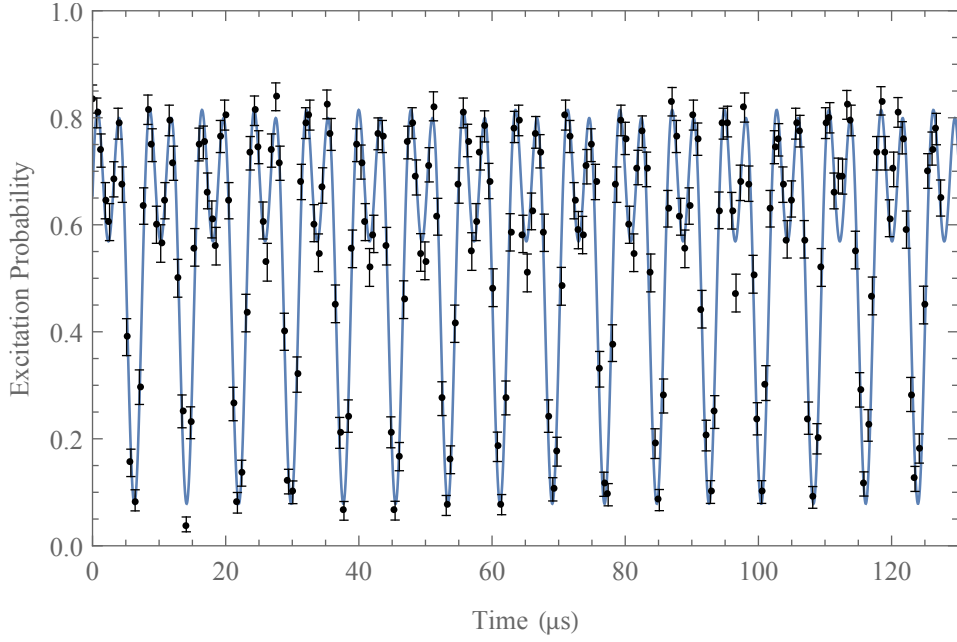


Figure VIII.15: Probability of the ion to be in the excited state as a function of the delay in a Ramsey experiment with a superposition of $|g, 0\rangle$, $|g, 1\rangle$ and $|g, 2\rangle$. The trapping frequency is 125 kHz.

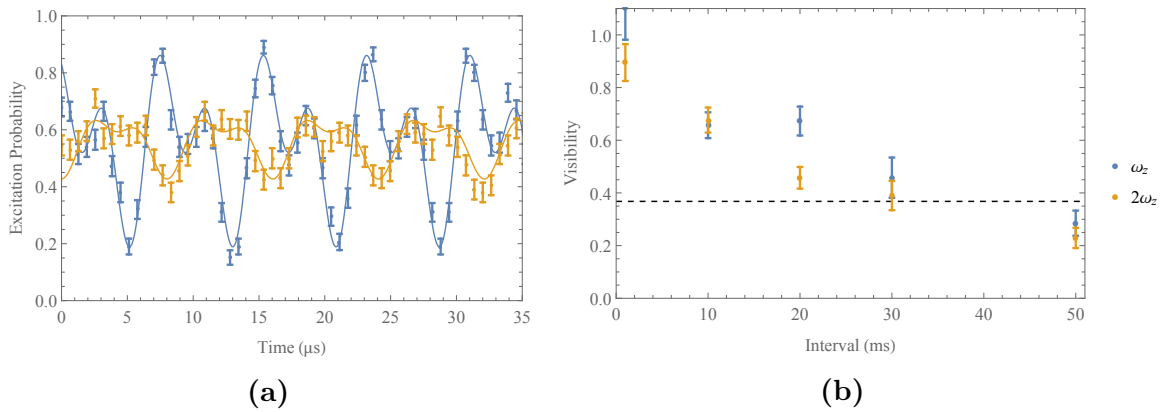


Figure VIII.16: (a) Ramsey experiment with a superposition of $|g, 0\rangle$, $|g, 1\rangle$ and $|g, 2\rangle$ with an initial delay of 1 ms (in blue) and 50 ms (in yellow) showing the loss of visibility. Normalised visibility as a function of time for the ω_z component (in blue) and the $2\omega_z$ component.

VIII.3 Coherent Manipulations in High Phonon States

This section concerns the preparation of non-thermal states well outside the Lamb-Dicke regime and coherent manipulations with these states. Being outside the Lamb-Dicke regime gives the possibility to address high order sidebands to create superpositions of motional states with large separations.

Sideband heating

Sideband heating (SBH) is a technique named in analogy with sideband cooling which consists in increasing the energy of the ion's motion by driving a blue sideband with the 729 nm laser (and with the 854 nm laser on). Sideband heating follows the same principles as sideband cooling, which have been already discussed, and is performed experimentally in a similar manner, the only major difference being of course the frequency of the laser.

Starting from a ground state cooled ion, sideband heating on the first blue sideband will increase the phonon number until the first zero coupling point is reached. The population is then distributed close to this zero coupling point giving rise to a characteristic spectrum where the first pair of sidebands (red and blue) are very low in amplitude and higher order sidebands high, consistent with the Rabi coupling strengths at this point. Figure VIII.17 shows the spectrum of a sideband heated ion where the first blue sideband was driven for 10 ms after an initial stage of Doppler and sideband cooling to the ground state. At the trapping frequency of 420 kHz used in this example, the lowest zero coupling point of the first blue sideband is located around $n = 163$. The spectrum is fitted assuming a Gaussian distribution of motional states, we obtain an average phonon number $\bar{n} = 146(2)$ with a standard deviation $\sigma = 14(1)$.

By addressing a higher order sideband, the ion can be further heated to higher motional states and the population will eventually be distributed next to the zero-coupling point of this sideband. There is *a priori* no limit to how high the phonon number can be taken by exciting different blue sidebands in a multiple-stage process. The spectrum in figure VIII.18 was obtained after 10 ms of heating on the first blue sideband followed by 20 ms on the second blue sideband. We find an average phonon number $\bar{n} = 294(2)$ and a standard deviation $\sigma = 25(3)$ (the zero point is at $n = 293$). The spread of the distribution is not negligible and increases for longer heating times. Nevertheless, it is possible to drive the ion coherently on certain sidebands and observe Rabi oscillations owing to limited variations in their coupling strengths over the range of populated motional states. For instance, the coupling strength of the fourth red sideband as a function of the phonon number around the zero coupling point of the second blue sideband is plotted in figure VIII.19. The probability distribution of the motional state after two-stage SBH as found from the

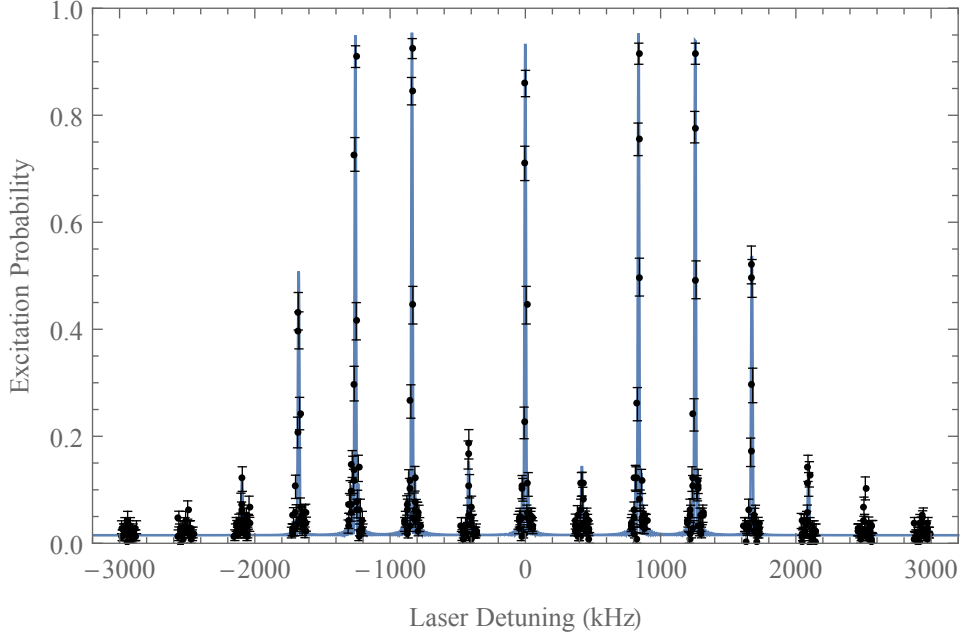


Figure VIII.17: Spectrum after 10 ms of sideband cooling followed by 10 ms of SBH on the first blue sideband. A fit to the data points finds an average phonon number $\bar{n} = 146(2)$ with a standard deviation $\sigma = 14(1)$ for a distribution assumed to be Gaussian. The Rabi frequency is about 18 kHz.

spectrum in figure VIII.18 is also represented. Notice that the coupling strength of the fourth red sideband is almost constant where the probability of occupation of the motional state is high. It is contained between $0.365 \times \Omega_0$ and $0.400 \times \Omega_0$ on a range comprising 95 % of the population ($n = 244$ to $n = 344$). This allows Rabi oscillations to be observed on the fourth red sideband as can be seen in figure VIII.20 and importantly there is an almost full population transfer between the ground and excited electronic states. This allows us to apply π and $\pi/2$ pulses with a good fidelity.

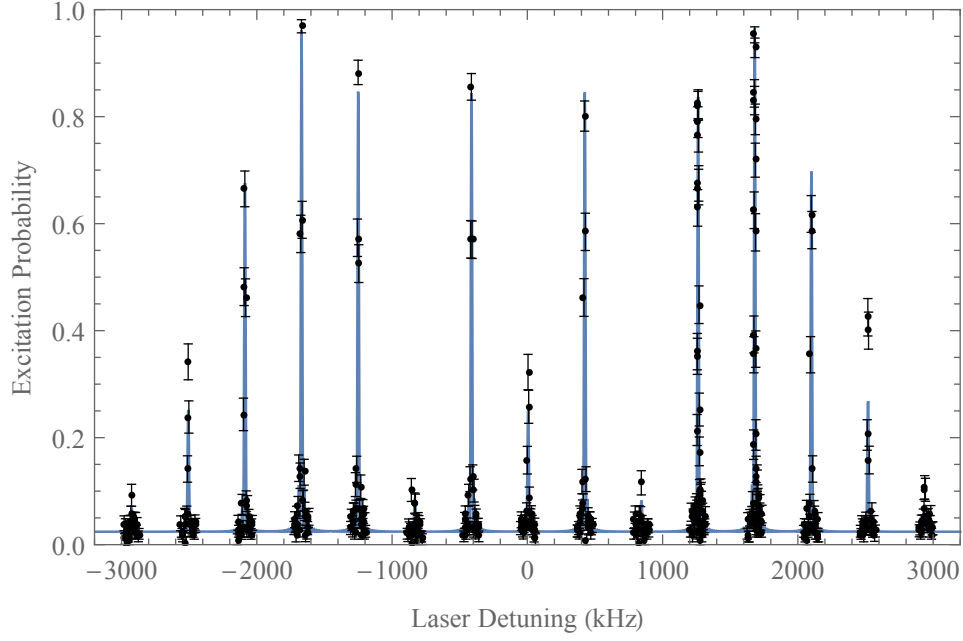


Figure VIII.18: Spectrum after 10 ms of sideband cooling followed by 10 ms of SBH on the first blue sideband and 20 ms on the second blue. A fit to the data points finds an average phonon number $\bar{n} = 294(2)$ with a standard deviation $\sigma = 25(3)$ for a distribution assumed to be Gaussian. The Rabi frequency is about 16 kHz.

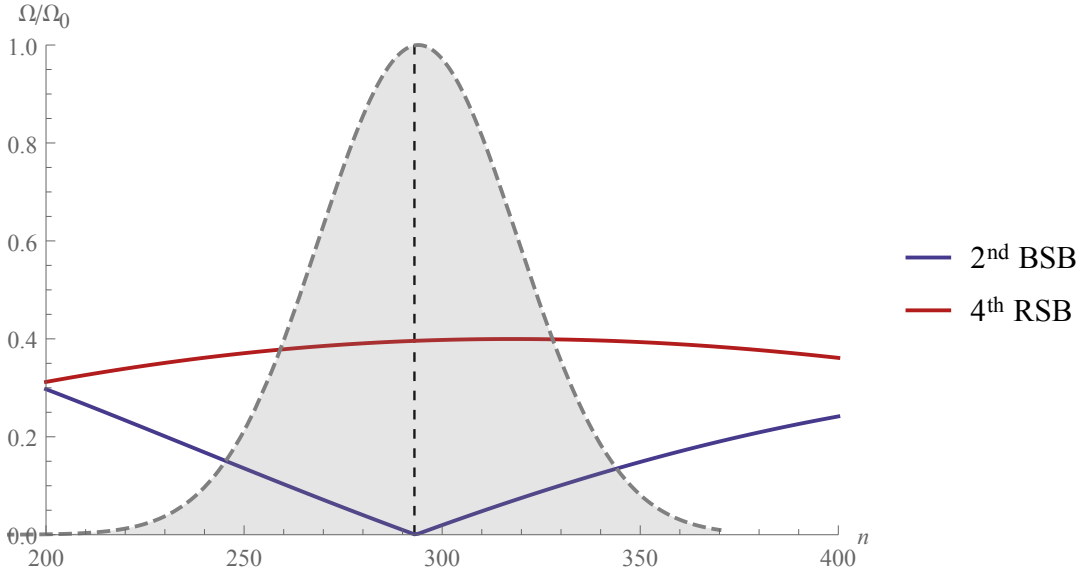


Figure VIII.19: Normalised coupling strengths for the second blue and fourth red sidebands for $\nu_z = 420$ kHz ($\eta = 0.15$). The dashed grey line represents the probability of occupation of the motional states for a Gaussian distribution centred at $n = 294$ with a standard deviation $\sigma = 25$ (normalised to 1 for $n = 294$).

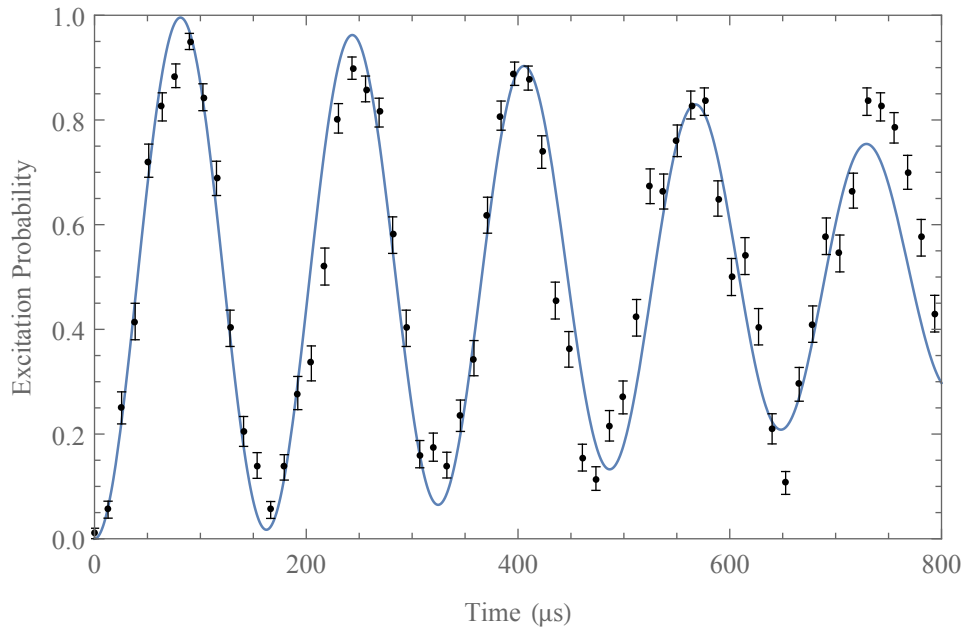


Figure VIII.20: Rabi oscillations on the fourth red sideband after two-stage SBH on the first and second blue sidebands. The fit assumes a Gaussian distribution of the motional states with the parameters found from the spectrum in figure VIII.18 and a Gaussian decay of the oscillations (due to decoherence). The Rabi frequency is about 16 kHz and the decay time constant $\tau_d = 0.73(7)$ ms.

Superpositions of high phonon states

We measure the motional coherence time when the ion is in a high motional state by creating superpositions of motional states after sideband heating and performing Ramsey experiments. The experiments are conducted at a trapping voltage of 420 kHz and two-stage SBH on the first and second blue sidebands is applied. This corresponds to the situation described earlier, the average phonon number of the ions is therefore around $n = 293$.

To create a motional superposition starting from $|g, 0\rangle$, the technique consisted in splitting the population between the electronic ground and excited states before transferring the population in the excited state to some other state $|g, n\rangle$ by addressing a red sideband which did not affect the population in $|g, 0\rangle$. This relied on the asymmetry in coupling strength between the red and blue sidebands close to $n = 0$. That technique is not applicable to high motional states because the ground state will be coupled to the laser light if the excited state is. Thus, if a $\pi/2$ pulse is applied on the carrier followed by a π pulse on the s^{th} sideband, a superposition of $|g, n - s\rangle$ and $|e, n + s\rangle$ will be formed. Although these two states have different phonon numbers, which we want, they also are in different electronic states which means this superposition is first and foremost subject to the decoherence of the optical qubit. Therefore the motional coherence time cannot be measured with such a superposition.

Instead, we create a superposition of four states by applying two consecutive $\pi/2$ pulses on different sidebands. For instance, a $\pi/2$ pulse on the carrier followed by a $\pi/2$ pulse on the third red sideband creates a superposition of $|e, n - 3\rangle$, $|g, n\rangle$, $|e, n\rangle$ and $|g, n + 3\rangle$. A reversed pulse sequence after a delay T completes the Ramsey experiment (see figure VIII.21).

