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UNIVERSITY OF CALIFORNIA

Los Angeles

Electric-field Gradients for Molecular Qubits and Quantum Sensing

A dissertation submitted in partial satisfaction

of the requirements for the degree

Doctor of Philosophy in Physics

by

Grant David Mitts

2026

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ABSTRACT OF THE DISSERTATION

Electric-field Gradients for Molecular Qubits and Quantum Sensing

by

Grant David Mitts

Doctor of Philosophy in Physics

University of California, Los Angeles, 2026

Professor Eric R. Hudson, Chair

This dissertation focuses on implementing electric-field gradients into a cryogenic ion trap for purposes related to quantum computing and sensing. The principal goal of this experiment is to introduce electric-field gradients as a new platform for molecular ion quantum logic. The work covered in this document discusses the progress made towards performing molecular spectroscopy on parity transitions within the hyperfine manifold of HCl^+ . A byproduct of this research has been the introduction of motional Raman transitions. Our research into this phenomenon demonstrates its application as a wideband, phase-sensitive electric field sensing scheme and, through quantum amplification, its ability to sense below the Standard Quantum Limit. The motional Raman scheme is also investigated as a nonlinear process, where probing a signal at subharmonics of a motional frequency can result in sensing below the linear Fourier Transform Limit. Sensing below the Fourier Transform Limit is further investigated in the context of two closely separated tones. By applying a single phase reversal during the pulse sequence, we demonstrate super-resolution of two 80 MHz tones separated 5 Hz. The final focus of this dissertation is on implementing quantum techniques into mass spectrometry. Specifically, we demonstrate how CAT interferometry can be used to perform a high-precision mass measurement on an H^{35}Cl^+ ion.

The dissertation of Grant David Mitts is approved.

Kenneth R. Brown

David R. Leibrandt

Wesley C. Campbell

Eric R. Hudson, Committee Chair

University of California, Los Angeles

2026

To my family, friends, and all the ions who died during the making of this PhD.

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ACKNOWLEDGMENTS

There exist several purposes in writing a PhD dissertation. Deep down we all want it to have some grandiose meaning, for example having it become the foremost resource that younger grad students go to in order to learn the methods and techniques that will allow them to quickly and efficiently build an experiment of their own. I think of this type as the cookbook. It will lay out the fundamentals of the experiment from first principle and include all the relevant bits and bobbles about what did what and what knobs the future grad students need to turn to get everything behaving properly. This would obviously be the goal but what we discover in the many years needed to build and run the experiment is that things don't go as expected and that makes the thesis a much more complex thing to write. The chronological order of events isn't the order that will make sense to readers because often the order gets jumbled up in the process of proving the initial hypothesis. Instead, along the way we uncover things by accident and have to figure out a way to make it feel like to readers that this was an intentional discovery through a deeper understanding that was not initially there. I like to express this as one of two philosophies about what makes good science.

This will be a brief monologue about two ways of defining the perfect science experiment. In one camp, I will affirm my own personal belief that the best experiment is one that went perfectly, or is one that follows the theory that outlines the experiment, as was the initial goal of my experiment. The other, a belief held strongly by my friend Brian Bostwick, is one where the best science is not by experimentally confirming a theory but instead is by observing a phenomenon and being able to describe it completely from first principles. What happened in this experiment was most definitely an attempt at the former that led us down the rabbit hole to the latter. Amid the development of EGGs came Green EGGs, leading to motional Raman in its various forms.

My advisor Eric falls into the same camp as my friend Brian and has expressed his enjoyment of the various endeavors we've gone on that were not EGGs. I was fortunate to have been able to join Eric's group as an undergrad the fall of my junior year. During my sophomore year, I became friends with P.J. Robinson, who had

been doing theoretical physical chemistry research since his freshman year and who told me that I needed to get involved in research. Knowing I liked the idea of laser-cooling, I looked at which professors did this and learned about Eric’s MOTION experiment through his website. Having never met Eric before, the obvious option was to email him. After several failed attempts at starting an email conversation with him, I decided I had to knock on his door. His response was “I was waiting for an undergrad that had the guts to do something like this.”

The second way in which I was extremely fortunate was that Eric put me under the wing of Elizabeth West. She was just starting her post-doctoral research on velocity map imaging, and could use an extra pair of hands. She helped guide me throughout my undergraduate experience in this group and was the truest definition of a mentor. Without her wonderful personality and patience, I may not have pursued graduate school. Under her supervision, not only was I able to learn more about AMO physics and building an experiment, but she also facilitated my integration into the larger UCLA AMO community, having me present in group meeting and in journal club.

I am thankful to the many generations of UCLA AMO students and staff that I have had the pleasure of meeting. When your tenure in a research group is 9 years, you really get to meet some amazing people. After choosing UCLA for grad school, Eric shifted me onto the MOTION experiment where I got to work with Prateek Puri and Mike Mills, who both graciously invited me into the ranks of MOTION researchers albeit my time was some of the shortest. Working on trying to drive a molecular transition within BaCl^+ with Mike taught me an important lesson, and that is to be superstitious, ESPECIALLY with molecular spectroscopy. After thinking we saw indication of a transition, Mike had a celebratory beer (not in lab of course...). It wasn’t something we could repeat afterwards, and I believe this curse followed me to EGGs.

The advent of EGGs came from Eric and Wes during my hiatus from lab, a.k.a. the first year of grad classes. Upon my return, Eric informed me that I would be building the experiment and asked if I thought I could work well with the other UCLA undergrad turned grad, Clayton Ho. I said “yeah, probably”, not understanding the

gravitas of what it meant to build an experiment with another person.

There are so many ways in which to describe Clayton, and yet he always feels like an enigma. His mannerisms have always been rather outlandish and contagious, such as introducing the phrase “hateful” into my vocabulary or the Atlas pose into the UCLA AMO community. And while we all have our Atlas moments, none of us can carry the weight of the world on his shoulders like Clayton can. As researchers, we operate very differently, not unlike the sun and the moon. I use this metaphor since there have been long periods of our tenure when I operate the day shift (9 a.m. to 7 p.m.) and Clayton the night shift (2 p.m. to 2 a.m.). I prefer light mode, whereas Clayton sets the computers to dark mode. I focused on building the vacuum chamber and Clayton captained the simulations and coded the entire experiment (I wrote a single labrad code to a-ramp and that was it). The term bus factor indicates how replaceable someone is in the event that they get hit by a bus. To understand Clayton’s bus factor, look at Fig. [2.2](#).

But luckily for us, we had Josh Rabinowitz join our experiment. With his prior experience with ARTIQ and desire to work with molecular ions, he was the perfect candidate to pick up the mantle. Josh has also been keen on getting into the California lifestyle and we have had numerous pre-work surf sessions. I have indefinitely loaned him a wavestorm that was gifted to me from Matt Boguslawski, who was a postdoc on the barium experiment during my first couple of years of grad school. It has been enjoyable having watched Josh nosedive in Venice but, unlike with surfing, he was extremely fast at integrating himself into our experiment. The initial project we tasked him with was building the 729nm filter cavity + TA system, so sideband cooling operations could be faster and have far less noise. This upgrade has been huge for the experiment, and Josh has had many great ideas for current and future techniques that can be implemented into the experiment. The only challenge working with Josh is that he likes country music.

The last member of our group that I must acknowledge is Hao Wu. Hao and I have had the longest overlap in lab, as he began his postdoc here working on the MOTION experiment. When Eric tasked me with designing the EGGs cryogenic system, Hao was able to answer many of the questions that I had (I must also thank

Elizabeth and Guozhu Zhu for their cryogenic knowledge). But it wasn't until after we loaded ions for the first time in our trap that Hao became OUR postdoc. Since then he has been an integral member of the experiment, coming up with new avenues for the experiment, providing insight into molecules, and keeping us caught up with relevant publications for techniques we should be aware of. We have been extremely fortunate to have Hao working with us and guiding the direction of EGGs.

To finish up, I have been very fortunate to have received so much guidance from those around me, near and far. In Eric's group, NICER (Guozhu Zhu and Guanming Lao), Barium (Dave Hucul, Justin Christensen, Matt Boguslawski, Zach Wall, Sam Vizvary, Michael Bareian, Steven Diaz, Elijah Mossman, and Jiaxiang Wang), Thorium (Christian Schneider, Justin Jeet, Ricky Elwell, James Terhune, and Alexandros Androutsos). In Wes Campbell's group, Ytterbium A-level (Tony Ransford, Conrad Roman, Thomas Dellert, and Drew Parks), Ytterbium B-level (Patrick McMillin, Hassan Farhat, and Will Liu), Molecular Chemistry (Changling Zhao), Thulium (Patrick Muller and Andrei Tretiakov). In Paul Hamilton's group, Sagnac Interferometer (Adam West and Randy Putnam), GNOME (Sami Khamis), HUNTER (Eddie Chang, Sami, Tori Palmaccio, and Arthur Miltner), Ytterbium Bloch Oscillations (Rob Niederriter, Chandel Schlupe-Rombes, Leah Phillips, and Raju Manthana). In Dave Leibbrandt's group, Thorium (Asad Contractor, Rick Matish, Nick Lackmann), Molecules (Chao Fang), Space Clocks (Stephan Randolph). And for discussions about molecular ions outside of UCLA, I am thankful for conversations we've had with Evan Reed, Lu Qi, and Ken Brown (Duke) as well as with Dalton Chaffee and James Chou (NIST). And since one of the greatest challenges in this experiment has been our cryogenic system, I am thankful to Andrew Laugharn (UMD), Hannah Knaack (Boulder), and Riley Ancona (Quantinuum) for their expertise/commiseration with the Stinger system.

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H. Wu, C. Z. C. Ho, G. D. Mitts, J. A. Rabinowitz, and E. R. Hudson, “Nonlinear-Enhanced Wideband Sensing via Subharmonic Excitation of a Quantum Harmonic Oscillator”, *Phys. Rev. Lett.* (2026)

H. Wu, G. D. Mitts, C. Z. C. Ho, J. A. Rabinowitz, and E. R. Hudson, “Wideband electric field quantum sensing via motional Raman transitions”, *Nat. Phys.* (2025)

CHAPTER 1

Introduction

Naïveté can often paint atoms and diatomic molecules as the simplest physical systems; experience warns against such notions. Ion trap systems combine the internal structure of the ion with the quantum harmonic oscillator modes of the ion trap, replicating the simplest systems described by quantization, such as a two-level system and the Fock basis. Interactions that couple the internal states to the motion present increased complexity that allows for the development of useful quantum systems for computing and metrology. The potential applications of these systems motivate continual study, as different atoms and molecules become the targets of such studies.

1.1 Molecular Qubits

The desire to understand and control molecular states does not stem from a singular focus, with different motivations for each experiment. Molecules offer a unique testbed for measurements of fundamental physics such as the electron dipole moment [And18] as well as possible violations of the Standard Model [HC13]. Additionally, an understanding of their internal structure, interactions, and reactions offers insight into the realm of chemistry and biology, which often appear abstract in the study of atoms. Other motivation stem from how the complexity and vast internal structure of molecules can also be leveraged for quantum computing capabilities [Dan25, CH20, HC21]. On the whole, we work with molecules because we think they are cool.

Quantum computing platforms require high-fidelity operations on enormous systems of identical qubits. Atomic systems have emerged as leading candidates since every atom of a given isotope is identical, and the highest fidelity operations have

been demonstrated with atomic ions. The current industrial standard consists of a single qubit described by a pair of ions of different species [RAA25]. Within an ion, different states are chosen to serve as the qubit OMG [D 21b]. While industrial focus is aimed primarily at improving the existing hardware, academic research is aimed at exploring novel avenues to overcome challenges that arise from fundamental limitations. One approach has been to continue investigating alternative atomic species with greater complexity within the internal states, while another has been to investigate alternative ways to introduce interactions between the internal states. With molecular ions, both avenues are under investigation [Cho17,F 24,HC21,MH00]. The greater complexity of molecules leads not only to a prolific amount of internal states but also introduces interactions that were previously forbidden.

1.2 Quantum Sensing

Limitations to qubit fidelity are often from external sources of noise, making the qubit a sensor for this noise. Zeeman shifts that detect the presence of magnetic fields, or small resonant electric fields that excite phonons into an ion chain, demonstrate some of the simplest ways a trapped ion can sense the environment around it. These examples clearly demonstrate how particles can interact with the fields around them but do not necessarily categorize the system as a quantum sensor. A more rigorous classification for a quantum sensor requires utilizing non-classical states. For example, although they may be described in a quantum picture, coherent states can be represented as classical states; utilizing squeezed states, Fock states, CAT states, or entanglement are ways in which a quantum advantage can be introduced [GJN23,BSB19,WSH19,C 13,ZUL18]. To have a quantum advantage, a classical limit must be defined. Referred to as the standard quantum limit, this represents the best achievable sensitivity that a classical state can produce independent of which estimator is chosen. The estimator will serve to convert a detection signal into a physical parameter, for example, how we convert photon counts into a measurement of displacement. Our choice of estimator may result in worse uncertainty than the standard quantum limit, which produces the Cramer-Rao bound. For a choice of estimator, this represents the best possible sensitivity measurement that

can be achieved.

1.3 Overview

The different chapters of this dissertation are categorized as follows.

1.3.1 Chapter 2: Ion Trap Fundamentals

This chapter will focus on ion trap fundamentals, both in the context of the confining potential and in how to visualize the motional modes in the phase space picture. An ion trap thesis is not complete without covering the Mathieu equations and how those lead into the pseudopotential picture of the motional modes of the trap. To prepare a reader for the experimental results captured in this thesis, the phase space picture of different classical and quantum states of a quantum harmonic oscillator are outlined. The Wigner quasi-probability function is derived for each state to facilitate an understanding of its physical implications.

1.3.2 Chapter 3: Atomic States and Interaction with Light

Within this chapter, the basics of atomic physics are covered using Ca^+ , including how its internal states interact with light. A description of the interaction picture for Hamiltonian physics is reviewed since most models we work with have been moved into this picture, and it allows us to easily demonstrate how light can couple a two-level system, i.e., Rabi flopping. Lastly, the different readout methods used in this work will be covered: blue sideband Rabi oscillations, sideband ratio, rapid adiabatic passage, and Wigner reconstruction.

1.3.3 Chapter 4: Diatomic Molecules

A general overview of the different components in the Hamiltonian of a diatomic molecule is provided to familiarize an atomic physicist with the world of molecules. The different quantum numbers associated with each term in the Hamiltonian are outlined, as well as how different Hund's cases represent different couplings between

momentum to determine what are the good quantum numbers in each case. Degenerate states in molecules are introduced in detail, focusing on the A -doublet parity states. Finally, it is brought together for the molecule used in this experiment, H^{35}Cl^+ , depicting the complex energy structure that this experiment was designed to perform spectroscopy on.

1.3.4 Chapter 5: Experimental Setup

This chapter covers all the experimental setup involved in this experiment: cryogenics, vacuum, ion trap, optics, and electronics. Within these pages are discussions about why the experiment is built the way it is, as well as comments on how it could be improved. I hope that grad students building a cryogenic experiment consult this section so as to avoid some of the pitfalls I fell into designing this experiment, notably with respect to joint resistances and black body radiation shielding.

1.3.5 Chapter 6: Electric-field Gradient Gates

The goal of this experiment is outlined within this chapter, as well as the progress made towards it. It has been the most challenging part of writing this work because of the numerous obstacles that we have encountered and how we do not believe we've been successful in implementing this scheme. However, this section covers many of the careful considerations that were made in order to overcome the obstacles, many of which were never considered in this design stage of this experiment. The results and discussion outlined in this chapter will hopefully lead to successful spectroscopy down the road.

1.3.6 Chapter 7: Motional Raman

This chapter represents the turning point in this experiment, and covers the results in our first publication [WMH25]. Birthed from a desire to improve the spectroscopic search of EGGs, the motional Raman technique describes a heterodyne process, whereby two photon can combine to lead to displacements far away from resonance. In addition to describing this process using the Magnus expansion theory, the prop-

erties of this scheme are deeply investigated as well as practical applications of it. Compatibility with quantum amplification allows for demonstration of sensitivities below the standard quantum limit.