Starting with an ion in $|\psi\rangle = |g, n\rangle$, after the first pulse on the carrier the state becomes:

$$|\psi\rangle = \frac{1}{\sqrt{2}} |g, n\rangle - \frac{i}{\sqrt{2}} |e, n\rangle. \quad (245)$$

The initial phase of the the first pulse was set to zero. For subsequent pulses, we denote the phases ϕ_2 , ϕ_3 , ϕ_4 . The application of a $\pi/2$ pulse on the third red sideband leads to the superposition of four states:

$$|\psi\rangle = \frac{1}{2} \left(-e^{i\phi_2} |e, n - 3\rangle + |g, n\rangle - i |e, n\rangle - ie^{-i\phi_2} |g, n + 3\rangle \right). \quad (246)$$

After a delay T during which the laser is blocked and set to the carrier frequency, the state of the ion is:

$$|\psi\rangle = \frac{1}{2} \left(-e^{i(\phi_2 + 3\omega_z T)} |e, n - 3\rangle + |g, n\rangle - i |e, n\rangle - ie^{-i(\phi_2 + 3\omega_z T)} |g, n + 3\rangle \right). \quad (247)$$

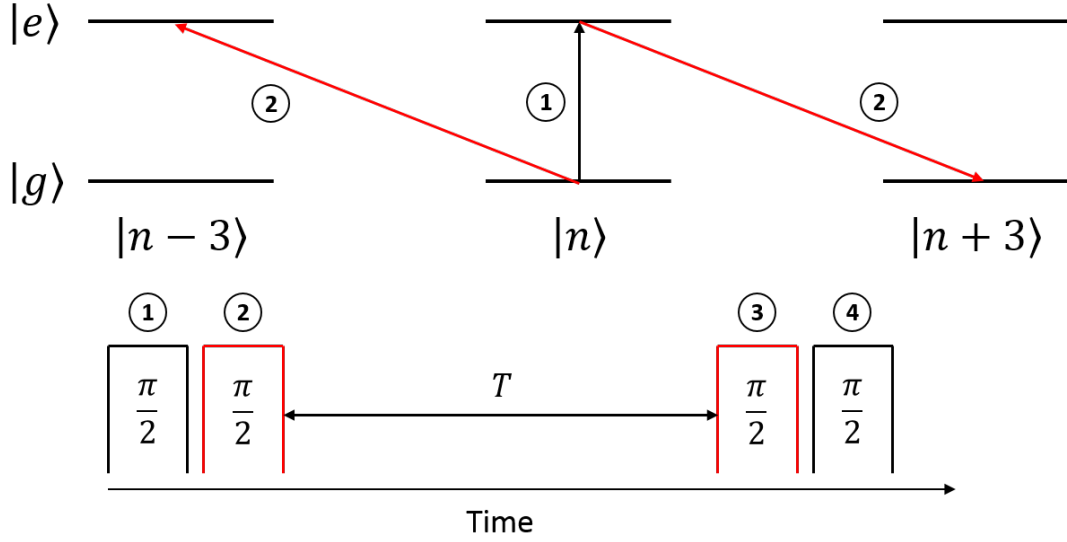


Figure VIII.21: Pulse sequence to create a superposition of $|e, n-3\rangle$, $|g, n\rangle$, $|e, n\rangle$ and $|g, n+3\rangle$ and perform a Ramsey experiment. Pulses 1 and 4 are tuned to the carrier frequency, pulses 2 and 3 to the third red sideband.

The second $\pi/2$ pulse on the third red sideband is then applied and yields:

$$|\psi\rangle = \frac{1}{2\sqrt{2}} \left(- (e^{i(\phi_2+3\omega_z T)} + e^{i\phi_3}) |e, n-3\rangle + (1 - e^{i(\phi_2-\phi_3+3\omega_z T)}) |g, n\rangle \right. \\ \left. + i(e^{i(\phi_3-\phi_2-3\omega_z T)} - 1) |e, n\rangle - i(e^{-i\phi_3} + e^{-i(\phi_2+3\omega_z T)}) |g, n+3\rangle \right). \quad (248)$$

The last pulse on the carrier couples $|g, n-3\rangle$ to $|e, n-3\rangle$, $|g, n\rangle$ to $|e, n\rangle$ and $|g, n+3\rangle$ to $|e, n+3\rangle$. The probability of excitation is therefore given by the sum of the probabilities to be in $|e, n-3\rangle$, $|e, n\rangle$ or $|e, n+3\rangle$. We obtain:

$$P_e(T) = \frac{1}{8} \left(4 - 2 \cos(3\omega_z T + \phi_2 - \phi_3 + \phi_4) + \cos(6\omega_z T + 2\phi_2 - 2\phi_3 + \phi_4) + \cos(\phi_4) \right). \quad (249)$$

The phases of the two frequency components in this expression have an important difference: $\phi_2 - \phi_3 + \phi_4$ remains unchanged by laser frequency noise whereas $2\phi_2 - 2\phi_3 + \phi_4$ does not so that the amplitude of the component at $6\omega_z$ will be affected by optical decoherence but not the one at $3\omega_z$. Figure VIII.22 shows the measured probability of excitation as a function of time for Ramsey experiments with different initial delays. In VIII.22.a, the initial delay is $10 \mu\text{s}$ which is well within the optical coherence domain and therefore the amplitude of both frequency components is maximum. As the delay gets longer, the $6\omega_z$ component fades away and the excitation probability eventually follows a simple sinusoidal function oscillating at $3\omega_z$ with a maximum visibility of one half. The overall visibility is taken

as the average of the amplitudes of the two frequency components. We fit it with a sum of two Gaussian functions corresponding to the optical (τ_1) and motional coherences (τ_2):

$$V(T) = \frac{1}{2} \left(V_1 \exp \left(\frac{-T^2}{2\tau_1^2} \right) + V_2 \exp \left(\frac{-T^2}{2\tau_2^2} \right) \right). \quad (250)$$

V_1 and V_2 respectively represent the amplitudes of the $6\omega_z$ and $3\omega_z$ components at $T = 0$. In principle, the amplitude of the $6\omega_z$ component should be affected by the motional decoherence but we can neglect it because the optical coherence time is much shorter than the motional one. The experiments are performed at high voltage (250 V) where we expect the motional coherence to be limited by dephasing which justifies the use of a Gaussian model. The optical coherence time is given by:

$$T_c^o = \tau_1 \sqrt{2(1 + \ln(V_1))} \quad (251)$$

and the motional coherence time by:

$$T_c^m = \tau_2 \sqrt{2(1 + \ln(V_2))}. \quad (252)$$

From the fit of the visibility (in figure VIII.23), we find $T_c^o = 700(60) \mu\text{s}$ and $T_c^m = 140(15) \text{ms}$. The initial visibility is not very high (about 0.87); this is most likely because of the spread in the probability distribution of the motional state. The motional coherence time corresponds to a standard deviation of the axial frequency approximately equal to 0.49(5) Hz.

We perform similar Ramsey experiments with different superpositions by changing the sidebands addressed in the pulse sequence. With a $\pi/2$ pulse on the first red sideband followed by a $\pi/2$ pulse on the third red sideband, we create a superposition of $|e, n-3\rangle$, $|e, n-1\rangle$, $|g, n\rangle$ and $|g, 2\rangle$. In this case, the excitation probability at the end of the Ramsey sequence (the laser is tuned to the first RSB during the delay) has a frequency component at $2\omega_z$ and one $4\omega_z$. A pulse on the third blue sideband followed by a pulse on the third red creates a superposition of $|e, n-3\rangle$, $|g, n\rangle$, $|e, n+3\rangle$ and $|g, n+6\rangle$ in which case the two frequency components of the excitation probability will be at $6\omega_z$ and $12\omega_z$ (with the laser at the third BSB during the delay). The visibility is measured for these two superpositions for different delays (only for latter times past the optical coherence for the second one), the results are shown in figure VIII.24. For the superposition of $|e, n-3\rangle$, $|e, n-1\rangle$, $|g, n\rangle$ and $|g, 2\rangle$, we find an optical coherence time $T_c^o = 665(90) \mu\text{s}$ consistent with the previous value and a motional coherence time $T_c^m = 280(40) \text{ms}$. With the $|e, n-3\rangle$, $|g, n\rangle$, $|e, n+3\rangle$, $|g, n+6\rangle$ superposition, the motional coherence time is $T_c^m = 120(20) \text{ms}$. These last two motional coherence times respectively correspond to standard deviations of the axial frequency of 0.37(5) Hz and 0.27(4) Hz which are in rough agreement

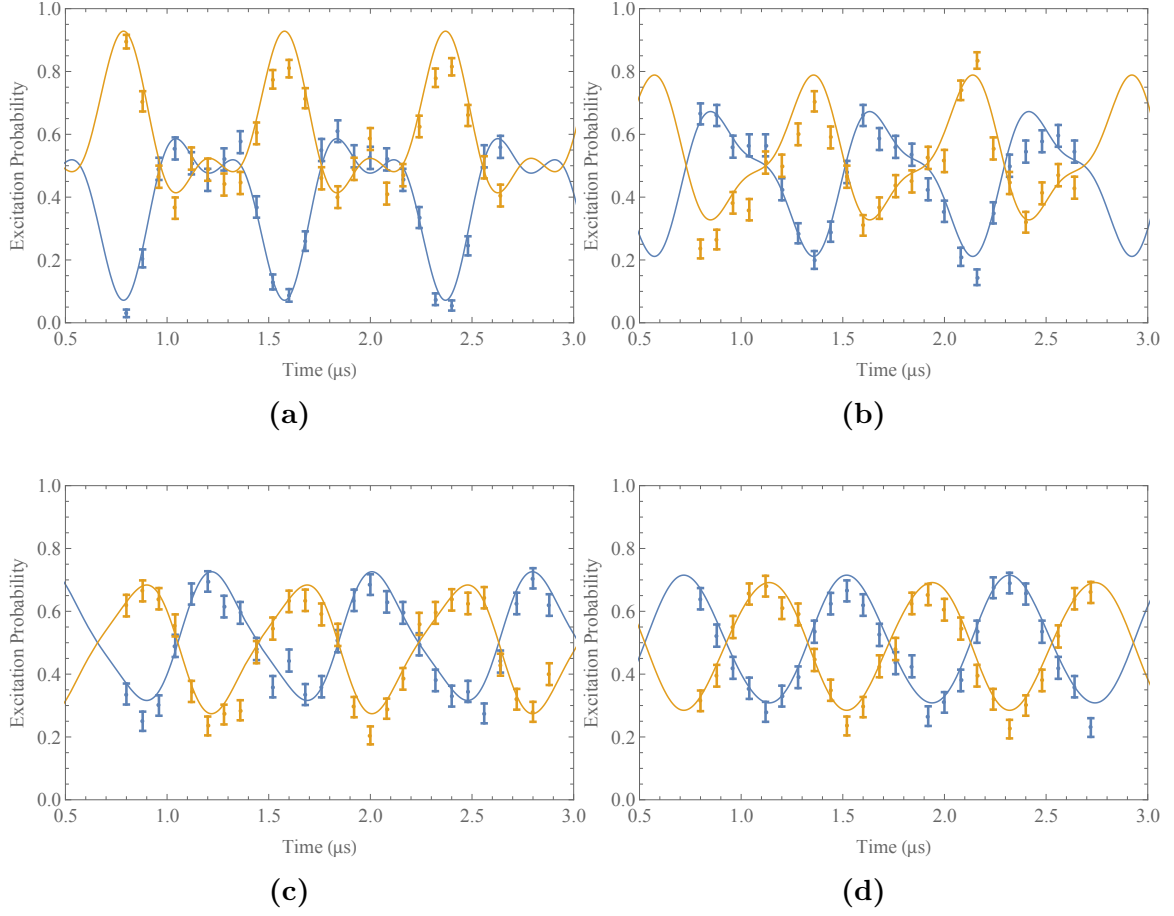


Figure VIII.22: Ramsey experiments with a superposition of $|e, n-3\rangle$, $|g, n\rangle$, $|e, n\rangle$ and $|g, n+3\rangle$ with initial delays of (a) 10 μs , (b) 600 μs , (c) 1 ms and (d) 20 ms. The experiment contains two interleaved sequences, at the origin of the two curves, differing by a π offset on the last laser pulse.

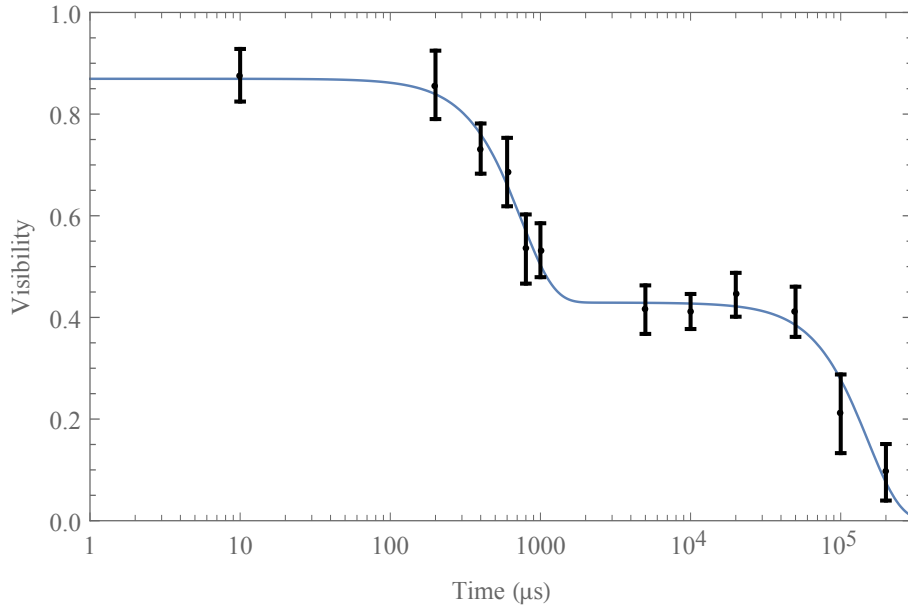


Figure VIII.23: Visibility in Ramsey experiments as a function of the delay time for a superposition of $|e, n - 3\rangle$, $|g, n\rangle$, $|e, n\rangle$ and $|g, n + 3\rangle$.

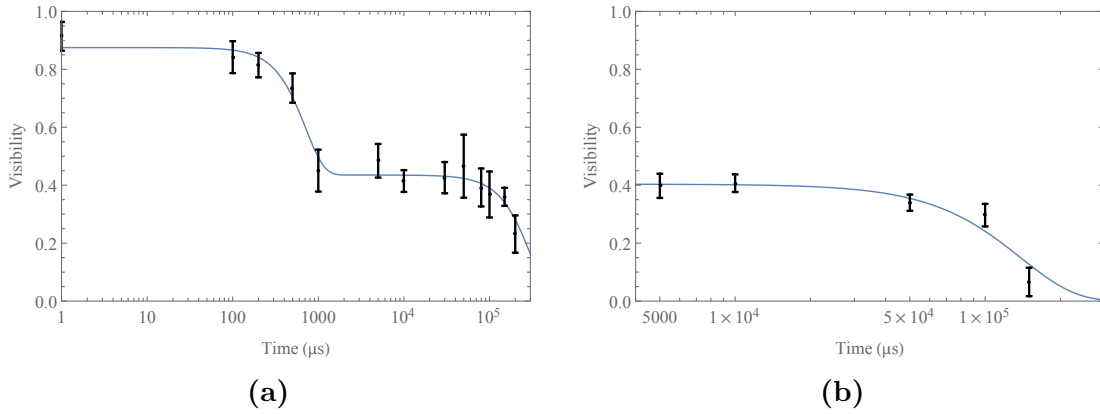


Figure VIII.24: Visibility in Ramsey experiments as a function of the delay time for **(a)** a superposition of $|e, n - 3\rangle$, $|e, n - 1\rangle$, $|g, n\rangle$ and $|g, 2\rangle$ and **(b)** a superposition of $|e, n - 3\rangle$, $|g, n\rangle$, $|e, n + 3\rangle$ and $|g, n + 6\rangle$.

with the value found for the superposition of $|e, n - 3\rangle$, $|g, n\rangle$, $|e, n\rangle$ and $|g, n + 3\rangle$ and with the results of the Ramsey experiments with low motional states in the previous section. Globally, we notice a decrease in the coherence time for larger separation of the motional states but no clear scaling can be deduced from our results. Additional measurements are needed to better characterise the motional decoherence and to find how it depends on factors such as the separation of the motional states and their energy. Improvements to the experimental set-up, in particular to the Doppler cooling lasers, would be welcome to offer the stability required to make sensible comparisons between different experiments. The sideband heating technique may be improved, for instance by choosing more carefully the pulse durations, to produce narrower distributions which would reduce systematic errors in the Ramsey experiments.