1.3.7 Chapter 8: Motional Raman: Subharmonics

A continuation of motional Raman, this chapter further investigates the possibility of multiphoton interactions within a quantum harmonic oscillator. While these processes can be understood in the context of the Magnus expansion, a dive into Floquet theory was chosen to provide an alternative tool toward describing how the different photons combine to produce displacements. What this leads to was a characterization of non-linear interactions through the lens of a quantum harmonic oscillator, which showed practical application in terms of reducing the uncertainty of frequency measurements beyond the linear Fourier transform limit. Further discussion is provided regarding the class of odd subharmonics which were not rigorously investigated, as well as superharmonic terms that indicated signals leaking out of our signal generator.

1.3.8 Chapter 9: Super-resolution

In a similar desire to overcome the Fourier transform limit and extend the practical application of the motional Raman technique, this chapter covers the scheme for resolving two closely separated tones. Referred to as super-resolution, this section outlines the problem of Rayleigh’s curse that stems from quantum noise fluctuations before describing the simple method we devised to suppress these fluctuations. The scheme is captured using Wigner reconstruction, a challenging technique that reproduces the wavepacket in phase space using the characteristic function. Ultimate resolution of this scheme is demonstrated by resolving two signals separated by 5Hz using a 1ms probe time, resulting in a $\times 200$ improvement over the Fourier transform limit.

1.3.9 Chapter 10: CAT Interferometry

In this final chapter, the current work of this experiment is discussed: implementing a quantum enhancement technique for mass spectrometry. Eric has wanted us to perform mass spectrometry in this experiment from the moment a Ca^+ ion was cooled to its ground motional state. Enhancement is performed using spin-motional CAT states to create an interferometer capable of measuring the secular frequencies below the standard quantum limit. For calibration-free measurement, an ion balance is implemented, whereby a comparison of two axial motional modes is made. The second logic ion in this chain also introduces the idea of trinomial statistics, further improving the sensitivity of our measurement. Lastly, we show that entangling both Ca^+ ions prior to the CAT interferometer can produce even more sensitivity, although a demonstration of this with the dark ion will be achieved shortly after the submission of this thesis.

CHAPTER 2

Ion Trap Fundamentals

Confined within an electric potential, a trapped ion has dynamics that result from combining a static field with an oscillating one [Daw76]. Solving the dynamics reveals two different categories of motion, one referred to as the secular motion of the ion and the other as the micromotion. The latter can result in a noise source that heats the ion, and the former allows us to consider the ion as a harmonic oscillator. Specifically, by cooling to the motional ground state, we enter a regime where we consider the behavior of the ion as a quantum harmonic oscillator, introducing a phase space picture characterized by the Wigner quasiprobability distribution [HR06].

2.1 Confining Potential

An ion trap is a tool consisting of electrodes that enable the capture of charged particles in an electric field. In order for charged particles to remain in a stable trapping region, Earnshaw's theorem must be obeyed

$$\nabla \cdot \vec{E} = 0. \quad (2.1)$$

The standard approach to solving begins with an ansatz that the electric field is linear in all three Cartesian directions,

$$\vec{E} = E_0(\lambda x \hat{x} + \sigma y \hat{y} + \gamma z \hat{z}). \quad (2.2)$$

Following Earnshaw's theorem, we see that $\lambda + \sigma + \gamma = 0$. Typically two solutions are often discussed [Daw76], $\lambda = -\sigma, \gamma = 0$ and $\lambda = \sigma, \gamma = -2\sigma$. I will present a slightly more complex solution that takes aspects from these two solutions and rewrites the electric field as

$$\vec{E} = E_0[(\lambda_x + \sigma_x)x\hat{x} + (\lambda_y + \sigma_y)y\hat{y} + \gamma z\hat{z}]. \quad (2.3)$$

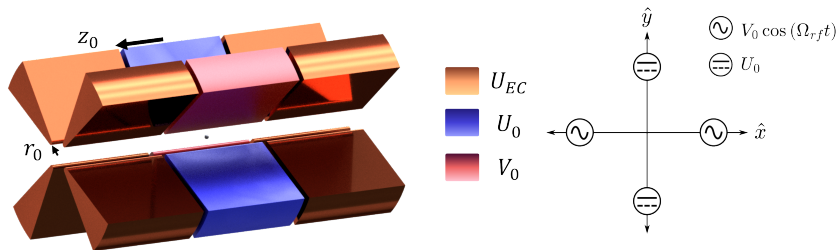


Figure 2.1: The typical linear Paul trap consists of a pair of radial RF electrodes oscillating at Ω_{rf} with an amplitude of V_0 , a pair of radial DC electrodes, and endcap electrodes for axial confinement. The endcap electrodes are shown here with matching voltages for ease. The distance between the ion at the center of the trap and a radial electrode is given by r_0 ; the distance between the ion and the endcaps is given by z_0 .

Earnshaw's theorem with this electric field will be $(\lambda_x + \lambda_y) + (\sigma_x + \sigma_y) + \gamma = 0$, and a nice solution to it will be $\lambda_x = -\lambda_y = \lambda, \sigma_x = \sigma_y = \sigma, \gamma = -2\sigma$. Rewriting Eqn. 2.3 with this substitution results in

$$\vec{E} = E_0[(\lambda + \sigma)x\hat{x} + (-\lambda + \sigma)y\hat{y} - 2\sigma z\hat{z}]. \quad (2.4)$$

When we solve for the potential using this electric field, we will separate the components related to λ and σ . The λ term is the potential of hyperbolic electrodes, a common starting point for many discussions of trapping potentials. As a result, the total electrical potential will be

$$\varphi = -\frac{E_0}{2}[\lambda(x^2 - y^2) + \sigma(x^2 + y^2 - 2z^2)] + const. \quad (2.5)$$

The conventional coordinate system of an ion trap involves defining $\hat{x}$ along the electrode axis that has the trapping voltage, defining $\hat{y}$ for the other radial electrodes, and defining $\hat{z}$ as the axial direction. The electrodes that do not have the trapping voltage applied to them are referred to throughout this thesis as the EGGS electrodes. By separating our potential into components dependent on λ and σ , we can clearly see that the first component is independent of axial position.

The superposition of potentials allows us to separate the RF components and the DC components to determine the unknown values, with the addition of a geometric factor to make up for the shielding from the electrodes. The first is the potential

produced by the RF and EGGs electrodes which will take the form

$$\varphi_{radial} = -\frac{E_0}{2}[\lambda(x^2 - y^2)] + A. \quad (2.6)$$

To solve for the constants $E_0\lambda$ and A , boundary conditions are introduced for each of the radial electrodes, where the RF electrodes produce a potential

$$\varphi_{RF}(x = r_0, y = 0) = -\frac{E_0}{2}\lambda(r_0^2) + A = V_0 \cos(\Omega_{rf}t) \quad (2.7)$$

at $(x_{RF}, y_{RF}) = (r_0, 0)$ and the EGGs electrodes have the potential

$$\varphi_{EGGS}(x = 0, y = r_0) = \frac{E_0}{2}\lambda(r_0^2) + A = U_0 \quad (2.8)$$

at $(x_{EGGS}, y_{EGGS}) = (0, r_0)$. This decision replicates how ion traps are typically operated, with RF applied to one pair of radial electrodes and DC applied to the other [Chr20].

The second potential is produced by the axial confining electrodes, the end caps, which are held at a potential U_{EC} . In the radial case, the end caps have negligible shielding of the RF voltages. However, in the axial case, the RF electrodes do shield the potential produced by the end caps. It's typical to treat the trapping potential as a purely axial component on axis held at κU_{EC} .

$$\varphi_{axial}(x = 0, y = 0, z = z_0) = \frac{E_0}{2}\sigma 2z_0^2 = \kappa U_{EC} \quad (2.9)$$

Solving the previous equations, we get our trapping potential to be

$$\begin{aligned} \varphi &= \varphi_{rad} + \varphi_{axial} \\ &= \frac{V_0 \cos(\Omega_{rf}t) - U_0}{2} \left(1 + \frac{(x^2 - y^2)}{r_0^2}\right) + U_0 + \frac{\kappa U_{EC}}{2z_0^2} (2z^2 - x^2 - y^2). \end{aligned} \quad (2.10)$$

If we assume that there isn't any voltage applied to the EGGs electrodes, then we will get the typical formula for the trapping potential

$$\varphi = \frac{V_0 \cos(\Omega_{rf}t)}{2} \left(1 + \frac{(x^2 - y^2)}{r_0^2}\right) + \frac{\kappa U_{EC}}{2z_0^2} (2z^2 - x^2 - y^2).$$

If instead, a DC voltage is applied to the RF electrodes as opposed to the EGGs electrodes, then the electric potential becomes the following

$$\varphi = \frac{(V_0 \cos(\Omega_{rf}t) + U_0)}{2} \left(1 + \frac{(x^2 - y^2)}{r_0^2}\right) + \frac{\kappa U_{EC}}{2z_0^2} (2z^2 - x^2 - y^2).$$

In order to determine the secular frequencies of ions trapped in this potential, we need to solve for the equations of motion in each element of the coordinate system using Eqn. 2.10. Recall that the Coulomb force will allow us to simply compute the acceleration of a charged particle in an electric field, $\mathbf{F} = e\mathbf{E} = -e\nabla\varphi$. Since our potential has dependence in each direction, we will produce three equations of motion:

$$\ddot{x} = -\left(\frac{eV_0}{mr_0^2}\cos(\Omega_{rf}t) - \frac{eU_0}{mr_0^2} - \frac{\kappa eU_{EC}}{mz_0^2}\right)x \quad (2.11)$$

$$\ddot{y} = \left(\frac{eV_0}{mr_0^2}\cos(\Omega_{rf}t) - \frac{eU_0}{mr_0^2} + \frac{\kappa eU_{EC}}{mz_0^2}\right)y \quad (2.12)$$

$$\ddot{z} = -\frac{2\kappa eU_{EC}}{mz_0^2}z. \quad (2.13)$$

2.1.1 Mathieu Equation

Differential equations of the form

$$\frac{d^2x_i}{d\varepsilon^2} + [a_i + 2q_i\cos(2\varepsilon)]x_i = 0 \quad (2.14)$$

are referred to as the Mathieu equations and have well-studied approximate solutions. In order to write equations 2.11-2.13 in the same form as the Mathieu equations, we will perform a change of variable, $\varepsilon = \frac{\Omega_{rf}t}{2}$. The Mathieu a and q parameters are important values that define how well a charged particle will remain trapped. Outside of the stable regime, particles will not remain confined, a fact which is often utilized for mass selection. In addition, these parameters determine the secular frequencies of the ions, or more fundamentally tell us what physical trap parameters are required for operation. For example, larger traps (large r_0 and z_0) require higher voltages to trap particles, which can sometimes pose challenges and limit stability. For the equations of motion 2.11-2.13, Mathieu a parameters are

$$a_x = -\left(\frac{4eU_0}{mr_0^2\Omega_{rf}^2} + \frac{4e\kappa U_{EC}}{mz_0^2\Omega_{rf}^2}\right) \quad (2.15)$$

$$a_y = \frac{4eU_0}{mr_0^2\Omega_{rf}^2} - \frac{4e\kappa U_{EC}}{mz_0^2\Omega_{rf}^2} \quad (2.16)$$

$$a_z = \frac{8e\kappa U_{EC}}{mz_0^2\Omega_{rf}^2} \quad (2.17)$$

and the q parameters are

$$q_x = \frac{2eV_0}{mr_0^2\Omega_{rf}^2} \quad (2.18)$$

$$q_y = -\frac{2eV_0}{mr_0^2\Omega_{rf}^2} \quad (2.19)$$

$$q_z = 0. \quad (2.20)$$

Approximate solutions to the Mathieu equations are achievable when we make what is known as the adiabatic approximation

$$a_i, q_i \ll 1. \quad (2.21)$$

Under this approximation, the solution to the equation of motion in the i direction is given by

$$x_i \approx A \left(1 - \frac{q_i}{2} \cos(\Omega_{rf}t) \right) \cos(\omega_i t) \quad (2.22)$$

where the trapping frequency Ω_{rf} describes the fast oscillations of the ion, and the slow oscillations are described by

$$\omega_i = \frac{\beta_i \Omega_{rf}}{2}. \quad (2.23)$$

The constant β_i is part of a recursive formula but is most often estimated to the first order term

$$\beta_i \approx \sqrt{a_i + \frac{q_i^2}{2}}. \quad (2.24)$$

Using this approximation, the slow frequency, hereafter referred to as the secular frequency, is

$$\omega_i \approx \frac{\Omega_{rf}}{2} \sqrt{a_i + \frac{q_i^2}{2}}. \quad (2.25)$$

The pseudopotential of the trap will now look like a harmonic oscillator, with the motion described by the secular frequency

$$\varphi = \frac{1}{2}m\omega_x^2 x^2 + \frac{1}{2}m\omega_y^2 y^2 + \frac{1}{2}m\omega_z^2 z^2. \quad (2.26)$$

2.1.2 Multiple Ions

As more particles become confined by the electric potential, they also feel the electric field introduced by the other ions present in the trap. The number of modes increases as more ions are added to the trap, given by $N_{modes} = 3N_{ions}$. In order to determine

these new modes, the potential between each charged particle must be considered. Coupling terms between the different ion positions will be introduced, leading to a new potential energy of the chain. The derivation of the new modes comes from investigating small changes from the equilibrium positions given by the Lagrangian of the system, seen in [Jam98]. A treatment of this as an eigenvalue problem results in the following: the eigenvalues represent the frequencies of each normal mode, and the eigenvectors represent the motional behavior of each mode. An alternative method is to add the ion-ion potential to the pseudopotential given by Eqn. 2.26 and calculate the eigenvalues and eigenvectors for the resulting equations of motion.

The most fundamental mode within the trap is the center-of-mass (COM) mode. For a single particle, these modes represent its motion in all three directions. The next set of modes are introduced when a second ion crystallizes with the first. These modes are commonly referred to as the Rocking or the Breathing modes. Additionally, since the mass of the two particles may not be equal, the COM mode may also change. For the simplest case, both ions will share the same mass, and the resulting mode frequencies are given by ω_x , ω_y , ω_z , $\sqrt{3}\omega_z$, $\sqrt{\omega_y^2 - \omega_x^2}$, and $\sqrt{\omega_x^2 - \omega_z^2}$. The normal mode eigenvectors for the like-ion chain will be $b_{com} = \frac{1}{\sqrt{2}}(1, 1)$ and $b_{rock} = \frac{1}{\sqrt{2}}(1, -1)$ for all three dimensions. The motion of each particle is represented as a different index in b_i . If the second ion had a different mass, not only would the mode frequencies change, the components of the mode eigenvectors will no longer have the same strength, such that the heavier particle would have smaller displacements.

Confinement of two different ions results in a mixed species crystal, with secular frequencies dependent on the mass of both ions. Estimation of the secular frequencies for multiple ions requires numerically solving the Lagrangian, but analytical solutions exist for simple cases, such as the two-ion mixed species crystal [JS12] or the three-ion mixed crystal with a single mismatched ion in the center of the chain [DWZ00]. Maintaining a linear chain is important for our applications, and when two different ions are confined within the same potential well, they can form a linear chain provided the radial confinement is greater than the axial confinement by

$$\left(\frac{\omega_z}{\omega_r}\right)_{crit}^2 = 2.53N^{-1.73} \quad (2.27)$$

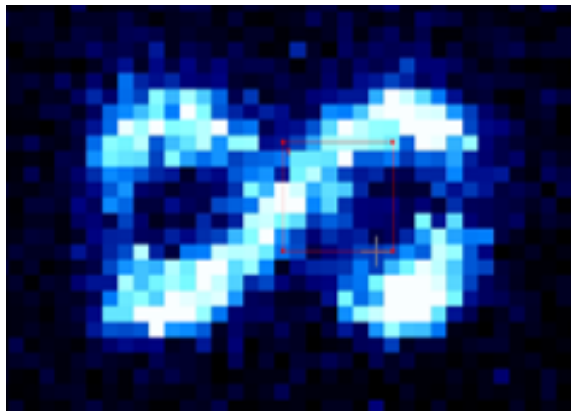


Figure 2.2: An ion chain can undergo a phase transition [Sch93] whereby the radial and axial modes undergo strong coupling. This chain is made up of two Ca^+ ions with an axial secular frequency of 690kHz, an RF secular frequency of 1200MHz, and an EGGs secular frequency of 1331MHz. The radial frequencies were split by applying $U_0 = 1\text{V}$ to the EGGs electrodes.

where $\omega_r = \sqrt{\omega_x^2 + \omega_y^2}$ [Ste97]. Given this condition, the two particles will have ion separation distances of

$$x_{eq} = \left(\frac{q^2}{4\pi\epsilon_0} \right)^{\frac{1}{3}} \left(\frac{m_1\omega_1^2 + m_2\omega_2^2}{(m_1\omega_1^2)(m_2\omega_2^2)} \right)^{\frac{1}{3}} \quad (2.28)$$

and for two ions of the same mass this simplifies to

$$x_{eq} = \left(\frac{q^2}{2\pi\epsilon_0 m\omega_z^2} \right)^{\frac{1}{3}}. \quad (2.29)$$

A more rigorous, numerical treatment provided in [Jam98] reveals that the scaling varies as

$$x_{eq}(N) = \left(\frac{q^2}{4\pi\epsilon_0 m\omega^2} \right)^{\frac{1}{3}} \frac{2.018}{N^{0.559}} \quad (2.30)$$

for N like-mass ions confined in a linear chain. For quantum computing applications with linear chains [GD09], this relation is important but Eqn. 2.29 works well enough for our applications.