VIII.4 Coherent States

In this section, we present the creation of coherent states of the ion's axial motion. A coherent state $|\alpha\rangle$ is defined as the eigenstate of the annihilation operator a_z : $a_z |\alpha\rangle = \alpha |\alpha\rangle$. Expressed in the basis of the Fock states $|n\rangle$, we have:

$$|\alpha\rangle = e^{-|\alpha|^2/2} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle \quad (253)$$

such that the probability of occupation of a given state $|n\rangle$ follows a Poisson distribution:

$$P(n) = \langle n|\alpha\rangle^2 = e^{-|\alpha|^2} \frac{\alpha^{2n}}{n!} \quad (254)$$

where the average phonon number $\bar{n}$ is equal to α^2 . Experimentally, we create coherent states by shining a bichromatic 729 nm laser beam⁵ on a ground state cooled ion where the two frequency components are tuned to the first red and first blue sidebands. In the Lamb-Dicke regime the interaction Hamiltonian with the bichromatic beam is, after a second application of the rotating wave approximation:

$$H_I = i \frac{\hbar \Omega_0}{2} \left[(a_z^\dagger + a_z) (e^{i\phi} |e\rangle \langle g| + e^{-i\phi} |g\rangle \langle e|) \right] \quad (255)$$

This interaction is equivalent to a displacement operator $D(\alpha)$ which, if applied to the motional ground state, creates a coherent state [120, 121]. The displacement operator is given by

$$D(\alpha) = \exp(\alpha a_z^\dagger - \alpha^* a_z) \quad (256)$$

where α is a function of the bichromatic interaction time related to the Rabi frequency and the Lamb-Dicke parameter by $\alpha(t) = i\eta\Omega_0 t$. Coherent states can be revealed with standard techniques like spectroscopy and Rabi oscillations. Figure VIII.25 shows the probability of excitation as a function of the probe pulse time (probe on first blue sideband). The ion is first sideband cooled to the ground state before being illuminated with bichromatic light for 200 μ s. The data is fitted assuming a Poissonian distribution of the motional states. We find $\bar{n} = \alpha^2 = 3.6(2)$. The oscillations display a characteristic collapse and revival. This is due to a small number of different coupling strengths for the blue sideband resulting in different Rabi oscillation frequencies for each occupied motional state.

The spectra in figure VIII.26 were taken after 1 ms of bichromatic excitation. A fit gives an average phonon number $\bar{n} = 49.2(7)$. An interesting feature can be

⁵The bichromatic beam is produced by feeding two RF signals to the AOM in single pass configuration.

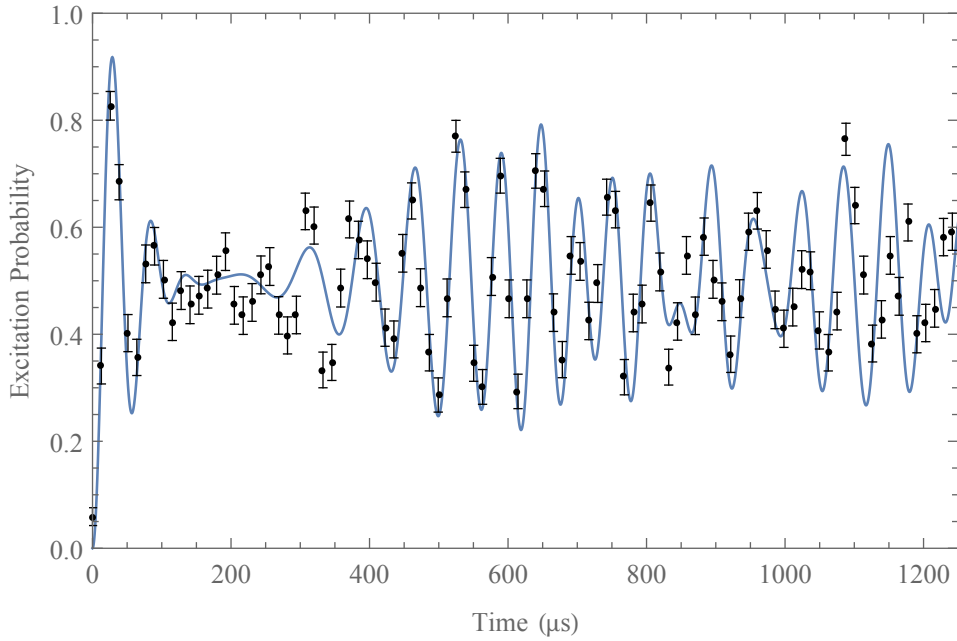
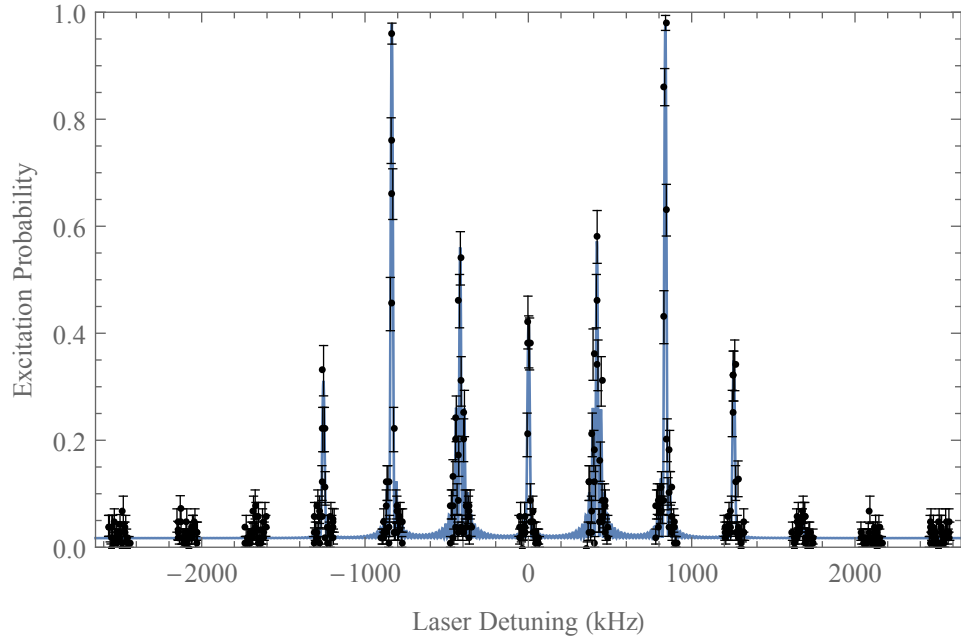


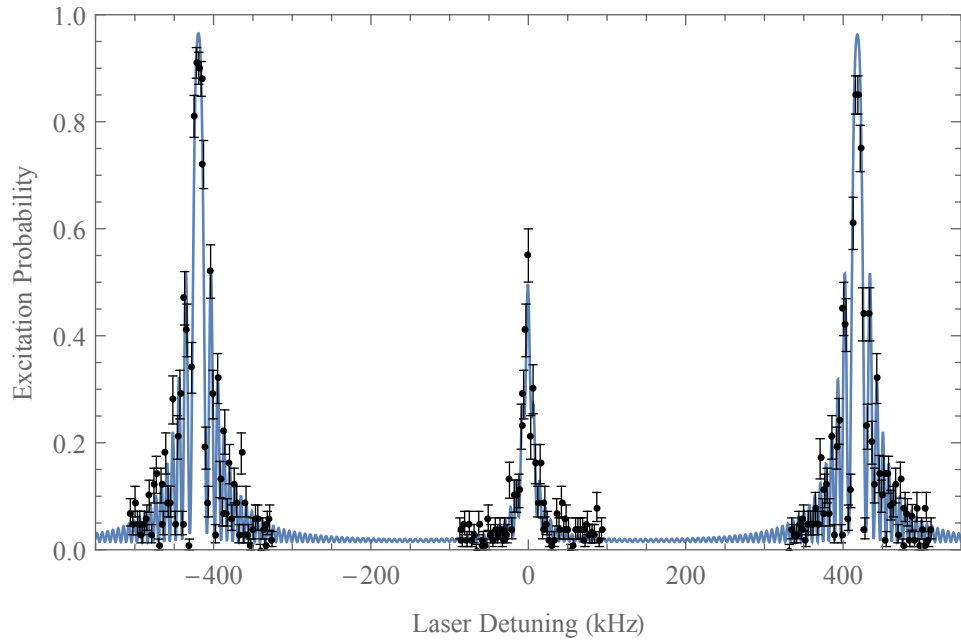
Figure VIII.25: Rabi oscillations on the first blue sideband after a $200\ \mu\text{s}$ bichromatic excitation. The fit suggests a coherent state with $\bar{n} = 3.6(2)$ is created. The axial frequency is $420\ \text{kHz}$.

noted: when the probe is tuned to the first red and blue sidebands, the ion is driven coherently as evidenced by the fringes on the sidebands. The carrier on the contrary shows no such sign of coherent drive. This is confirmed by the time-domain data in figure VIII.27: Rabi oscillations are observed with the blue sideband whereas the probability of excitation quickly converges to one half when addressing the carrier. As for a sideband heated ion, this is well understood when looking at the coupling strengths of the different sidebands as a function of the phonon state (see figure VIII.28). Unlike the carrier's, the coupling strength of the blue sideband undergoes little variation over the range of populated motional states.

The evolution of the coherent state with the bichromatic interaction time can be monitored by applying a probe pulse of a fixed length after increasingly long bichromatic interactions and measuring the probability of excitation. This is shown in figure VIII.29. We perform two experiments: one with the probe pulse on the carrier and one with the probe on the first blue sideband. The powers and durations of the probe are set to perform a 4π pulse on the carrier and a π pulse on the first BSB in the motional ground state. We notice an interesting phenomenon: after some interaction time t_1 , the average phonon number of the coherent state reaches a maximum before decreasing and eventually going back to zero at $2t_1$. This is evidenced on the plots in figure VIII.29 by a reflection symmetry with respect to a vertical line at $t_1 = 467\ \mu\text{s}$. For longer interaction times, after $2t_1$, the average phonon number increases again and we observe a sinusoidal evolution of the coherent state. This behaviour is similar to that of a classical harmonic oscillator driven



(a)



(b)

Figure VIII.26: (a) Spectrum taken after a 1 ms bichromatic excitation. The fit gives $\bar{n} = 49.2(7)$. (b) Close up on the carrier and the first pair of sidebands with a longer probe pulse. Fringes on the sides of the sidebands indicate a coherent drive.

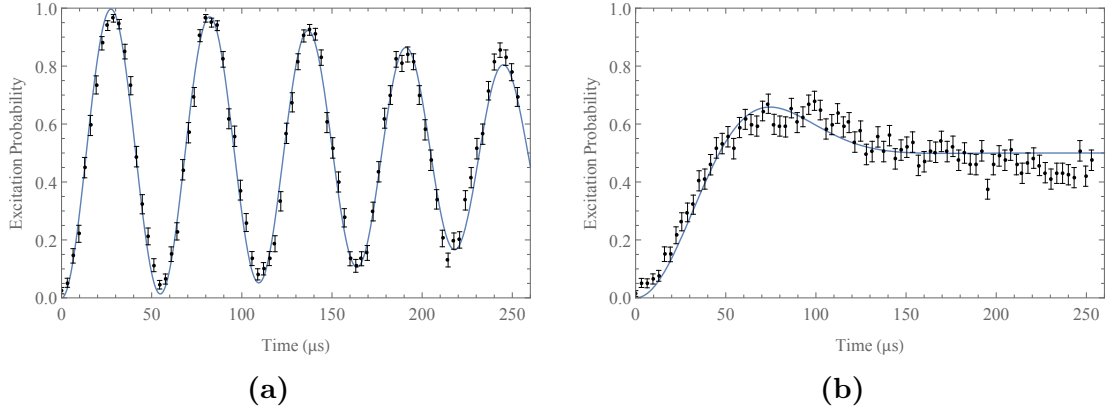


Figure VIII.27: Rabi oscillations after a 1 ms bichromatic excitation on (a) the first blue sideband and (b) the carrier. The fits indicate respectively $\bar{n} = 52.7(6)$ and $\bar{n} = 49.7(6)$. The Rabi frequency (Ω_0) is about 33 kHz.

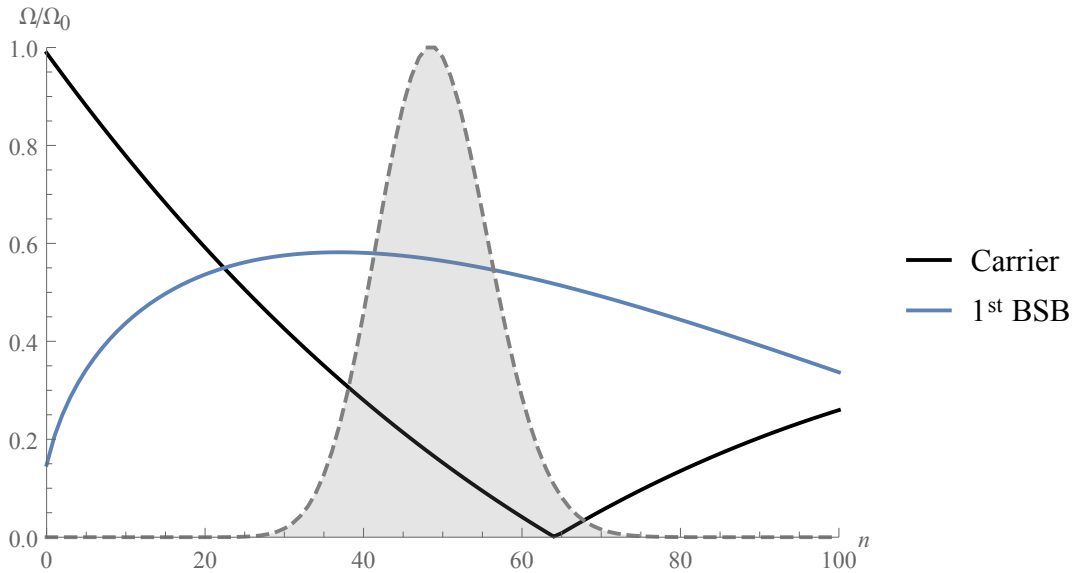


Figure VIII.28: Normalised coupling strengths for the carrier and first blue sideband for $\nu_z = 420$ kHz ($\eta = 0.15$). The dashed grey line represents the probability of occupation of the motional states for a Poisson distribution with a mean value equal to 49 (normalised to 1 for $n = 49$).

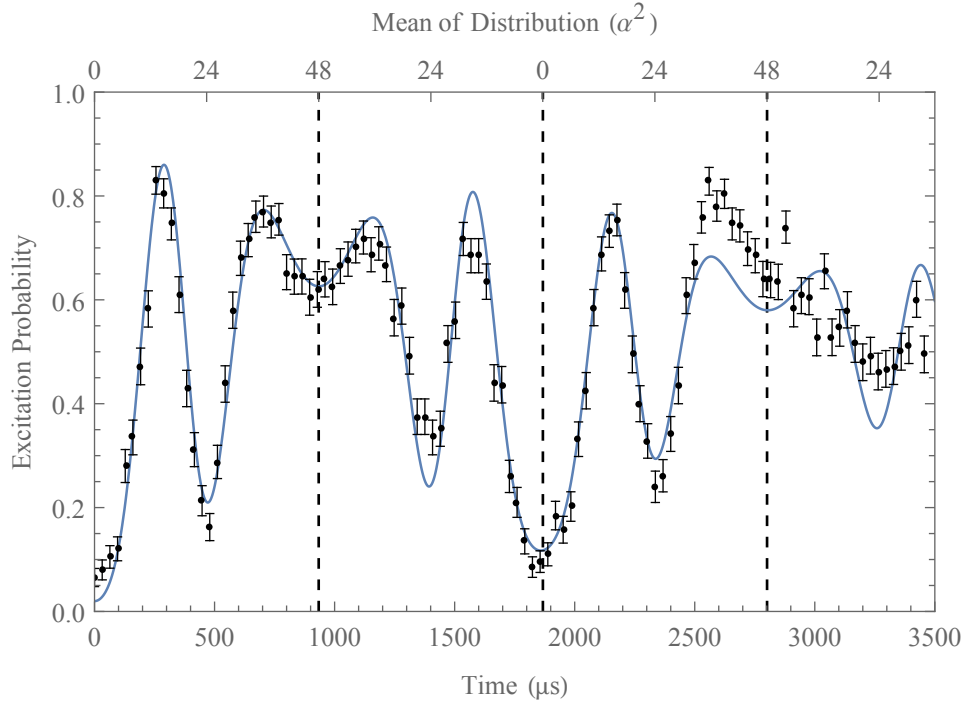
off-resonantly where the envelope of the amplitude oscillates at the frequency of the beat note. We therefore fit the data considering a coherent state with an average phonon number evolving sinusoidally:

$$P_e(t) = \sum_{n=0}^{100} \frac{e^{-\bar{n}(t)} \bar{n}(t)^n}{n!} \frac{1}{2} \left(1 - \cos(\Omega_{n,n+s} \tau_p) e^{-t^2/(2\tau_d^2)} \right) \quad (257)$$

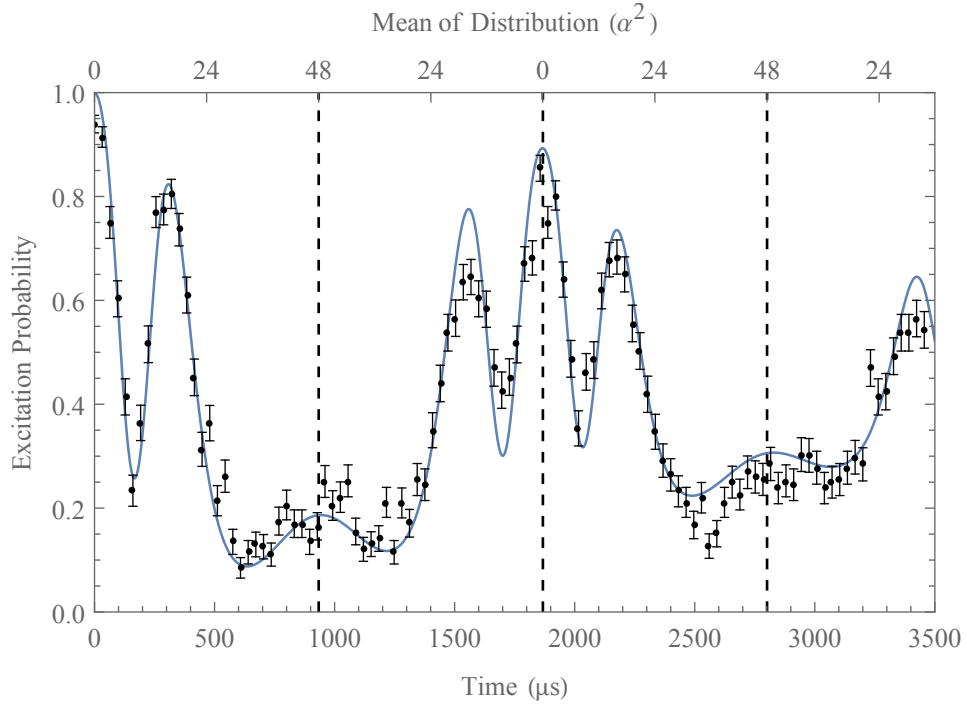
with

$$\bar{n}(t) = \frac{1}{2} \bar{n}_{max} (1 - \cos(\omega_b t)) \quad (258)$$

where τ_p is the duration of the probe pulse and ω_b the oscillation (beat) frequency characterising the evolution of the coherent state. The Gaussian decay factor is here to take decoherence into account. We find maximum average phonon numbers of 49.0(5) and 47.9(6) respectively for the carrier and blue sideband probes and $\omega_b/2\pi = 536(1)$ Hz. This indicates that there is some detuning between the frequency components of the bichromatic beam and the resonant frequencies of the first blue and red sidebands.