2.1.3 Excess Micromotion

So far the discussion has been limited to the motion described by the pseudopotential Eqn. 2.26. However, the motion of the ion described in Eqn. 2.22 indicates that the fast frequency oscillations can only be ignored for small Mathieu- q . Typical

confinement is provided by $q \approx 0.1$, which means that these oscillations are still noticeable. The motion, seen in Fig.2.3, is referred to as intrinsic micromotion. When there are static fields that push the ion off of the trapping null, additional motion is produced, referred to as excess micromotion. Changes to the static potential are typically introduced by patches of dielectric material on the electrodes that get charged by the UV laser light and discharged by collisions with background gasses (most noticeably during a warmup cycle). Shimming procedures that compensate for the stray field and push the ion back to the micromotion null are an important step to limit unnecessary heating from excess micromotion [BMB98], [NDM21], [SH11]. The displacement from the null for a stray electric field U_i^{EM} in the i^{th} dimension will be given by

$$\delta x_i = \frac{eU_i^{EM}}{m\omega_i^2} \quad (2.31)$$

which modifies Eqn.2.22

$$x_i \approx \left(1 - \frac{q_i}{2} \cos(\Omega_{rf}t)\right) (\delta x_i + A \cos(\omega_i t)). \quad (2.32)$$

An in-depth analysis of the effects of excess micromotion is performed in [BMB98], showing that this new equilibrium position will result in an increase in the kinetic energy for a period of oscillation by

$$\delta E_i = \frac{1}{2}m\left(\frac{q_i\delta x_i^2\Omega_{rf}^2}{2\sqrt{2}}\right)^2. \quad (2.33)$$

For large displacements from the trap null, the increase in heating can severely limit experiments, as the motional state will quickly decohere.

2.2 Quantum Harmonic Oscillator

The confining potential generated by the ion trap creates a unique playground for physicists because our particle's motion now represents its own quantum system, the quantum harmonic oscillator. The quantum harmonic oscillator (QHO) plays an important role in the study of quantum physics because it is one of the most fundamental examples of quantum physics, and these systems can be found throughout different branches of physics, such as in superconducting QC, Nitrogen Vacancy Centers, and trapped ion experiments. What makes these systems so exciting is that,

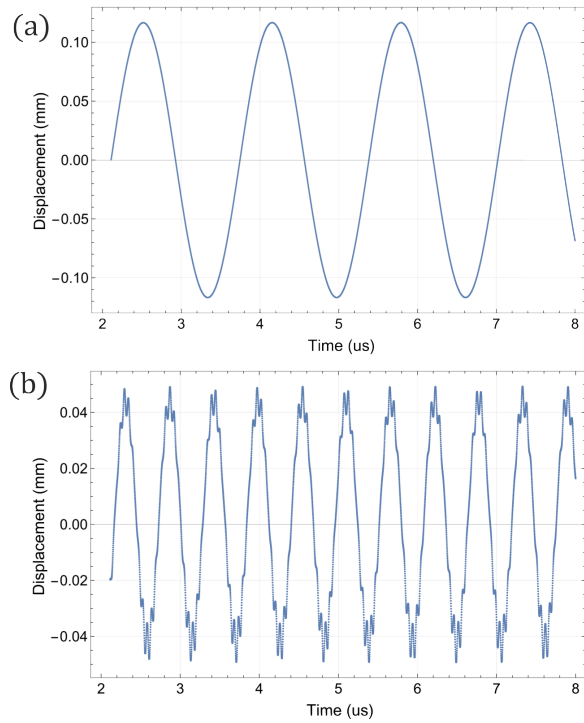


Figure 2.3: Simulated results for the motion of the ion in our trap. Simulations were generated using SIMION software. Axial motion (a) of the ion is generated by an electrostatic potential, whereas radial motion (b) comes from an oscillating potential, resulting in intrinsic micromotion. Parameters for the simulations were $V_0 = 150\text{V}$, $U_0 = 0\text{V}$, $U_{EC} = 200\text{V}$, and $\Omega = 2\pi \cdot 20\text{MHz}$ for a single Ca^+ in our trap ($r_0 = 550\mu\text{m}$ and $z_0 = 2.2\text{mm}$). From these simulations, and an estimate of $\kappa = 0.15$ was made. While the real values were measured to be slightly different, these estimates served to confirm ion stability with our new trap design.

regardless of what physics is present to create the Hamiltonian, the behavior that results is all the same.

To begin, the Hamiltonian for the QHO is given by

$$\hat{H}_{QHO} = \frac{1}{2}m\omega^2\hat{x}^2 + \frac{\hat{p}^2}{2m} \quad (2.34)$$

which will look nearly identical to the classical Hamiltonian of a harmonic oscillator, with the subtle difference stemming from the fact that position and momentum are now operators

$$\hat{x} = \sqrt{\frac{\hbar}{2m\omega}} (\hat{a}^\dagger + \hat{a}) \quad (2.35)$$

and

$$\hat{p} = i\sqrt{\frac{\hbar m\omega}{2}} (\hat{a}^\dagger - \hat{a}). \quad (2.36)$$

The raising and lowering operators $\hat{a}^\dagger$ and $\hat{a}$ are the fundamental operators associated with the QHO, resulting in a single phonon (photon in optical systems) being added or removed from the oscillator [Gri05]. Phonons are the unit of energy associated with a QHO; the quanta forms its own operator given by $\hat{n} = \hat{a}^\dagger\hat{a}$. With this definition the Hamiltonian can be rewritten as

$$\hat{H} = \hbar\omega \left(\hat{n} + \frac{1}{2} \right), \quad (2.37)$$

where $\hat{n}$ is the number of phonons in the system with energy $\hbar\omega$ and the term $\hbar\omega/2$ is the ground state energy of the system, which exists even when there are no phonons present in the system. Throughout this work the ground state energy term will frequently be dropped since it does not contribute to the system dynamics.

2.2.1 Wigner Phase Space

Phase space is introduced as a way of visualizing the kinematics of a harmonic oscillator over the course of its period. Classically this is depicted as position on one axis and momentum on the other. The oscillator could start at any point in phase space, but for pedagogical reasons it's chosen to be on the positive position axis. In the most common way of visualizing this, a pendulum starts held still at some height (assuming small angle). Upon its release, it will start moving towards the equilibrium position, reaching its peak speed at this point, before starting to slow

down again. In phase space, the oscillator forms a circle and the initial energy is transferred back and forth between position and momentum.

By painting a similar picture for a QHO, the time evolution behavior and probability distributions can be visualized [WM08]. Consider that a system is described by a density matrix $\hat{\rho} = \sum_i p_i |\psi_i\rangle \langle \psi_i|$, then the expectation value of an operator $\hat{A}$ is given by

$$\langle \hat{A} \rangle = \langle \psi | \hat{\rho} \hat{A} | \psi \rangle = \sum_i p_i \langle \psi_i | \hat{A} | \psi_i \rangle \quad (2.38)$$

with the uncertainty given by

$$\sigma(\hat{A}) = \sqrt{\langle \hat{A}^2 \rangle - \langle \hat{A} \rangle^2}. \quad (2.39)$$

For a QHO in the ground state $|n=0\rangle$, the expected position and momentum are $\langle \hat{x} \rangle = \langle 0 | \hat{x} | 0 \rangle = 0$ and $\langle \hat{p} \rangle = \langle 0 | \hat{p} | 0 \rangle = 0$, respectively. This should seem reasonable; ignoring the ground state energy, the system does not have any energy associated with motion. For sensing and metrological experiments, high precision is sought after, so it's important to picture the uncertainty of the different axes. Continuing from the ground state example, $\sigma(\hat{x}) = \sqrt{\frac{\hbar}{2m\omega}} = x_0$ and $\sigma(\hat{p}) = \sqrt{\frac{\hbar m\omega}{2}} = p_0$. The product of these two uncertainties notably is the Heisenberg uncertainty limit $\sigma_x \sigma_p \geq \hbar/2$.

Wigner space is the typical way that phase space will be visualized for QHO systems. A Wigner function can be calculated from its characteristic function using a variation of Eqn. 2.38,

$$\chi(\beta) = \langle \hat{D}(\beta) \rangle = \int dx' \langle x' | \hat{\rho} \hat{D}(\beta) | x' \rangle \quad (2.40)$$

which represents the average displacement of the state described by the density operator $\hat{\rho}$ [HR06]. Transforming from the characteristic function to the Wigner function takes the form

$$W(\gamma) = \frac{1}{\pi^2} \int d^2\beta e^{\beta\gamma^* - \beta^*\gamma} \chi(\beta) \quad (2.41)$$

where $\gamma = \gamma_r + i\gamma_i$ represents the coordinates of the Wigner space. The Wigner function is a quasi-probability distribution function and is frequently defined as

$$W(x, p) = \frac{1}{\pi} \int_{-\infty}^{\infty} du \langle x + \frac{u}{2} | \hat{\rho} | x - \frac{u}{2} \rangle e^{-2ipu}, \quad (2.42)$$

with $u = 2(x' - x)$. While the Wigner function can be organized in the x and p frame, it is typically most convenient to transform into a different frame given by the real and imaginary parts of γ , γ_r and γ_i . The most common transform is to define $\gamma = \frac{1}{2}(\frac{x}{\sigma_x} + i\frac{p}{\sigma_p})$, for real position and momentum values.

The Wigner function is referred to as a quasi-probability distribution since it can produce negative values, which are forbidden in a true probability distribution. Negative values are indicative of a truly quantum state, and must exist when the probability density of finding an oscillator at position x ($P(x) = \langle x | \hat{\rho} | x \rangle$) or with momentum p ($P(p) = \langle p | \hat{\rho} | p \rangle$) has nodes. These marginal distributions are related to the Wigner function by

$$P(x) = \int dp W(x, p) \quad (2.43)$$

$$P(p) = \int dx W(x, p) \quad (2.44)$$

and at a position (or momentum) that has $P(x_0) = 0$, this forces a negative value in the Wigner function [HR06].

2.2.2 Fock States

Since the Hilbert space of the QHO is given by the Fock (number) states, it makes the most sense to start by describing these states first. These states are orthogonal to each other ($\langle i | j \rangle = \delta_{ij}$) so it is a convenient basis to work from. The wavefunction of these states is given by [Gri05]

$$\psi_n(x) = \langle x | n \rangle = \frac{1}{\sqrt{2^n n!}} \left(\frac{1}{2\pi\sigma_x^2} \right)^{\frac{1}{4}} e^{-\frac{x^2}{4\sigma_x^2}} H_n\left(\frac{x}{\sqrt{2}\sigma_x}\right), \quad (2.45)$$

where $H_n(x)$ are the Hermite polynomials. By replacing x_0 with σ_x we can see a clear physical meaning behind this wavefunction. For the simplest case, if the QHO is in its motional ground state, $n=0$, then the probability of measuring the position $P_x = |\langle x | n=0 \rangle|^2$ is the normalized Gaussian distribution centered at 0, with an uncertainty of σ_x . Since all other states can be made up of a linear combination of these states, so can the wavefunctions of those states.

Fock states have a density operator represented by $\hat{\rho} = |n\rangle \langle n|$, producing an

average displacement of

$$\chi_n(\beta) = \langle n | \hat{D}(\beta) | n \rangle = e^{-\frac{|\beta|^2}{2}} \mathcal{L}_n(|\beta|^2) \quad (2.46)$$

after expanding the displacement operator and removing all vanishing elements. $\mathcal{L}_n(|\beta|^2)$ is the n^{th} Laguerre polynomial. Transforming produces a Wigner distribution of the form

$$W(x, p, n) = \frac{2}{\pi} (-1)^n \mathcal{L}_n \left(\left(\frac{x}{\sigma_x} \right)^2 + \left(\frac{p}{\sigma_p} \right)^2 \right) e^{-\frac{1}{2} \left(\left(\frac{x}{\sigma_x} \right)^2 + \left(\frac{p}{\sigma_p} \right)^2 \right)}. \quad (2.47)$$

While in the classical phase space picture, position and momentum are convenient axes, as mentioned earlier switching to a different frame, given by $\gamma = \frac{1}{2} \left(\frac{x}{\sigma_x} + i \frac{p}{\sigma_p} \right)$ is better for visualizing the different states in Wigner space, allowing the Fock state Wigner distribution to be written as

$$W(\gamma, n) = \frac{2}{\pi} (-1)^n \mathcal{L}_n(4|\gamma|^2) e^{-2|\gamma|^2}. \quad (2.48)$$

2.2.3 Coherent States

The orthogonality of the Fock states makes them a convenient basis to work in; however, one of the most utilized types of states is the coherent state. These states are eigenstates of the lowering operator ($\hat{a} |\alpha\rangle = \alpha |\alpha\rangle$) and are generated from the displacement operator,

$$\hat{D}(\alpha) = e^{\alpha \hat{a}^\dagger - \alpha^* \hat{a}}. \quad (2.49)$$

A coherent state is defined by its linear combination of Fock states,

$$|\alpha\rangle = e^{-\frac{|\alpha|^2}{2}} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle. \quad (2.50)$$

When a displacement operator is applied to the ground state it generates the coherent state $\hat{D}(\beta) |0\rangle = |\beta\rangle$, but more generally when it is applied onto another coherent state $|\alpha\rangle$, it will further displace it in phase space

$$\hat{D}(\beta) |\alpha\rangle = \hat{D}(\beta) \hat{D}(\alpha) |0\rangle = e^{\frac{\beta \alpha^* - \beta^* \alpha}{2}} |\alpha + \beta\rangle. \quad (2.51)$$

Coherent states can be described entirely classically, and are still a type of minimal uncertainty state. In phase space a coherent state can easily be visualized as

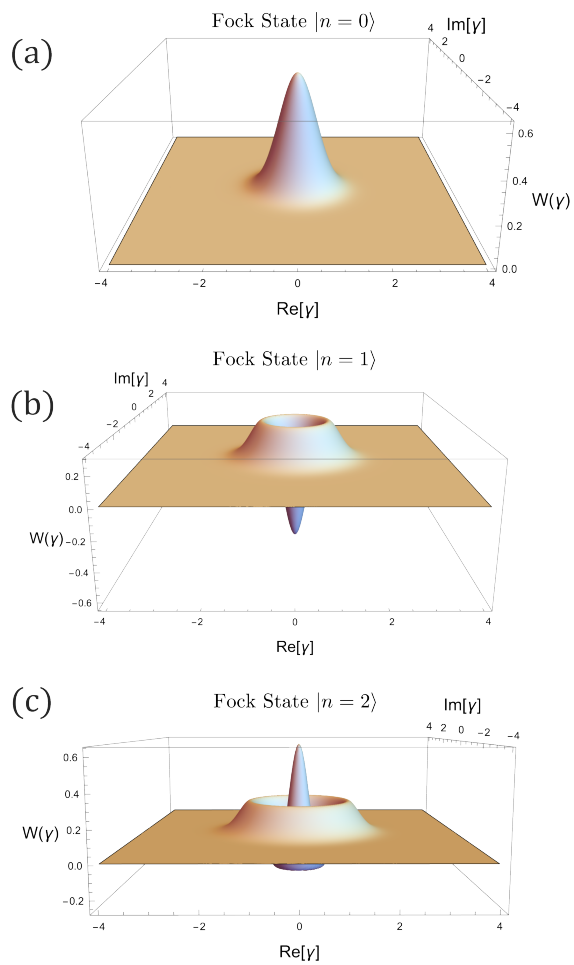


Figure 2.4: Wigner distribution for the (a) $|n = 0\rangle$, (b) $|n = 1\rangle$, and (c) $|n = 2\rangle$ Fock states. Negative Wigner values in the $|n \neq 0\rangle$ cases indicate non-classical behavior.