(a)



(b)

Figure VIII.29: Probability of the ion being in the excited state as a function of the bichromatic interaction time. In (a) the probe pulse is tuned to the carrier with power and duration calibrated to be a 4π pulse in the ground state. In (b) the probe pulse is tuned to the first blue sideband with power and duration calibrated to be a π pulse in the ground state. The details of the fits are given in the text.

IX

Conclusion

Previous theses of the group have detailed the development of the Penning trap experiment at Imperial College which eventually led to the demonstration of sideband cooling of the axial motion of a single $^{40}\text{Ca}^+$ ion to the ground state in the summer of 2014 [74, 76, 77, 78, 89, 90]. This thesis has presented improvements to the experimental set-up implemented since as well as new results in laser cooling and coherent manipulation of calcium ions in a Penning trap.

Although the experiment had already been well developed, with a tried and tested trap, functional Doppler and spectroscopy lasers and a control system based on a FPGA and a computer interface, modifications were necessary to conduct more involved experiments. I introduced a new radio-frequency driver based on direct digital synthesis to fine-tune the parameters of the 729 nm laser which allowed for more flexibility and complexity in the design of experiments. In particular this gave us the ability to carry out sophisticated multiple-stage sideband cooling (MSSBC) sequences which have been widely used in our experiments. The addition of a “noise eater” in the optical path of the spectroscopy laser improved the quality and repeatability of the results, especially for coherent manipulations, while the installation of a new EMCCD camera offered region specific detection. Other smaller improvements were made to enhance the reliability of the systems, the quality of the results and the ergonomics of the set-up.

As discussed in length in the previous chapters, a major difficulty of sideband cooling in Penning traps resides in the typically low trapping frequencies, compared to Paul traps, leading to large Lamb-Dicke parameters. To cool even the simple harmonic axial motion of a single ion to the ground state often requires several motional sidebands to be addressed. Using multiple-stage sideband cooling and adiabatic cooling, we were able to achieve ground state cooling with a high occupation probability (at least 93 %) over a wide range of axial frequencies (67 to 450 kHz). These techniques are a priori applicable to different ion species in both Penning and Paul traps.

MSSBC was also employed to sideband cool the radial motion of a single ion and

attain average phonon numbers below one for both the modified cyclotron and magnetron modes. We found that applying an axialisation field was necessary during Doppler cooling to reduce the magnetron phonon number sufficiently to initiate sideband cooling. Very high radial heating rates of several hundred phonons per second were measured for both modes. The reason for this large anomalous heating remains undetermined and should be investigated as it constitutes a limit to the cooling efficiency of the radial motion. There is certainly room for improvement for the radial Doppler cooling through a fine-tuning of certain experimental parameters concerning the radial Doppler beam (focus, offset, intensity) and the axialisation drive.

Our achievements in sideband cooling of a single ion opened the way for coherent manipulations of the electronic and motional states. The ability to prepare an ion in the ground state at very different trapping frequencies has allowed us to perform Ramsey experiments to study the coherence of the axial motion in different regimes. We found that the motional coherence of the ion at high frequency (420 kHz) is limited by instabilities in the trapping frequency but the coherence time is nevertheless large with a maximum of 565(21) ms measured with a superposition of $|g, 0\rangle$ and $|g, 1\rangle$. The effect of dephasing on a coherent superposition of motional states can be partially compensated by dynamical decoupling techniques. At low frequency (125 kHz), decoherence was dominated by anomalous heating and reduced to 77(2) ms for the same superposition. Systematic measurements of the coherence time and the heating rate on a bigger sample of axial frequencies would provide us with valuable information about the frequency dependence of anomalous heating and its contribution to decoherence which constitutes a significant obstacle for QI experiments. A decrease in the coherence time was observed as the separation of the Fock states gets larger in the motional superpositions, both for low and high motional states. The data are however insufficient to establish precise trends.

Progress in our sideband cooling techniques has allowed us to cool ion Coulomb crystals close to the ground state. This was demonstrated on a two-ion chain as well as on planar crystals of up to ten ions. We found that sideband cooling of ion chains is very challenging in the Penning trap due to the low trapping frequency required for the stability of this configuration and is probably not scalable to larger crystals. It is, in my opinion, not worth pursuing sideband cooling of long ion chains in Penning traps since it can be done much more easily in linear Paul traps which are naturally better suited for this purpose. The resonant frequency shift between the ions due to magnetic field inhomogeneities poses an other problem with chains. A careful compensation with additional coils may be necessary to cancel the shifts and be able to drive all the ions coherently.

On the contrary, sideband cooling of small planar crystals, up to five or six ions, turned out to be relatively simple for two reasons. The slow rotation of the crystal,

close to the magnetron frequency under normal trapping conditions, results in a near degeneracy of the axial modes which can be addressed simultaneously with one laser frequency. The higher trapping frequencies where the planar configuration is stable are synonymous with smaller Lamb-Dicke parameters, reducing the need to address high order sidebands. MSSBC was however necessary with the ten-ion crystal for which the axial modes are more numerous and more separated and extending this technique to larger crystals may prove challenging.

A possible improvement to the existing trap could be a ring electrode with more segments (8 for instance) which would give us the ability to apply a rotating wall to fix the rotation frequency of the crystals. This may allow us to perform sideband cooling on three-dimensional crystals, something we have been unable to do so far. A better fluorescence collection would also be useful to determine the motional state of the crystals more accurately. Some gains could be obtained by using more modern PMTs with a higher quantum efficiency and by increasing the numerical aperture of the imaging system.

At the time of writing (April 2018) and to our knowledge, our results are the only reported (published) demonstration of near ground state cooling of two-dimensional crystals in a Penning trap. However, recent work carried out at NIST has shown sub-Doppler cooling of large planar crystals of beryllium ions using EIT cooling [122]. Sub-Doppler cooling techniques of ICCs in Penning traps may be particularly useful to the fields of quantum information and quantum simulations where low temperatures improve the fidelity of the operations and where it is often necessary to operate in the Lamb-Dicke regime [123] which is not always attainable with Doppler cooling alone.

Bibliography

- [1] Nobel Media AB. The Nobel prize in physics 1989. http://www.nobelprize.org/nobel_prizes/physics/laureates/1989/.
- [2] H. Häffner *et al.* Quantum computing with trapped ions. *Phys. Rep.*, **469**, 155-203, (2008).
- [3] J. W. Britton *et al.* Engineered two-dimensional Ising interactions in a trapped-ion quantum simulator with hundreds of spins. *Nature*, **484**, 489-492, (2012).
- [4] A. Bylinskii *et al.* Tuning friction atom by atom in an ion crystal simulator. *Science*, **384** (6239), 1115-1118, (2015).
- [5] H. S. Margolis. Optical frequency standard and clocks. *Contemp. Phys.*, **51**, 1, 37-58, (2010).
- [6] C. W. Chou *et al.* Frequency comparison of two high-accuracy Al^+ optical clocks. *Phys. Rev. Lett.*, **104**, 070802, (2010).
- [7] A. E. Learnhardt *et al.* High-resolution spectroscopy on trapped molecular ions in rotating electric fields: a new approach for measuring the electron electric dipole moment. *J. Mol. Spectrosc.*, **270**, 1-25, (2011).
- [8] W. B. Cairncross *et al.* Precision measurement of the electron's electric dipole moment using trapped molecular ions. *Phys. Rev. Lett.*, **119**, 153001, (2017).
- [9] W. Quint *et al.* Laser-microwave double-resonance technique for g -factor measurements in highly charged ions. *Phys. Rev. A*, **78**, 032517, (2008).
- [10] H. Dehmelt. Experiments with an isolated subatomic particle at rest. *Rev. Mod. Phys.*, **62**, 525, (1990).

- [11] P. F. Staannum *et al.* Rotational laser cooling of vibrationally and translationally cold molecular ion. *Nat. Phys.*, **6**, 271-274, (2010).
- [12] W. Nörtershäuser *et al.* First observation of the ground-state hyperfine transition in $^{209}\text{Bi}^{80+}$. *Phys. Scr.*, **T156**, 014016, (2013).
- [13] L. Schmöger *et al.* Coulomb crystallization of highly charged ions. *Science*, **347** (6227), 1233-1236, (2015).
- [14] G. Andresen *et al.* Antimatter plasmas in a multipole trap for antihydrogen. *Phys. Rev. Lett.*, **98**, 023402, (2007).
- [15] R. C. Thompson. Ion Coulomb crystals. *Contemp. Phys.*, **56**, 63-79, (2015).
- [16] J. I. Cirac & P. Zoller. Quantum computation with cold trapped ions. *Phys. Rev. Lett.*, **74**, 20, (1995).
- [17] B. Lekitsch *et al.* Blueprint for a microwave trapped ion quantum computer. *Sci. Adv.*, **3**:e1601540, (2017).
- [18] A. Bermudez *et al.* Assessing the progress of trapped-ion processors towards fault-tolerant quantum computation. *Phys. Rev. X*, **7**, 041061, (2017).
- [19] J. P. Gaebler *et al.* High-fidelity universal gate for $^9\text{Be}^+$ ion qubits. *Phys. Rev. Lett.*, **117**, 060505, (2016).
- [20] T. P. Harty *et al.* High-fidelity trapped-ion quantum logic using near-field microwaves. *Phys. Rev. Lett.*, **117**, 140501, (2016).
- [21] IonQ Inc. IonQ is developing world-leading general-purpose quantum information processors. <https://ionq.co/>.
- [22] Austrian Business Agency. Austria promotes quantum computer spin-off. <https://investinaustria.at/en/news/2018/03/quantum-computer.php>.
- [23] A. Sørensen & K. Mølmer. Quantum computation with ions in thermal motion. *Phys. Rev. Lett.*, **82**, 9, (1999).
- [24] Nobel Media AB. The Nobel prize in physics 2012. http://www.nobelprize.org/nobel_prizes/physics/laureates/2012/.
- [25] C. Monroe *et al.* Demonstration of a fundamental quantum logic gate. *Phys. Rev. Lett.*, **75**, 254, (1995).
- [26] D. Leibfried *et al.* Experimental demonstration of a robust, high-fidelity geometric two ion-qubit phase gate. *Nature*, **422**, 412-415, (2003).

- [27] F. Schmidt-Kaler *et al.* Realization of the Cirac-Zoller controlled-NOT quantum gate. *Nature*, **422**, 408-411, (2003).
- [28] T. Monz *et al.* Realization of a scalable Shor algorithm. *Science*, **351**, 1068-1070, (2016).
- [29] S. Debnath *et al.* Demonstration of a small programmable quantum computer with atomic qubits. *Nature*, **536**, 63-66, (2016).
- [30] N. Friis *et al.* Observation of entangled states of a fully controllable 20-qubit system. *Phys. Rev. X*, **8**, 021012, (2018).
- [31] A. Friedenauer *et al.* Simulating a quantum magnet with trapped ions. *Nat. Phys.*, **4**, 757-761, (2008).
- [32] R. Islam *et al.* Onset of a quantum phase transition with a trapped ion quantum simulator. *Nat. Commun.*, **2**:377, (2011).
- [33] J. G. Bohnet *et al.* Emergence and frustration of magnetism with variable-range interactions in a quantum simulator. *Science*, **340** (6132), 583-587, (2013).
- [34] P. Jurcevic *et al.* Direct observation of dynamical quantum phase transitions in an interacting many-body system. *Phys. Rev. Lett.*, **119**, 080501, (2017).
- [35] J. Zhang *et al.* Observation of a many-body dynamical phase transition with a 53-qubit quantum simulator. *Nature*, **551**, 601-604, (2017).
- [36] B. Yoshimura *et al.* Creation of two-dimensional coulomb crystals of ions in oblate Paul traps for quantum simulations. *EPJ Quantum Technology*, **1**:14, (2014).
- [37] M. Mielenz *et al.* Arrays of individually controlled ions suitable for two-dimensional quantum simulations. *Nat. Commun.*, **7**:11839, (2016).
- [38] Michael J. Biercuk *et al.* Experimental Uhrig dynamical decoupling using trapped ions. *Phys. Rev. A*, **79**, 062324, (2009).
- [39] P. Zou *et al.* Implementation of local and high-fidelity quantum conditional phase gates in a scalable two-dimensional ion trap. *Phys. Lett. A*, **374**, 1425-1430, (2010).
- [40] D. Porras & J. I. Cirac. Quantum manipulation of trapped ions in two dimensional Coulomb crystals. *Phys. Rev. Lett.*, **96**, 250501, (2006).
- [41] R. Blümel *et al.* Chaos and order of laser-cooled ions in a paul trap. *Phys. Rev. A*, **40**, 2, (1989).

- [42] J. M. Taylor & T. Calarco. Wigner crystals of ions as quantum hard drives. *Phys. Rev. A*, **78**, 062331, (2008).
- [43] J. D. Baltrusch *et al.* Fast and robust quantum computation with ionic Wigner crystals. *Phys. Rev. A*, **83**, 042319, (2011).
- [44] J. F. Goodwin *et al.* Trapped-ion quantum error-correcting protocols using only global operations. *Phys. Rev. A*, **92**, 032314, (2015).
- [45] J. G. Bohnet *et al.* Quantum spin dynamics and entanglement generation with hundreds of trapped ions. *Science*, **352** (6291), 1297-1301, (2016).
- [46] J. F. Goodwin *et al.* Resolved-sideband laser cooling in a penning trap. *Phys. Rev. Lett.*, **116**, 143002, (2016).
- [47] C. Monroe *et al.* Resolved-sideband Raman cooling of a bound atom to the 3d zero-point energy. *Phys. Rev. Lett.*, **75**, 22, (1995).
- [48] M. Bohman *et al.* Sympathetic cooling of protons and antiprotons with a common endcap Penning trap. *J. Mod. Opt.*, **65**:5-6, 568-576, (2018).
- [49] C. Smorra *et al.* Base – the baryon antibaryon symmetry experiment. *Eur. Phys. J.*, **224**, 3055-3108, (2015).
- [50] A. Derevianko *et al.* Highly charged ions as a basis of optical atomic clockwork of exceptional accuracy. *Phys. Rev. Lett.*, **109**, 180801, (2012).
- [51] M. Vogel & W. Quint. Aspects of fundamental physics in precision spectroscopy of highly charged ions in Penning traps. *Ann. Phys.*, **525**, 505-513, (2013).
- [52] Z. Andelkovic *et al.* Laser cooling of externally produced Mg ions in a Penning trap for sympathetic cooling of highly charged ions. *Phys. Rev. A*, **87**, 033423, (2013).
- [53] T. Murböck *et al.* Rapid crystallization of externally produced ion in a Penning trap. *Phys. Rev. A*, **94**, 043410, (2016).
- [54] G. Stutter *et al.* Sideband cooling of small ion Coulomb crystals in a Penning trap. *J. Mod. Opt.*, **65**, 64-74, (2018).
- [55] R. C. Thompson & J. Papadimitriou. Simple model for the laser cooling of an ion in a Penning trap. *J. Phys. B: At. Mol. Opt. Phys.*, **33**, 3393-3405, (2000).
- [56] J. R. Castrejón-Pita *et al.* Novel designs for Penning ion traps. *J. Mod. Opt.*, **54**, 1581-1594, (2006).

- [57] M. Brownnutt *et al.* Monolithic microfabricated ion trap chip design for scaleable quantum processors. *New J. Phys.*, **8**, 232, (2006).
- [58] W. Paul. Electromagnetic traps for charged and neutral particles. *Rev. Mod. Phys.*, **62**, 3, (1990).
- [59] Pradip K. Ghosh. *Ion traps*. Oxford university press, (1995).
- [60] L. S. Brown & G. Gabrielse. Geonium theory: Physics of a single electron or ion in a Penning trap. *Rev. Mod. Phys.*, **58**, 1, (1986).
- [61] D. F. V. James. Quantum dynamics of cold trapped ions with application to quantum computation. *Appl. Phys. B*, **66**, 181-190, (1998).
- [62] C.-C. Joseph Wang *et al.* Phonon-mediated quantum spin simulator employing a planar ionic crystal in a Penning trap. *Phys. Rev. A*, **87**, 013422, 2013.
- [63] Gilbert Grynberg & Alain Aspect. *Introduction to quantum optics*. Cambridge University Press, (2010).
- [64] D. Leibfried *et al.* Quantum dynamics of single trapped ions. *Rev. Mod. Phys.*, **75**, 281-324, (2003).
- [65] S. Stenholm. The semiclassical theory of laser cooling. *Rev. Mod. Phys.*, **58**, 699-739, (1986).
- [66] D. J. Wineland & H. Dehmelt. Proposed $10^{14} \delta\nu < \nu$ laser fluorescence spectroscopy on Ti^+ mono-ion oscillator. *Bull. Am. Phys. Soc*, **20**, 637, (1975).
- [67] W. Neuhauser *et al.* Optical sideband cooling of visible atom cloud confined in parabolic well.
- [68] D. J. Wineland & W. M. Itano. Laser cooling of atoms. *Phys. Rev. A*, **20**, 4, (1979).
- [69] W. M. Itano & D. J. Wineland. Laser cooling of ions stored in harmonic and Penning traps. *Phys. Rev. A*, **25**, 1, (1982).
- [70] V. Zhelyazkova *et al.* Laser cooling and slowing of CaF molecules. *Phys. Rev. A*, **89**, 053416, (2014).
- [71] C. Guet & W. R. Johnson. Relativistic many-body calculations of transition rates for Ca^+ , Sr^+ and Ba^+ . *Phys. Rev. A*, **44**, 3, (1991).
- [72] R. Gerritsma *et al.* Precision measurement of the branching ration of the $4p\ ^2P_{3/2}$ decay of Ca II. *Eur. Phys. J. D*, **50**, 13-19, (2008).