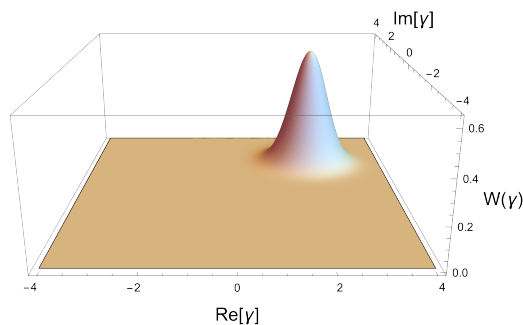


Figure 2.5: Wigner distribution for a coherent state. Using a displacement given by $\alpha_r = 2$ and $\alpha_i = 2$ for $\alpha = \alpha_r + i\alpha_i$.

displacing the Gaussian probability of the ground state to a different point in space without changing the uncertainties. Conveniently, it has the same general behavior when described by the Wigner picture. To prove this picture, it should be intuitive that for the coherent state $|\alpha\rangle$, the mean displacement will be

$$\chi_{coh}(\beta) = \langle \alpha | \hat{D}(\beta) | \alpha \rangle = e^{\frac{\beta \alpha^* - \beta^* \alpha}{2}} \langle \alpha | \alpha + \beta \rangle = e^{-\frac{|\beta|^2}{2}} e^{\beta \alpha^* - \beta^* \alpha} \quad (2.52)$$

where we used the definition Eqn. 2.50 to solve for the inner product $\langle \alpha | \alpha + \beta \rangle$. The Wigner function for a coherent state is then described by

$$W(\gamma, \alpha) = \frac{2}{\pi} e^{-2|\gamma - \alpha|^2} \quad (2.53)$$

whereas with Fock states $\gamma = \frac{1}{2} \left(\frac{x}{\sigma_x} + i \frac{p}{\sigma_p} \right) = \gamma_r + i\gamma_i$, and the displacement $\alpha = \alpha_r + i\alpha_i$ has real component α_r and imaginary component α_i . Substituting these values reproduces another common form of the Wigner function

$$W(\gamma, \alpha) = \frac{2}{\pi} e^{-2((\gamma_r - \alpha_r)^2 + (\gamma_i - \alpha_i)^2)}. \quad (2.54)$$

2.2.4 Thermal States

Thermal states describe an oscillator that has been interacting with its environment and as such has become a mixed state. An ion that is Doppler cooled but not sideband cooled can correctly be described by a thermal distribution, as can an ion that is being heated by a chaotic field within the trap after being prepared in its motional ground state [WM08]. The Maxwell-Boltzmann distribution for a quantum

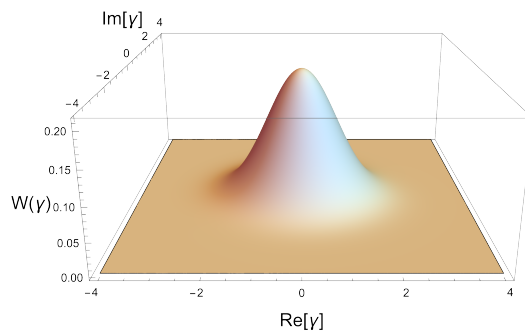


Figure 2.6: Wigner distribution for a thermal state of $\bar{n} = 2$.

harmonic oscillator will take the form

$$\hat{\rho}_{th} = (1 - e^{-\frac{\hbar\omega_0}{k_B T}}) \sum_{n=0}^{\infty} e^{-\frac{n\hbar\omega_0}{k_B T}} |n\rangle \langle n| \quad (2.55)$$

and the Bose-Einstein distribution for phonons is

$$\bar{n} = \frac{1}{e^{\frac{\hbar\omega_0}{k_B T}} - 1}. \quad (2.56)$$

Substituting Eqn. 2.56 into 2.55, the probability distribution of a thermal state is

$$P_n = \langle n | \rho | n \rangle = \frac{1}{(1 + \bar{n})} \left(\frac{\bar{n}}{1 + \bar{n}} \right)^n. \quad (2.57)$$

Similarly, when the average displacement is calculated using the density operator, the resulting characteristic function is

$$\chi_{th}(\beta) = \langle n | \hat{\rho}_{th} \hat{D}(\beta) | n \rangle = \sum_{n=0}^{\infty} \frac{\bar{n}^n}{(\bar{n} + 1)^{n+1}} \chi_n(\beta) \quad (2.58)$$

where we've substituted the characteristic function for the n^{th} Fock state. Simplifying this result takes several lines of algebra and the generating function of the Laguerre polynomial to produce

$$\chi_{th}(\beta) = e^{-(\bar{n} + \frac{1}{2})|\beta|^2}. \quad (2.59)$$

By transforming the characteristic function, it can be shown that the Wigner function for the thermal state takes the form

$$W(\gamma, \bar{n}) = \frac{2}{\pi} \frac{1}{1 + 2\bar{n}} e^{-\frac{2|\gamma|^2}{1 + 2\bar{n}}} \quad (2.60)$$

where once again γ is the coordinate in phase space and $\bar{n}$ is the average phonon number of the thermal state.

2.2.5 Squeezed States

A state cooled to its ground state i.e. an $n=0$ Fock state, $\alpha = 0$ coherent state, or $\bar{n} = 0$ thermal state, will be a circle in phase space with minimal uncertainty in both quadratures, $\sigma(\gamma_r)$ and $\sigma(\gamma_i)$. Reducing the uncertainty in one quadrature at the expense of the other indicates squeezing of the wavefunction [Bur20]. Squeezing is given by the operator

$$\hat{S}(\xi) = e^{\frac{1}{2}(\xi^* \hat{a}^2 - \xi \hat{a}^{\dagger 2})} \quad (2.61)$$

where the squeezing parameter is $\xi = r e^{i\vartheta}$, r is the strength of the squeeze and ϑ is the angle that is getting squeezed ($\vartheta = 0$ squeezes in $Re(\gamma)$ and $\vartheta = \pi$ squeezes in $Im(\gamma)$). Applying the squeezing operator on vacuum state $|0\rangle$, will produce the squeezed vacuum state, and applying it to a coherent state will produce a squeezed coherent state. The effect in phase space will remain the same in either case. A squeezed vacuum state

$$|\xi\rangle = \frac{1}{\sqrt{\cosh r}} \sum_{n=0}^{\infty} \frac{(-e^{i\vartheta} \tanh r)^n \sqrt{(2n)!}}{2^n n!} |2n\rangle \quad (2.62)$$

will have non-vanishing Fock state populations for only even Fock numbers

$$P_n = |\langle \xi | n \rangle|^2 = \begin{cases} 0, & \text{if } n=\text{odd} \\ \frac{(\tanh r)^{2n}}{\cosh r} \frac{(2n)!}{(2^n n!)^2}, & \text{if } n=\text{even} \end{cases} \quad (2.63)$$

although this will not hold for a squeezed coherent state. To calculate the characteristic function, recognize that the squeezing and displacement operators do not commute, but instead obey $\hat{D}(\beta)\hat{S}(\xi) = \hat{S}(\xi)\hat{D}(\zeta)$ where $\zeta = \beta \cosh r + \beta^* e^{i\vartheta} \sinh r$, such that

$$\chi_{sq}(\beta) = \langle \xi | \hat{D}(\beta) | \xi \rangle = \langle 0 | \hat{S}(\xi)^\dagger \hat{D}(\beta) \hat{S}(\xi) | 0 \rangle = \chi_{n=0}(\zeta). \quad (2.64)$$

The squeezed vacuum state's Wigner function is written as either

$$W(\gamma, \xi) = \frac{2}{\pi} e^{(-2e^{2r} Re(e^{-i\frac{\vartheta}{2}} \gamma)^2 - 2e^{-2r} Im(e^{-i\frac{\vartheta}{2}} \gamma)^2)} \quad (2.65)$$

or as

$$W(x, p, \xi) = \frac{2}{\pi} e^{-2e^{2\xi} (\frac{x}{2\sigma_x})^2} e^{-2e^{-2\xi} (\frac{p}{2\sigma_p})^2}. \quad (2.66)$$