- [73] A. Kreuter *et al.* Experimental and theoretical study of the $3d\ ^2D$ -level lifetimes of $^{40}\text{Ca}^+$. *Phys. Rev. A*, **71**, 032504, (2005).
- [74] Daniel R. Crick. *Manipulation of individual laser cooled Ca^+ ions in a Penning trap*. PhD thesis, Imperial College London, (2009).
- [75] D. R. Crick *et al.* Magnetically induced electron shelving in a trapped Ca^+ ion. *Phys. Rev. A*, **81**, 052503, (2010).
- [76] Sean Donnellan. *Towards sideband cooling of a single $^{40}\text{Ca}^+$ ion in a Penning trap*. PhD thesis, Imperial College London, (2011).
- [77] Sandeep Mavadia. *Motional sideband spectra and Coulomb crystals in a Penning trap*. PhD thesis, Imperial College London, (2013).
- [78] Joseph Francis Goodwin. *Sideband cooling to the quantum ground state in a Penning trap*. PhD thesis, Imperial College London, (2015).
- [79] D. M. Segal & C. Wunderlich. *Cooling techniques for trapped ions*. Imperial College Press, (2013).
- [80] E. S. Phillips *et al.* Dynamics of axialized laser-cooled ions in a Penning trap. *Phys. Rev. A*, **78**, 032307, (2008).
- [81] D. M. Segal H. F. Powell and R. C. Thompson. Axialization of laser cooled magnesium ions in a Penning trap. *Phys. Rev. Lett.*, **89**, 9, (2002).
- [82] R. J. Hendricks *et al.* Laser cooling in the Penning trap: an analytical model for cooling rates in the presence of an axializing field. *J. Phys. B: At. Mol. Opt. Phys.*, **41**, 035301, (2008).
- [83] Hebe Frances Powell. *Quantum optics with a single trapped ion*. PhD thesis, Imperial College London, (2002).
- [84] Richard James Hendricks. *Spectroscopy and dynamics of laser cooled Ca^+ ions in a Penning trap*. PhD thesis, Imperial College London, (2006).
- [85] L. Schweikhard & A. G. Marshall. Excitation modes for Fourier transform-ion cyclotron resonance mass spectrometry. *J. Am. Soc. Mass. Spectrom.*, **4**, 433-452, (1993).
- [86] F. Diedrich *et al.* Laser cooling to the zero-point energy of motion. *Phys. Rev. Lett.*, **62**, 4, (1989).
- [87] I. Marzoli *et al.* Laser cooling of trapped three-level ions: Designing two-level systems for sideband cooling. *Phys. Rev. A*, **49**, 4, (1994).

- [88] Christian Felix Roos. *Controlling the quantum state of trapped ions*. PhD thesis, Leopold-Franzens-Universität Innsbruck, (2000).
- [89] Shailen Bharadia. *Towards laser spectroscopy of highly charged ions: dynamics of $^{40}\text{Ca}^+$ ions in a Penning trap*. PhD thesis, Imperial College London, (2011).
- [90] Graham Stutter. *Quantum dynamics of small numbers of ion in a Penning trap*. PhD thesis, Imperial College London, (2015).
- [91] C. E. Shannon. Communication in the presence of noise. *Proceedings of the IRE*, **86**, pp 10-21, (1949).
- [92] Alice Heather Burrell. *High fidelity readout of trapped ion qubits*. PhD thesis, Exeter College, University of Oxford, (2010).
- [93] Franziska Beck. Theory and simulation of ground state laser cooling in a Penning trap. Master’s thesis, Imperial College London, (2014).
- [94] M. K. Joshi *et al.* Population dynamics in sideband cooling of trapped ions outside the Lamb-Dicke regime. Manuscript in preparation, (2017).
- [95] D. Comparat. General conditions for quantum adiabatic evolution. *Phys. Rev. A*, **80**, 012106, (2009).
- [96] G. Poulsen & M. Drewsen. Adiabatic cooling of a single trapped ion. *arXiv*, 1210.4309v1, (2012).
- [97] M. Abramowitz & I. A. Stegun. *Handbook of mathematical functions with formulas, graphs and mathematical tables*. National Bureau of Standards, (1964).
- [98] B. E. King *et al.* Cooling the collective motion of trapped ions to initialize a quantum register. *Phys. Rev. Lett.*, **81**, 8, (1998).
- [99] C. A. Sackett *et al.* Experimental entanglement of four particles. *Nature*, **404**, 256-259, (2000).
- [100] F. Schmidt-Kaler *et al.* How to realize a universal quantum gate with trapped ions. *Appl. Phys. B*, **77**, 789-796, (2003).
- [101] K. Kim *et al.* Quantum simulation of frustrated ising spins with trapped ions. *Nature*, **465**, 590-594, (2010).
- [102] D. Gangloff *et al.* Velocity tuning of friction with two trapped atoms. *Nat. Phys.*, **11**, 915-919, (2015).
- [103] Jonathan Home. *Entanglement of two trapped-ion spin qubits*. PhD thesis, Linacre College, Oxford, (2006).

- [104] M. J. Biercuk *et al.* High-fidelity quantum control using ion crystals in a Penning trap. *Quantum Inf. Comput.*, **9**, 920-949, (2009).
- [105] A. Khan *et al.* Theoretical basis for quantum simulation with a planar ionic crystal in a penning trap using a triangular rotating wall. *Phys. Rev. A*, **92**, 043405, (2015).
- [106] Y. Seis *et al.* Large-scale error-correction protocols using global operations in ion traps. *arXiv*, 1606.00056v1, (2016).
- [107] R. Lechner *et al.* Electromagnetically-induced-transparency ground-state cooling of long ion strings. *Phys. Rev. A*, **93**, 053401, (2016).
- [108] L. Viola and S. Lloyd. Dynamical suppression of decoherence in two-state quantum systems. *Phys. Rev. A*, **58**, 4, (1998).
- [109] E. Knill L. Viola and S. Lloyd. Dynamical decoupling of open quantum systems. *Phys. Rev. Lett.*, **82**, 12, (1999).
- [110] E. L. Hahn. Spin Echoes. *Phys. Rev.*, **80**, 4, (1950).
- [111] H. Y. Carr & E. M. Purcell. Effects of diffusion in nuclear magnetic resonance experiments. *Phys. Rev.*, **94**, 3, (1954).
- [112] S. Meiboom & D. Gill. Modified spin-echo method for measuring nuclear relaxation times. *Rev. Sci. Instrum.*, **29**, 8, (1958).
- [113] Gotz S. Uhrig. Keeping a quantum bit alive by optimized pi-pulse sequences. *Phys. Rev. Lett.*, **98**, 100504, (2007).
- [114] Gotz S. Uhrig. Exact results on dynamical decoupling by pi-pulses in quantum information piprocesses. *New J. Phys.*, **10**, 083024, (2008).
- [115] Z-Y. Wang W. Yang and R-B. Liu. Preserving qubit coherence by dynamical decoupling. *Front. Phys.*, **6**, 1-13, (2011).
- [116] D. J. Szwer *et al.* Keeping a single qubit alive by experimental dynamic decoupling. *J. Phys. B: At. Mol. Opt. Phys.*, **44**, 025501, (2011).
- [117] Y. Wang *et al.* Single-qubit quantum memory exceeding ten-minute coherence time. *Nat. Photonics*, **11**, 646-650, (2017).
- [118] Q. A. Turchette *et al.* Decoherence and decay of motional quantum states of a trapped atom coupled to engineered reservoirs. *Phys. Rev. A*, **62**, 053807, (2000).
- [119] Analog Devices Inc. *AD9858 data sheet rev C*.

- [120] E. Solano *et al.* Entangled coherent states and squeezing in N trapped ions. *J. Opt. B Quantum Semiclassical Opt.*, **4**, 324-327, (2002).
- [121] Gerhard Kirchmair. *Quantum non-demolition measurements and quantum simulations*. PhD thesis, Leopold-Franzens-Universität Innsbruck, (2010).
- [122] John J. Bollinger. Private Communication, (2018).
- [123] B. C. Sawyer *et al.* Spin dephasing as a probe of mode temperature, motional state distributions, and heating rates in a two-dimensional crystal. *Phys. Rev. A*, **89**, 033408, (2014).

Appendices

A

Mathematics

Note to the reader: the derivations made here are not necessary to understand the main body of the thesis but can be useful if the reader cares to verify the equations in the text.

A.1 Classical Motion of a Single Ion

Derivation of the equations of motion and the total energy

The derivations of the radial mode frequencies and equations of motion are carried out here. We start with:

$$\begin{aligned}\ddot{x} - \frac{1}{2}\omega_z^2 x - \omega_c \dot{y} &= 0 \\ \ddot{y} - \frac{1}{2}\omega_z^2 y + \omega_c \dot{x} &= 0\end{aligned}\tag{259}$$

Let $u = x + iy$, we have:

$$\ddot{u} = \ddot{x} + i\ddot{y} = \frac{1}{2}\omega_z^2 x + \omega_c \dot{y} + \frac{1}{2}\omega_z^2 iy - \omega_c i\dot{x}\tag{260}$$

$$\Leftrightarrow \ddot{u} + i\omega_c \dot{u} - \frac{1}{2}\omega_z^2 u = 0.\tag{261}$$

This is a linear differential equation of the second order whose characteristic equation is:

$$\lambda^2 + i\omega_c \lambda - \frac{1}{2}\omega_z^2\tag{262}$$

and discriminant:

$$\Delta = -\omega_c^2 + 2\omega_z^2.\tag{263}$$

We assume Δ negative. The solutions are:

$$\lambda_{\pm} = \frac{1}{2}(-i\omega_c \pm i\sqrt{\omega_c^2 - 2\omega_z^2}). \quad (264)$$

Let $\lambda = -i\omega$,

$$\omega_{\pm} = \frac{1}{2}(\omega_c \pm \sqrt{\omega_c^2 - 2\omega_z^2}). \quad (265)$$

ω_+ is the modified cyclotron frequency and ω_- the magnetron frequency. The general solution of the differential equation is:

$$u = \tilde{\rho}_+ e^{-i\omega_+ t} + \tilde{\rho}_- e^{-i\omega_- t} \quad (266)$$

where $\tilde{\rho}_+, \tilde{\rho}_- \in \mathbb{C}$ or otherwise:

$$u = R_+ e^{-i\omega_+ t + \phi_+} + R_- e^{-i\omega_- t + \phi_-} \quad (267)$$

with $R_c, R_m \in \mathbb{R}$. The equations for x and y can now be found by taking the real and imaginary parts of u :

$$\begin{aligned} x &= \Re[u] = R_+ \cos(\omega_+ t + \phi_+) + R_- \cos(\omega_- t + \phi_-) \\ y &= \Im[u] = -R_+ \sin(\omega_+ t + \phi_+) - R_- \sin(\omega_- t + \phi_-). \end{aligned} \quad (268)$$

We are now able to calculate the total energy:

$$\begin{aligned} E &= E_{kin} + E_{pot} \\ \Leftrightarrow E &= \frac{1}{2}mv^2 + e\phi \\ \Leftrightarrow E &= \frac{1}{2}m[\dot{x}^2 + \dot{y}^2 + \dot{z}^2] + \frac{m\omega_z^2}{4}[2z^2 - x^2 - y^2] \\ \Leftrightarrow E &= \frac{1}{2}m[\omega_+^2 R_+^2 + \omega_-^2 R_-^2 + 2\omega_+ \omega_- R_+ R_- \cos((\omega_+ - \omega_-)t) + \omega_z^2 Z^2 \sin^2(\omega_z t)] \\ &\quad + \frac{m\omega_z^2}{4}[2Z^2 \cos^2(\omega_z t) - R_+^2 - R_-^2 - 2R_+ R_- \cos((\omega_+ - \omega_-)t)] \\ \Leftrightarrow E &= \frac{1}{2}m[\omega_+^2 R_+^2 + \omega_-^2 R_-^2 + \omega_z^2 R_+ R_- \cos(\omega_1 t) + \omega_z^2 Z^2 \sin^2(\omega_z t)] \\ &\quad + \frac{m\omega_z^2}{4}[2Z^2 \cos^2(\omega_z t) - R_+^2 - R_-^2 - 2R_+ R_- \cos(2\omega_1 t)] \\ \Leftrightarrow E &= \frac{1}{2}m[\omega_z^2 Z^2 + 2\omega_1(R_+^2 \omega_+ - R_-^2 \omega_-)]. \end{aligned} \quad (269)$$

The phases were dropped for simplicity.

A.2 Quantum Description

Derivation of equation 29

The expression of the radial Hamiltonian as a function of the vectors $\vec{V}_+$ and $\vec{V}_-$ is derived here. By definition:

$$\vec{V}_+ = \vec{\rho} + \omega_- \hat{z} \times \vec{\rho} \quad (270a)$$

$$\vec{V}_- = \vec{\rho} + \omega_+ \hat{z} \times \vec{\rho} \quad (270b)$$

therefore:

$$\vec{V}_+ - \vec{V}_- = -(\omega_+ - \omega_-) \hat{z} \times \vec{\rho} \quad (271)$$

$$\hat{z} \times \frac{\vec{V}_+ - \vec{V}_-}{2\omega_1} = -\hat{z} \times (\hat{z} \times \vec{\rho}) \quad (272)$$

and $\hat{z} \times (\hat{z} \times \vec{\rho}) = -\vec{\rho}$, hence:

$$\vec{\rho} = \hat{z} \times \frac{\vec{V}_+ - \vec{V}_-}{2\omega_1}. \quad (273)$$

We now calculate the time derivatives of $\vec{V}_+$ and $\vec{V}_-$:

$$\begin{aligned} \dot{\vec{V}}_+ &= \frac{d}{dt}(\vec{\rho} + \omega_- \hat{z} \times \vec{\rho}) \\ \Rightarrow \dot{\vec{V}}_+ &= \dot{\vec{\rho}} + \omega_- \hat{z} \times \vec{\rho}. \end{aligned} \quad (274)$$

From Newton's second law, we have:

$$\dot{\vec{\rho}} = -\omega_c \hat{z} \times \vec{\rho} + \frac{1}{2}\omega_z^2 \vec{\rho}. \quad (275)$$

Replacing $\dot{\vec{\rho}}$ in equation 274 with its above expression gives:

$$\begin{aligned} \dot{\vec{V}}_+ &= -\omega_c \hat{z} \times \vec{\rho} + \omega_- \hat{z} \times \vec{\rho} + \frac{1}{2}\omega_z^2 \vec{\rho} \\ \Leftrightarrow \dot{\vec{V}}_+ &= -\omega_+ \hat{z} \times \vec{\rho} + \frac{1}{2}\omega_z^2 \vec{\rho}. \end{aligned} \quad (276)$$

Besides:

$$\begin{aligned} \omega_+ \hat{z} \times \vec{V}_+ &= \omega_+ \hat{z} \times (\vec{\rho} + \omega_- \hat{z} \times \vec{\rho}) \\ \Leftrightarrow \omega_+ \hat{z} \times \vec{V}_+ &= \omega_+ (\hat{z} \times \vec{\rho} - \omega_- \vec{\rho}) \\ \Leftrightarrow \omega_+ \hat{z} \times \vec{V}_+ &= \omega_+ \hat{z} \times \vec{\rho} - \frac{1}{2}\omega_z^2 \vec{\rho} \end{aligned} \quad (277)$$

where we have used that $2\omega_+\omega_- = \omega_z^2$. Therefore:

$$\vec{V}_+ = -\omega_+\hat{z} \times \vec{V}_+. \quad (278)$$

Similarly, we can show that:

$$\vec{V}_- = -\omega_-\hat{z} \times \vec{V}_-. \quad (279)$$

We can now express $\vec{\rho}$ as a function of $\vec{V}_+$ and $\vec{V}_-$:

$$\begin{aligned} \vec{\rho} &= \frac{1}{2\omega_1} \left(\frac{d\hat{z}}{dt} \times (\vec{V}_+ - \vec{V}_-) + \hat{z} \times \frac{d}{dt}(\vec{V}_+ - \vec{V}_-) \right) \\ \Rightarrow \vec{\rho} &= \frac{1}{2\omega_1} (\hat{z} \times (\vec{V}_+ - \vec{V}_-)) \\ \Leftrightarrow \vec{\rho} &= \frac{1}{2\omega_1} (-\hat{z} \times \omega_+\hat{z} \times \vec{V}_+ + \hat{z} \times \omega_-\hat{z} \times \vec{V}_-) \\ \Leftrightarrow \vec{\rho} &= \frac{\omega_+\vec{V}_+ - \omega_-\vec{V}_-}{2\omega_1}. \end{aligned} \quad (280)$$

The radial Hamiltonian is given by:

$$H_r = \frac{1}{2}m(\dot{\rho}^2 - \frac{1}{2}\omega_z^2\rho^2) \quad (281)$$

and

$$\begin{aligned} \rho^2 &= \left(\hat{z} \times \frac{\vec{V}_+ - \vec{V}_-}{2\omega_1} \right)^2 \\ \Leftrightarrow \rho^2 &= \frac{1}{4\omega_1^2} (\|\hat{z}\|^2 \|\vec{V}_+ - \vec{V}_-\|^2 + (\hat{z} \cdot (\vec{V}_+ - \vec{V}_-))^2) \\ \Rightarrow \rho^2 &= \frac{1}{4\omega_1^2} \|\vec{V}_+ - \vec{V}_-\|^2 \\ \Leftrightarrow \rho^2 &= \frac{1}{4\omega_1^2} (V_+^2 + V_-^2 - 2\vec{V}_+ \cdot \vec{V}_-) \end{aligned} \quad (282)$$

since the norm of $\hat{z}$ is unity and $\hat{z}$ and $\vec{V}_+ - \vec{V}_-$ are orthogonal. It is straightforward to find that:

$$\dot{\rho}^2 = \frac{1}{4\omega_1^2} (\omega_+^2 V_+^2 + \omega_-^2 V_-^2 - \omega_z^2 \vec{V}_+ \cdot \vec{V}_-). \quad (283)$$

Replacing 282 and 283 in 281 yields equation 29:

$$H_r = \frac{1}{4}m \frac{\omega_+ V_+^2 - \omega_- V_-^2}{\omega_1}. \quad (284)$$

Derivation of the commutation relations

Derivation of equation 30

$$\begin{aligned} [\rho_k, \dot{\rho}_l] &= \rho_k \frac{P_l - eA_l}{m} - \frac{P_l - eA_l}{m} \rho_k \\ \Leftrightarrow [\rho_k, \dot{\rho}_l] &= \frac{1}{m} [\rho_k, P_l] - \frac{e}{m} [\rho_k, A_l] \end{aligned} \quad (285)$$

and:

$$[\rho_k, A_l] = 0, \quad (286)$$

therefore:

$$[\rho_k, \dot{\rho}_l] = \frac{i\hbar}{m} \delta_{kl}. \quad (287)$$

Derivation of equation 31

$$\begin{aligned} m^2 [\dot{\rho}_k, \dot{\rho}_l] &= (P_k - eA_k)(P_l - eA_l) - (P_l - eA_l)(P_k - eA_k) \\ \Leftrightarrow m^2 [\dot{\rho}_k, \dot{\rho}_l] &= [P_k, P_l] + e[A_l, P_k] - e[A_k, P_l] + e^2[A_k, A_l] \\ \Rightarrow m^2 [\dot{\rho}_k, \dot{\rho}_l] &= e[A_l, P_k] - e[A_k, P_l]. \end{aligned} \quad (288)$$

Obviously:

$$\begin{aligned} [\dot{\rho}_x, \dot{\rho}_x] &= 0 \\ [\dot{\rho}_y, \dot{\rho}_y] &= 0 \end{aligned} \quad (289)$$

and we have:

$$\begin{aligned} A_x &= -\frac{B}{2}y \\ A_y &= \frac{B}{2}x, \end{aligned} \quad (290)$$

therefore:

$$\begin{aligned} [\dot{\rho}_x, \dot{\rho}_y] &= \frac{e}{m^2} ([A_y, P_x] - [A_x, P_y]) \\ \Leftrightarrow [\dot{\rho}_x, \dot{\rho}_y] &= \frac{eB}{2m^2} ([x, P_x] - [-y, P_y]) \\ \Leftrightarrow [\dot{\rho}_x, \dot{\rho}_y] &= \frac{\omega_c}{2m} (i\hbar + i\hbar) \\ \Leftrightarrow [\dot{\rho}_x, \dot{\rho}_y] &= i \frac{\hbar \omega_c}{m}. \end{aligned} \quad (291)$$

Similarly:

$$[\dot{\rho}_y, \dot{\rho}_x] = -i \frac{\hbar \omega_c}{m}. \quad (292)$$

Derivation of equation 32

$$\begin{aligned} [V_{+,x}, V_{-,x}] &= (\dot{\rho}_x - \omega_- \rho_y)(\dot{\rho}_x - \omega_+ \rho_y) - (\dot{\rho}_x - \omega_+ \rho_y)(\dot{\rho}_x - \omega_- \rho_y) \\ \Leftrightarrow [V_{+,x}, V_{-,x}] &= \dot{\rho}_x^2 - \omega_+ \dot{\rho}_x \rho_y - \omega_- \rho_y \dot{\rho}_x + \omega_- \omega_c' \rho_y^2 - \dot{\rho}_x^2 + \omega_- \dot{\rho}_x \rho_y + \omega_+ \rho_y \dot{\rho}_x - \omega_+ \omega_- \rho_y^2 \\ \Leftrightarrow [V_{+,x}, V_{-,x}] &= (\omega_+ - \omega_-)[\rho_y, \dot{\rho}_x] \\ \Leftrightarrow [V_{+,x}, V_{-,x}] &= 0. \end{aligned} \quad (293)$$

Similarly,

$$[V_{+,y}, V_{-,y}] = 0. \quad (294)$$

$$\begin{aligned} [V_{+,x}, V_{-,y}] &= (\dot{\rho}_x - \omega_- \rho_y)(\dot{\rho}_y + \omega_+ \rho_x) - (\dot{\rho}_y + \omega_+ \rho_x)(\dot{\rho}_x - \omega_- \rho_y) \\ \Leftrightarrow [V_{+,x}, V_{-,y}] &= [\dot{\rho}_x, \dot{\rho}_y] - \omega_+ [\rho_x, \dot{\rho}_x] - \omega_- [\rho_y, \dot{\rho}_y] \\ \Leftrightarrow [V_{+,x}, V_{-,y}] &= i \frac{\hbar \omega_c}{m} - i \frac{\hbar \omega_+}{m} - i \frac{\hbar \omega_-}{m} \\ \Leftrightarrow [V_{+,x}, V_{-,y}] &= 0. \end{aligned} \quad (295)$$

Similarly,

$$[V_{+,y}, V_{-,x}] = 0. \quad (296)$$

Derivation of equation 33

$$\begin{aligned} [V_{+,x}, V_{+,y}] &= (\dot{\rho}_x - \omega_- \rho_y)(\dot{\rho}_y + \omega_- \rho_x) - (\dot{\rho}_y + \omega_- \rho_x)(\dot{\rho}_x - \omega_- \rho_y) \\ \Leftrightarrow [V_{+,x}, V_{+,y}] &= [\dot{\rho}_x, \dot{\rho}_y] - \omega_- [\rho_x, \dot{\rho}_x] - \omega_- [\rho_y, \dot{\rho}_y] \\ \Leftrightarrow [V_{+,x}, V_{+,y}] &= i \frac{\hbar \omega_c}{m} - i \frac{\hbar \omega_-}{m} - i \frac{\hbar \omega_-}{m} \\ \Leftrightarrow [V_{+,x}, V_{+,y}] &= i \frac{2\hbar \omega_1}{m} \end{aligned} \quad (297)$$

and

$$\begin{aligned}
[V_{-,x}, V_{-,y}] &= (\dot{\rho}_x - \omega_+ \rho_y)(\dot{\rho}_y + \omega_+ \rho_x) - (\dot{\rho}_y + \omega_+ \rho_x)(\dot{\rho}_x - \omega_+ \rho_y) \\
\Leftrightarrow [V_{-,x}, V_{-,y}] &= [\dot{\rho}_x, \dot{\rho}_y] - \omega_+ [\rho_x, \dot{\rho}_x] - \omega_+ [\rho_y, \dot{\rho}_y] \\
\Leftrightarrow [V_{-,x}, V_{-,y}] &= i \frac{\hbar \omega_c}{m} - i \frac{\hbar \omega_+}{m} - i \frac{\hbar \omega_+}{m} \\
\Leftrightarrow [V_{-,x}, V_{-,y}] &= -i \frac{2\hbar \omega_1}{m}.
\end{aligned} \tag{298}$$

A.3 Ion Coulomb Crystals

Two-ion chain

Here we calculate the frequencies of the normal modes of a two-ion chain. For the general case of N ions, the potential at equilibrium is:

$$\mathcal{V} = \sum_{j=1}^N \frac{1}{2} m \omega_z^2 z_j^2 + \frac{e^2}{8\pi\epsilon_0} \sum_{k=1, k \neq j}^N \frac{1}{|z_k - z_j|}. \tag{299}$$

We calculate the partial derivatives:

$$\begin{aligned}
\frac{\partial^2 \mathcal{V}}{\partial z_j^2} &= m \omega_z^2 + \frac{e^2}{2\pi\epsilon_0} \sum_{k=1, k \neq j}^N \frac{1}{|z_k - z_j|^3} \\
\Leftrightarrow \frac{\partial^2 \mathcal{V}}{\partial z_j^2} &= m \omega_z^2 + \frac{e^2}{2\pi\epsilon_0 (l_z)^3} \sum_{k=1, k \neq j}^N \frac{1}{|w_k - w_j|^3} \\
\Leftrightarrow \frac{\partial^2 \mathcal{V}}{\partial z_j^2} &= m \omega_z^2 \left(1 + 2 \sum_{k=1, k \neq j}^N \frac{1}{|w_k - w_j|^3} \right)
\end{aligned} \tag{300}$$

and similarly,

$$\frac{\partial^2 \mathcal{V}}{\partial z_j \partial z_k} = -m \omega_z^2 \frac{2}{|w_k - w_j|^3}, \quad j \neq k. \tag{301}$$

We know the equilibrium positions for two ions:

$$w_j^0 = \pm \left(\frac{1}{2} \right)^{2/3} \tag{302}$$

so we can construct the matrix K^z

$$\begin{aligned}
K^z &= \omega_z^2 \begin{pmatrix} 1 + \frac{2}{|w_1^0 - w_2^0|^3} & -\frac{2}{|w_2^0 - w_1^0|^3} \\ -\frac{2}{|w_1^0 - w_2^0|^3} & 1 + \frac{2}{|w_2^0 - w_1^0|^3} \end{pmatrix} \\
\Rightarrow K^z &= \omega_z^2 \begin{pmatrix} 2 & -1 \\ -1 & 2 \end{pmatrix}.
\end{aligned} \tag{303}$$

To find the eigenvalues of K^z , we calculate the determinant of $K^z - \mu I$ where I is the identity matrix and find its roots. Those are $\mu = \omega_z^2$ and $\mu = 3\omega_z^2$. Finding the eigenvectors and normal modes' frequencies from there is trivial.

Planar crystal

Here we show how to obtain the system of equations to determine the equilibrium positions of the ions in a planar crystal as we as how to construct the matrix K^ρ to find the frequencies of the normal modes. Starting from the expression of the potential in the rotating frame at equilibrium, taking into account the ions lie in the radial plane:

$$\mathcal{V}^R = \sum_{j=1}^N \frac{1}{2} m \omega_{eff}^2 (\rho_J^R)^2 + \frac{e^2}{8\pi\epsilon_0} \sum_{k=1, k \neq j}^N \frac{1}{r_{jk}}, \quad (304)$$

we calculate the derivative with respect to x_j^R (it is similar for y_j^R):

$$\frac{\partial \mathcal{V}^R}{\partial x_j^R} = m \omega_{eff}^2 x_j^R - \frac{e^2}{4\pi\epsilon_0} \sum_{k=1, k \neq j} \frac{x_j^R - x_k^R}{((x_j^R - x_k^R)^2 + (y_j^R - y_k^R)^2)^{3/2}} \quad (305)$$

or else, in terms of u_j and v_j :

$$\frac{\partial \mathcal{V}^R}{\partial x_j^R} = m \omega_{eff}^2 \ell_\rho u_j - \frac{e^2}{4\pi\epsilon_0 (\ell_\rho)^2} \sum_{k=1, k \neq j} \frac{u_j - u_k}{((u_j - u_k)^2 + (v_j - v_k)^2)^{3/2}}. \quad (306)$$

Equating this equation to zero gives the first line of the system 61:

$$u_j - \sum_{k=1, k \neq j}^N \frac{u_j - u_k}{((u_j - u_k)^2 + (v_j - v_k)^2)^{3/2}} = 0. \quad (307)$$

The second line is found the same way by deriving with respect to y_j^R .

In order to construct the matrix K^ρ , we need to derive the potential with respect to z_j^R so we start from the full expression of the equilibrium potential:

$$\mathcal{V}^R = \sum_{j=1}^N \frac{1}{2} m \omega_z^2 (z_j^R)^2 + \frac{1}{2} m \omega_{eff}^2 (\rho_J^R)^2 + \frac{e^2}{8\pi\epsilon_0} \sum_{k=1, k \neq j}^N \frac{1}{r_{jk}}. \quad (308)$$

We first derive the second derivatives of the potential and then simplifies the expressions by considering that the ions are in the radial plane and so that their axial coordinate is zero. We can make this approximation considering the motion of the ions in the axial direction is small compared to the spacing between the ions. We

obtain:

$$\begin{aligned} \left[\frac{\partial^2 \mathcal{V}^R}{(\partial z_j^R)^2} \right]_0 &= m\omega_z^2 - m\omega_{eff}^2 \sum_{k=1, k \neq j} \frac{1}{((u_j - u_k)^2 + (v_j - v_k)^2)^{3/2}} \\ \left[\frac{\partial^2 \mathcal{V}^R}{\partial z_j^R \partial z_k^R} \right]_0 &= m\omega_{eff}^2 \frac{1}{((u_j - u_k)^2 + (v_j - v_k)^2)^{3/2}} \quad , \quad k \neq j \end{aligned} \quad (309)$$

The elements of k^ρ are found by dividing these derivatives by m .

A.4 Motional Superposition

The probability of excitation given by equation 244 is derived here. The ion is initially in the motional and electronic ground state $|\psi\rangle = |g, 0\rangle$. The first pulse on the carrier with a duration $\tau = 2 \arcsin(\sqrt{2/2})/\Omega_{0,0}$ couples $|e, 0\rangle$ and $|g, 0\rangle$ and the corresponding propagator is:

$$T_0^0 = \frac{1}{\sqrt{3}} \begin{pmatrix} 1 & -i\sqrt{2} \\ -i\sqrt{2} & 1 \end{pmatrix}. \quad (310)$$

Thus after the first pulse:

$$|\psi\rangle = -i\sqrt{\frac{2}{3}}|e, 0\rangle + \frac{1}{\sqrt{3}}|g, 0\rangle. \quad (311)$$

The second pulse is a $\pi/2$ on the first red sideband affecting the population in $|e, 0\rangle$ and $|g, 1\rangle$. We have:

$$T_1^{-1} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & e^{i\phi_2} \\ -e^{-i\phi_2} & 1 \end{pmatrix} \quad (312)$$

which yields

$$|\psi\rangle = \frac{1}{\sqrt{3}}(-i|e, 0\rangle + |g, 0\rangle + ie^{-i\phi_2}|g, 1\rangle). \quad (313)$$

A π pulse then transfers the population in $|e, 0\rangle$ to $|g, 2\rangle$.

$$T_2^{-2} = \begin{pmatrix} 0 & ie^{i\phi_3} \\ ie^{-i\phi_3} & 0 \end{pmatrix}. \quad (314)$$

The resulting wave function describes an equal superposition of the first three motional states, in the ground electronic state:

$$|\psi\rangle = \frac{1}{\sqrt{3}}(|g, 0\rangle + ie^{-i\phi_2}|g, 1\rangle + e^{-i\phi_3}|g, 2\rangle). \quad (315)$$

The ion then evolves freely for a duration T . The wave function becomes:

$$|\psi\rangle = \frac{1}{\sqrt{3}} (|g, 0\rangle + ie^{-i(\phi_2+\omega_z T)} |g, 1\rangle + e^{-i(\phi_3+2\omega_z T)} |g, 2\rangle). \quad (316)$$

After the second pulse on the second red sideband, we have:

$$|\psi\rangle = \frac{1}{\sqrt{3}} (ie^{i(\phi_4-\phi_3-2\omega_z T)} |e, 0\rangle + |g, 0\rangle + ie^{-i(\phi_2+\omega_z T)} |g, 1\rangle). \quad (317)$$

And after the second pulse on the first red sideband:

$$\begin{aligned} |\psi\rangle = & \frac{i}{\sqrt{6}} (e^{i(\phi_5-\phi_2-\omega_z T)} + e^{i(\phi_4-\phi_3-2\omega_z T)}) |e, 0\rangle \\ & + \frac{1}{\sqrt{3}} |g, 0\rangle \\ & + \frac{i}{\sqrt{6}} (e^{-i(\phi_2+\omega_z T)} - e^{i(\phi_4-\phi_3-\phi_5-2\omega_z T)}) |g, 1\rangle. \end{aligned} \quad (318)$$

The last pulse on the carrier couples $|e, 0\rangle$ with $|g, 0\rangle$ and $|e, 1\rangle$ with $|g, 1\rangle$. Because the state $|g, 1\rangle$ is populated, the probabilities for the ion to be in $|e, 0\rangle$ and $|e, 1\rangle$ have to be summed to calculate the total probability of excitation. The coefficient associated with $|e, 0\rangle$ is:

$$c_{e,0}(T) = \frac{i}{3} \left(\frac{1}{\sqrt{2}} (e^{i(\phi_5-\phi_2-\omega_z T)} + e^{i(\phi_4-\phi_3-2\omega_z T)}) - \sqrt{2} e^{i\phi_6} \right). \quad (319)$$

Thus the probability of the ion being in $|e, 0\rangle$ is:

$$\begin{aligned} |c_{e,0}(T)|^2 = & \frac{1}{9} \left(3 + \cos(\phi_3 + \phi_5 - \phi_2 - \phi_4 + \omega_z T) \right. \\ & \left. - 2 \cos(\phi_5 - \phi_2 - \phi_6 - \omega_z T) - 2 \cos(\phi_4 - \phi_3 - \phi_6 - 2\omega_z T) \right). \end{aligned} \quad (320)$$

The length of the pulse is calibrated for the carrier transition at the motional ground state, however $\Omega_{1,1} \simeq \Omega_{0,0}$ so we make the approximation that the propagator governing the transition between $|e, 1\rangle$ and $|g, 1\rangle$ is the same as that for the one between $|e, 0\rangle$ and $|g, 0\rangle$:

$$T_1^0 = \frac{1}{\sqrt{3}} \begin{pmatrix} 1 & -i\sqrt{2}e^{i\phi_6} \\ -i\sqrt{2}e^{-i\phi_6} & 1 \end{pmatrix}. \quad (321)$$

We find:

$$c_{e,1}(T) = \frac{1}{3} e^{i\phi_6} (e^{-i(\phi_2+\omega_z T)} - e^{i(\phi_4-\phi_3-\phi_5-2\omega_z T)}) \quad (322)$$

and the associated probability:

$$|c_{e,1}(T)|^2 = \frac{1}{9} \left(2 - 2 \cos (\phi_3 + \phi_5 - \phi_2 - \phi_4 + \omega_z T) \right). \quad (323)$$

Introducing $\Phi_1 = \phi_6 + \phi_2 - \phi_5$ and $\Phi_2 = \phi_6 + \phi_3 - \phi_4$, the total probability of excitation is:

$$P_e(T) = \frac{1}{9} \left(5 - \cos (\omega_z T + \Phi_2 - \Phi_1) - 2 \cos (\omega_z T + \Phi_1) - 2 \cos (2\omega_z T + \Phi_2) \right). \quad (324)$$

B

Planar Coulomb Crystals

B.1 Equilibrium Positions of Planar ICC

The tables below give the normalised equilibrium positions of ions in planar crystals. These were calculated numerically using the software Mathematica 10.0 except for the two-ion crystal where the positions were derived analytically. The configuration is found by finding a root of the system of equations and calculating the associated potential energy. The operation is repeated many times and the lowest energy configuration is retained. There is no guarantee to find an absolute minimum but the large number of repetitions (up to ten thousand) and the good agreement with experiments make the results relevant. The positions are given in units of ℓ_ρ defined as

$$\ell_\rho = \left(\frac{e^2}{4\pi\epsilon_0 m \omega_{eff}} \right)^{1/3} \quad (325)$$

2	3	4	5
(0, 0.62996) (0, -0.62996)	(0, -0.83268) (-0.72112, 0.41634) (0.72112, 0.41634)	(0, 0.98549) (0.98549, 0) (0, -0.98549) (-0.98549, 0)	(0, 1.11236) (-1.05792, 0.34374) (-0.65383, -0.89992) (0.65383, -0.89992) (1.05792, 0.34374)
6	7	8	9
(0, -1.33446) (0, 0) (-1.26915, -0.41237) (-0.78438, 1.07960) (1.26915, -0.41237) (0.78438, 1.07960)	(0, 1.41403) (-1.22459, -0.70702) (0, 0) (0, -1.41403) (1.22459, -0.70702) (1.22459, 0.70702) (-1.22459, 0.70702)	(0, -1.48952) (-1.45218, 0.33145) (-0.64628, 1.34201) (1.16456, -0.92870) (-1.16456, -0.92870) (0, 0) (1.45218, 0.33145) (0.64628, 1.34201)	(0, -0.56159) (-1.28992, 0.97936) (0, 0.52727) (0, 1.74132) (1.50386, -0.31842) (-0.80118, -1.51444) (0.80118, -1.51444) (1.28992, 0.97936) (-1.50386, -0.31842)
10	11	12	13
(0, 1.79773) (0, -1.79773) (1.23567, 1.18547) (-1.57501, 0) (-1.23567, -1.18547) (0, 0.53734) (1.57501, 0) (1.23567, -1.18547) (-1.23567, 1.18547) (0, -0.53734)	(0, 1.80060) (-0.26124, -0.65842) (1.23571, -1.21559) (1.91361, 0.00434) (-1.39010, 1.31512) (-1.32945, -1.31656) (1.23449, 1.31080) (-1.73298, -0.03772) (-0.43156, 0.55742) (0.69633, 0.10991) (0.06519, -1.86990)	(0, 1.75391) (1.22716, 1.50061) (-1.51893, -0.87695) (-1.91315, 0.31244) (1.51893, -0.87695) (-0.68599, -1.81305) (0.61034, 0.35238) (0, -0.70477) (-1.22716, 1.50061) (1.91315, 0.31244) (-0.61034, 0.35238) (0.68599, -1.81305)	(0, 0.83491) (-1.93803, 0.58680) (2.05830, 0.14051) (0.50185, -1.97390) (-0.89238, -1.76025) (1.54822, -1.15167) (0.83538, 0.00257) (0.23471, 2.04535) (1.38286, 1.32631) (-1.81084, -0.82054) (-1.09043, 1.60369) (0.00311, -0.83321) (-0.83275, -0.00056)
14	15	16	17
(0, 0.83965) (-2.11810, 0) (-0.82220, 0) (-1.57670, -1.15179) (0.65566, 1.96460) (1.57670, -1.15179) (0.65566, -1.96460) (-1.57670, 1.15179) (2.11810, 0) (0, -0.83965) (0.82220, 0) (-0.65566, -1.96460) (-0.65566, 1.96460) (1.57670, 1.15179)	(0, 2.08386) (1.21052, 1.83029) (-0.37173, -0.74349) (1.63827, -1.40954) (-2.00152, 0.89103) (1.69123, 0.77526) (0.75322, -0.33531) (0.55569, -1.77552) (-1.08182, 1.67751) (2.14374, -0.27410) (-1.97049, -0.31871) (-1.54848, -1.39452) (0.30372, 0.77377) (-0.77236, 0.34383) (-0.54998, -2.12435)	(0, -1.14780) (-1.25748, -1.73077) (-2.20299, -0.71580) (-2.03464, 0.66110) (1.36152, 1.87398) (0, -2.31636) (1.25748, -1.73077) (2.03464, 0.66110) (-0.67466, 0.92859) (-1.36152, 1.87398) (1.09162, -0.35469) (0, 2.13935) (-1.09162, -0.35469) (0.67466, 0.92859) (2.20299, -0.71580) (0, 0)	(0, -2.36240) (0, 2.36240) (1.34196, 1.82656) (0, 0) (1.31815, -1.89153) (-2.25441, -0.70609) (-1.34196, -1.82656) (-0.98817, 0.72601) (0.13131, 1.21216) (-1.31815, 1.89153) (2.19905, -0.69254) (1.11750, 0.48761) (0.98817, -0.72601) (-2.19905, 0.69254) (-0.13131, -1.21216) (2.25441, 0.70609) (-1.11750, -0.48761)

Table B.1: Equilibrium coordinates (x,y) of planar Coulomb crystals in the radial plane.

B.2 Axial Modes of Planar ICC

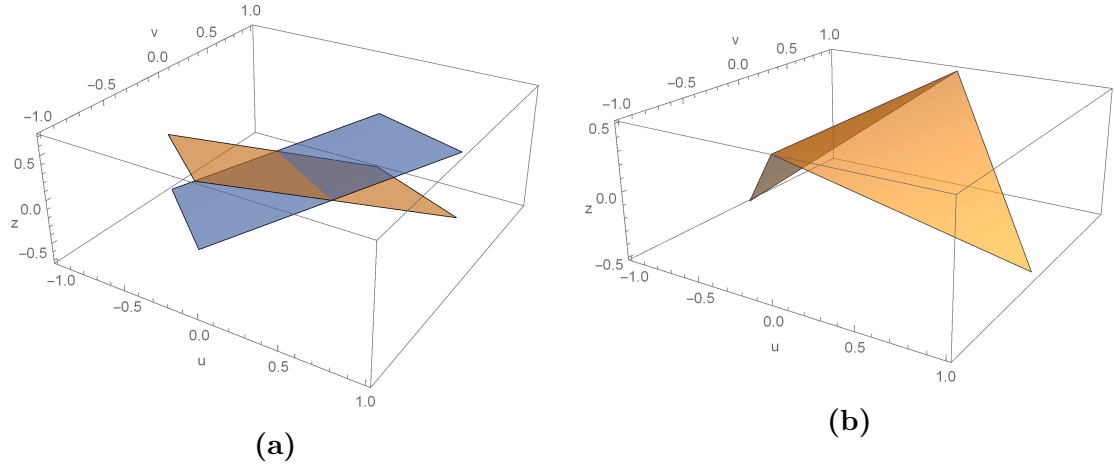


Figure B.1: Representation of the axial modes (except COM) of a four-ion planar crystal. (a) Modes 2 and 3 (degenerate). (b) Mode 4.

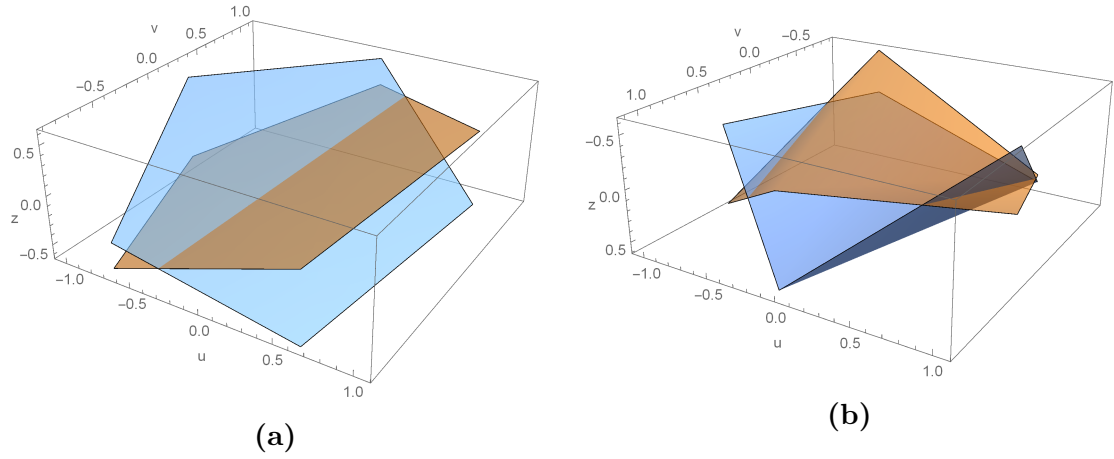


Figure B.2: Representation of the axial modes (except COM) of a five-ion planar crystal. (a) Modes 2 and 3 (*tilt*, degenerate). (b) Modes 4 and 5 (*folding*, degenerate).

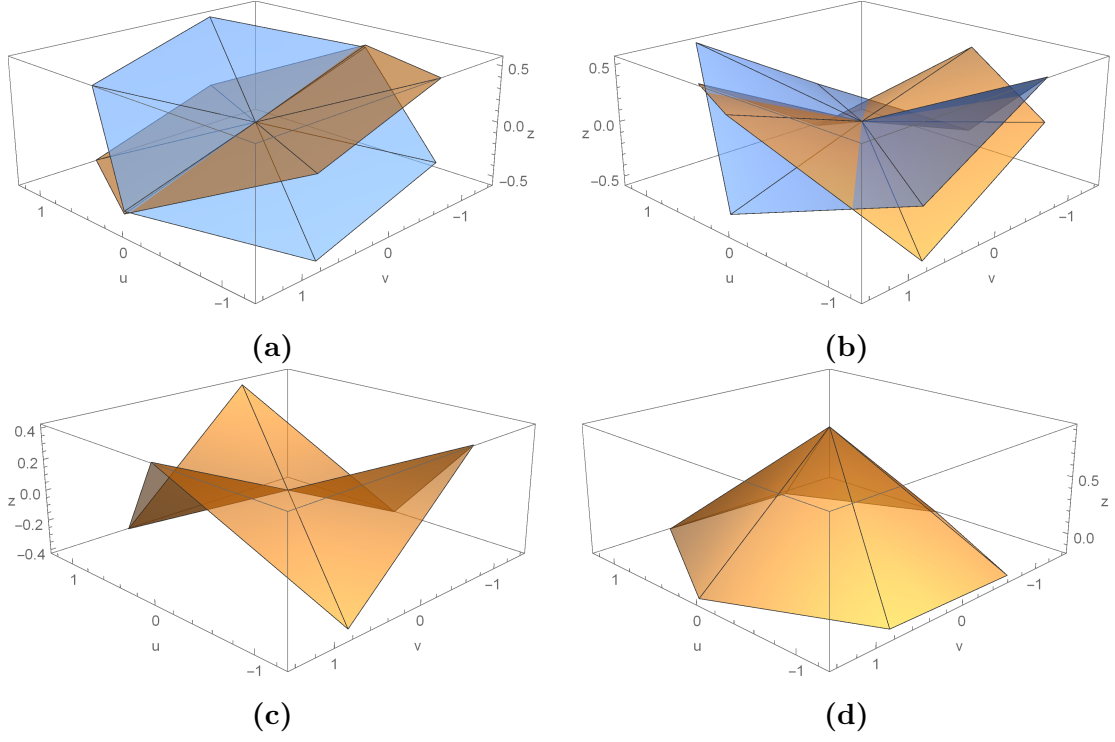


Figure B.3: Representation of the axial modes (except COM) of a seven-ion planar crystal. (a) Modes 2 and 3 (degenerate). (b) Modes 4 and 5 (degenerate). (c) Mode 6. (d) Mode 7.

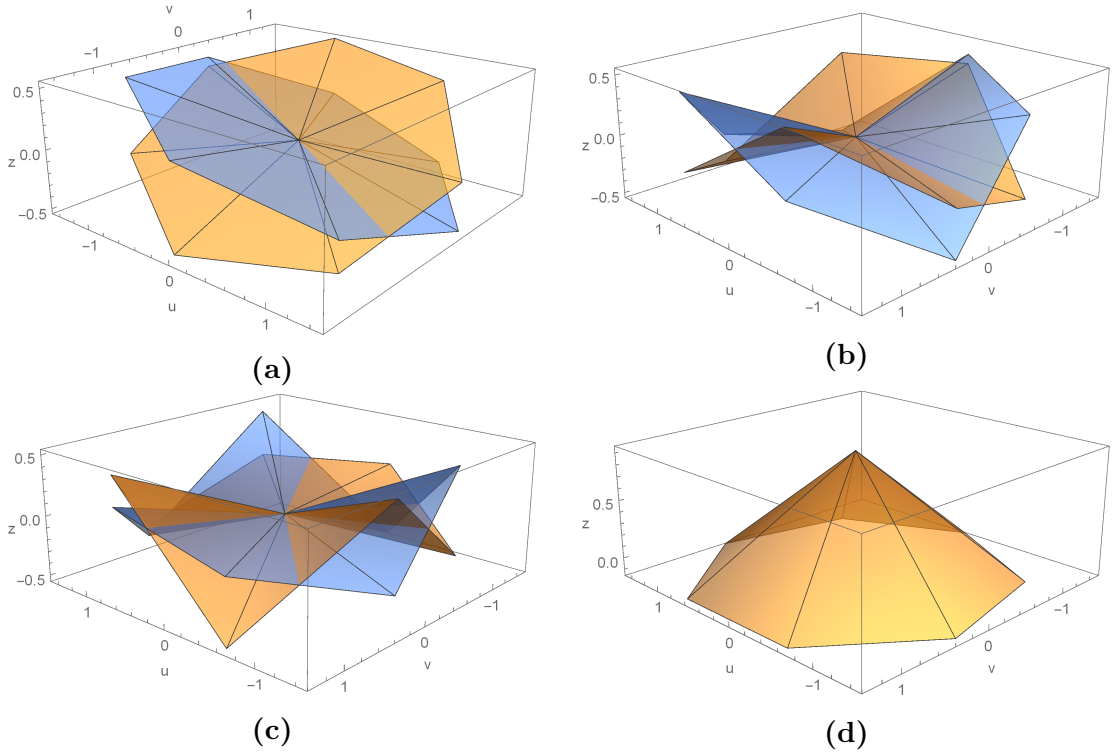


Figure B.4: Representation of the axial modes (except COM) of a eight-ion planar crystal. (a) Modes 2 and 3 (degenerate). (b) Modes 4 and 5 (degenerate). (c) Mode 6 and 7 (degenerate). (d) Mode 8.

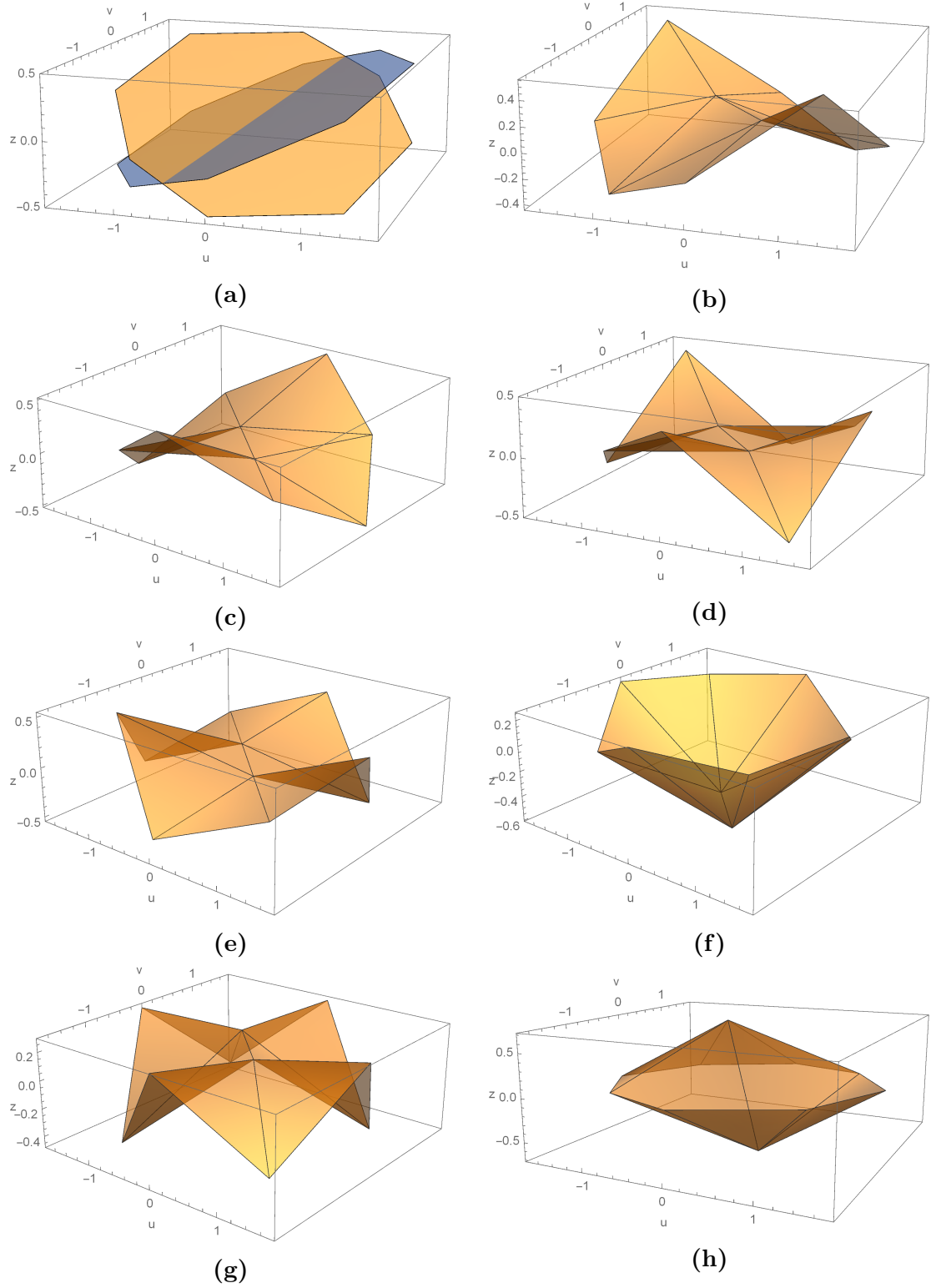
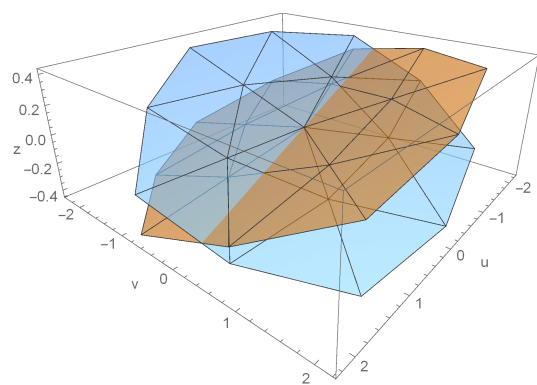
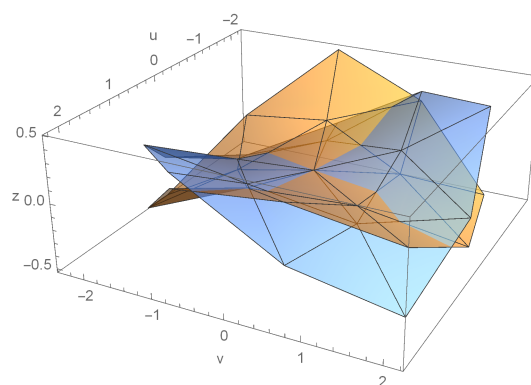


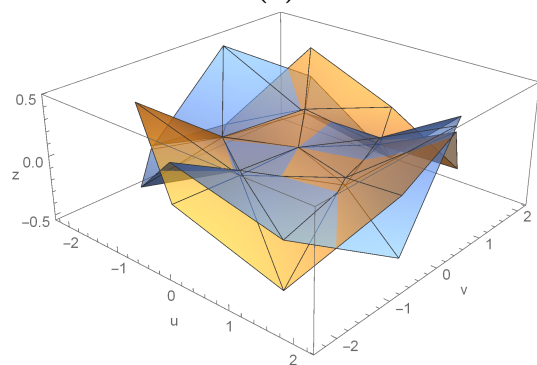
Figure B.5: Representation of the axial modes (except COM) of a ten-ion planar crystal. (a) Modes 2 and 3 (degenerate). (b) Mode 4. (c) Mode 5. (d) Mode 6. (e) Mode 7. (f) Mode 8. (g) Mode 9. (h) Mode 10.



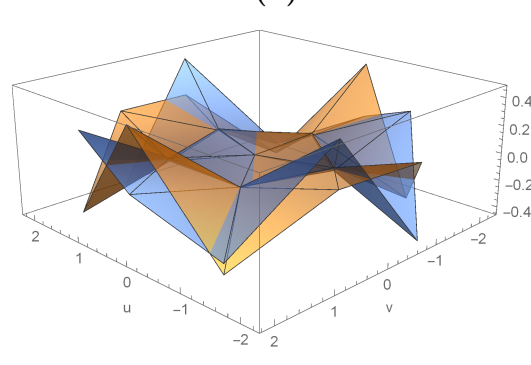
(a)



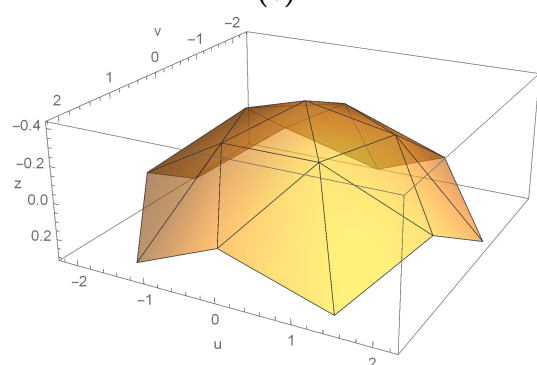
(b)



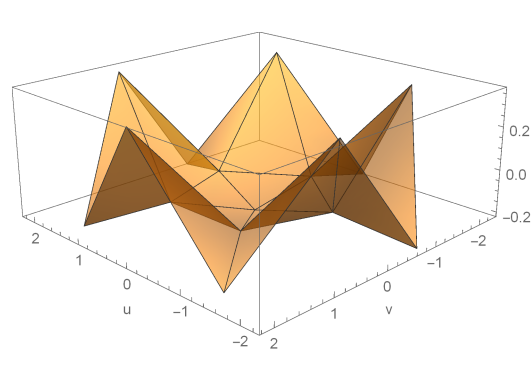
(c)



(d)



(e)



(f)

Figure B.6

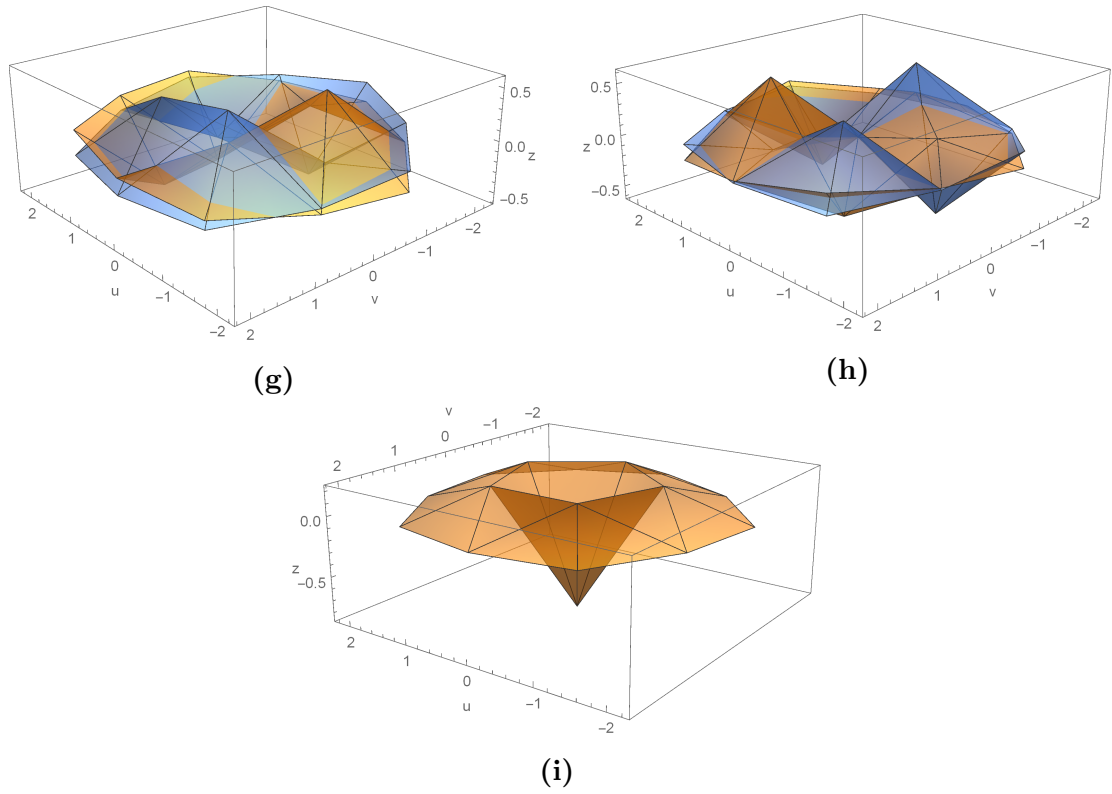
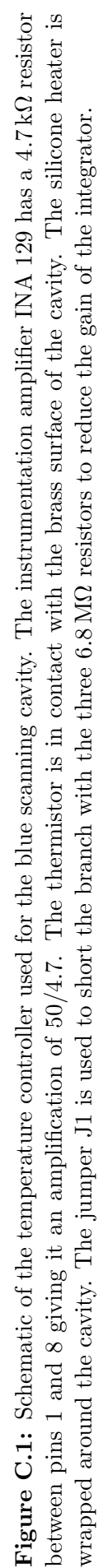


Figure B.6: Representation of the axial modes (except COM) of a sixteen-ion planar crystal. **(a)** Modes 2 and 3 (degenerate). **(b)** Mode 4 and 5 (degenerate). **(c)** Mode 6 and 7 (degenerate). **(d)** Mode 8 and 9 (degenerate). **(e)** Mode 10. **(f)** Mode 11. **(g)** Mode 12 and 13 (degenerate). **(h)** Mode 14 and 15 (degenerate). **(i)** Mode 16

C

Electronic Circuits



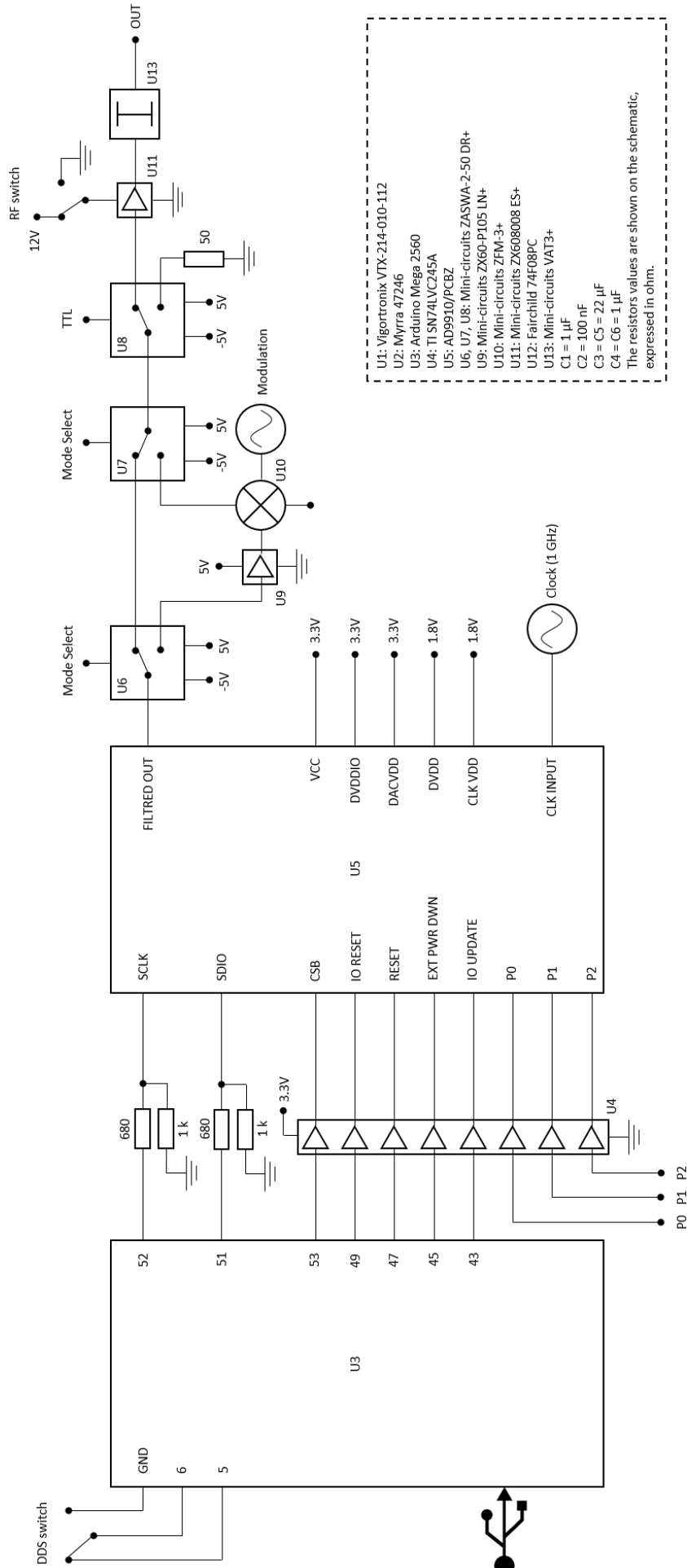


Figure C.3: 729 AOM driver MkII. Control and radio-frequency. NB: a second stage amplification is performed by an external RF amplifier not represented on this schematic.

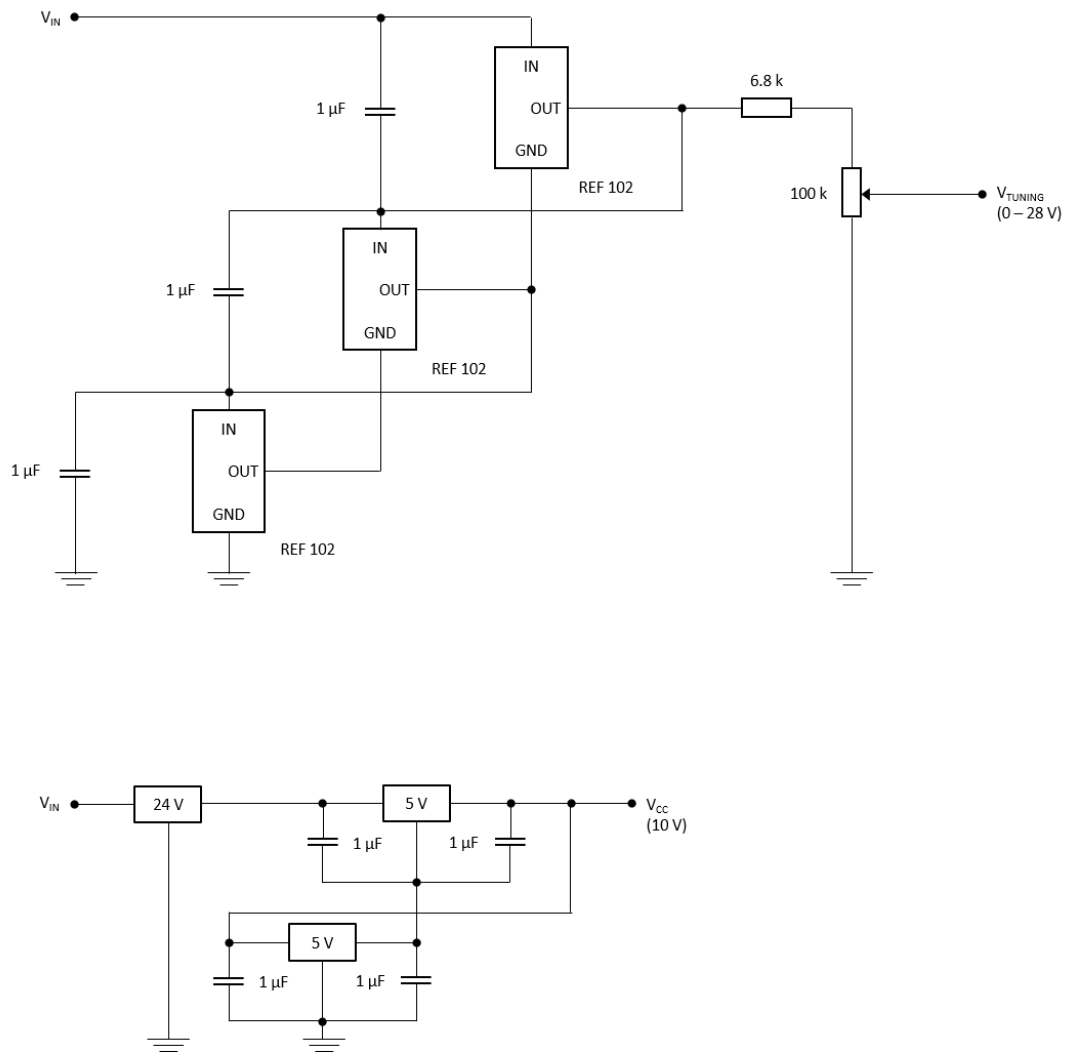


Figure C.4: Power supply of the VCO used for one of the RF signals sent to the EOM.