From the form of the Wigner function for the squeezed vacuum state, it is clear that the squeezing produces a two-dimensional Gaussian from the initial Gaussian

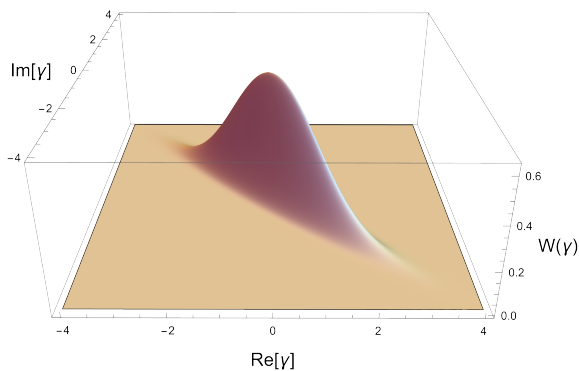


Figure 2.7: Wigner distribution for a squeezed vacuum state. Squeezing is generated with $\xi = e^{\frac{\pi}{2}}$.

distribution. An effective uncertainty in position and momentum goes as $e^{-\xi}\sigma_x$ and $e^{\xi}\sigma_p$, such that as we reduce the uncertainty in one dimension, we end up enhancing it in the other direction by the same amount. When the squeezed operator is applied onto a coherent state, a Gaussian with some displacement in phase space, it will make that distribution into a two-dimensional Gaussian as well.

2.2.6 CAT States

The final distribution relevant to the work of this thesis is the Schrödinger CAT state given by a linear superposition of two oppositely signed coherent states

$$|cat_+\rangle = \frac{|\alpha\rangle + |-\alpha\rangle}{\sqrt{2(1 + e^{-2|\alpha|^2})}} \quad (2.67)$$

$$|cat_-\rangle = \frac{|\alpha\rangle - |-\alpha\rangle}{\sqrt{2(1 - e^{-2|\alpha|^2})}} \quad (2.68)$$

for an even and an odd CAT state, respectively. Using the density operator $\hat{\rho}_{cat}^{\pm} = |cat_{\pm}\rangle \langle cat_{\pm}|$ to calculate the characteristic function,

$$\begin{aligned} \chi_{cat}^{\pm}(\beta) &= \langle cat_{\pm} | \hat{D}(\beta) | cat_{\pm} \rangle \\ &= \frac{\langle \alpha | \hat{D}(\beta) | \alpha \rangle + \langle -\alpha | \hat{D}(\beta) | -\alpha \rangle \pm \langle \alpha | \hat{D}(\beta) | -\alpha \rangle \pm \langle -\alpha | \hat{D}(\beta) | \alpha \rangle}{2(1 \pm e^{-2|\alpha|^2})}, \end{aligned} \quad (2.69)$$

where it is apparent that the characteristic functions for both $|\alpha\rangle$ and $|-\alpha\rangle$ are present, as well as a pair of interference terms. Likewise, the Wigner distribution will reflect this as well

$$W(\gamma, \alpha) = \frac{2}{\pi(1 \pm e^{-2|\alpha|^2})} \left(e^{-2|\gamma-\alpha|^2} + e^{-2|\gamma+\alpha|^2} \pm 2e^{-2|\gamma|^2} \cos(4(\gamma_i\alpha_r - \gamma_r\alpha_i)) \right) \quad (2.70)$$

where the first two terms are coherent states related to the two components of the cat, and the last term represents the interference fringes between them. The calculation done here was for a CAT state generated with equal displacements about the origin. Similar calculations can be done for two arbitrary coherent states.

2.2.7 Spin-Motional CAT States

When working with a system that consisted of both a spin and a motional component, we must consider a different type of CAT, which is referred to as a spin-motional CAT. This type is what we produce in our experiment with bichromatic tones on our quadrupole laser [Hem14]. These CAT states are given by

$$|cat_+^{SM}\rangle = \frac{|+, \alpha\rangle + |-, -\alpha\rangle}{\sqrt{2}} \quad (2.71)$$

$$|cat_-^{SM}\rangle = \frac{|+, \alpha\rangle - |-, -\alpha\rangle}{\sqrt{2}} \quad (2.72)$$

where the spin state is represented by $|\pm\rangle$ for the σ_x eigenvectors, and the motional state is represented by $|\pm\alpha\rangle$. The density matrix is expanded as

$$\hat{\rho}_{SM}^{\pm} = |+, +\alpha\rangle \langle +, +\alpha| + |-, -\alpha\rangle \langle -, -\alpha| \pm |-, -\alpha\rangle \langle +, +\alpha| \pm |+, +\alpha\rangle \langle -, -\alpha|. \quad (2.73)$$

To calculate the characteristic function we need to consider both the spin and the motional spaces, designated by x and y , respectively,

$$\chi_{SM}^{\pm}(\beta) = \iint dx dy \langle x, y | \hat{\rho}_{SM}^{\pm} \hat{D}(\beta) | x, y \rangle = \frac{1}{2} (\langle \alpha | \hat{D}(\beta) | \alpha \rangle + \langle -\alpha | \hat{D}(\beta) | -\alpha \rangle). \quad (2.74)$$

We see that the interference fringes are actually removed by considering the spin component as well. Not surprisingly, when we transform to the Wigner distribution, we see that it no longer possesses the interference behavior

$$W(\gamma, \alpha) = \frac{1}{\pi} \left(e^{-2|\gamma-\alpha|^2} + e^{-2|\gamma+\alpha|^2} \right) \quad (2.75)$$

and just looks like a mixture of two coherent states.

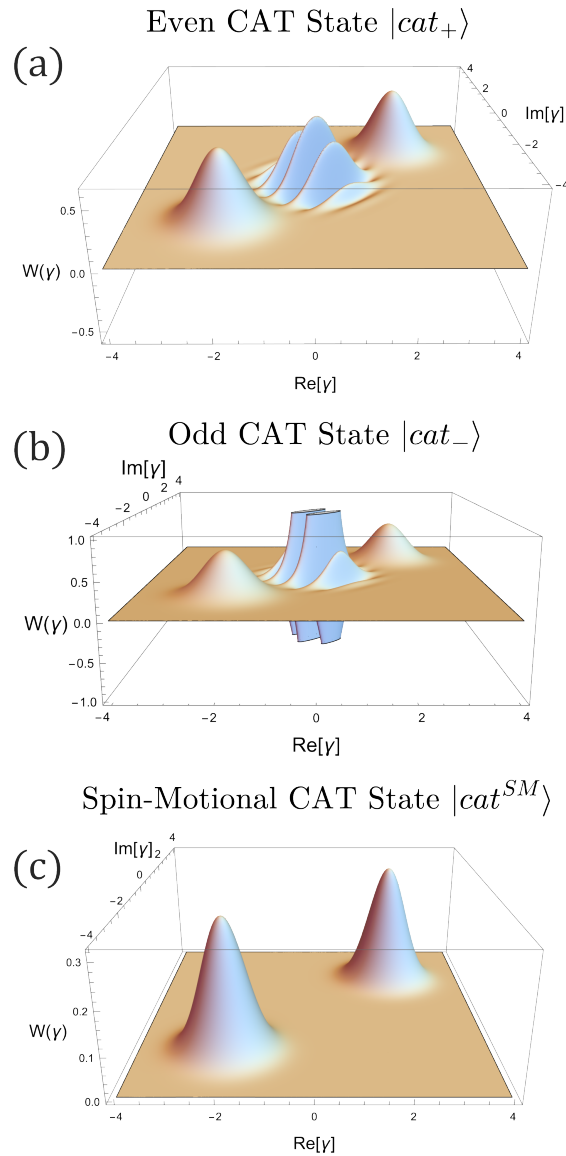


Figure 2.8: Wigner Distribution functions for (a) even CAT, (b) odd CAT, and (c) Spin-Motional CAT state. Each state is generated using a displacement strength of $\alpha_r = 2$ and $\alpha_i = 2$ for $\alpha = \alpha_r + i\alpha_i$. Note the presence of interference fringes in the even and odd cases, that are not present in the Spin-Motional CAT case. The normalization term in the odd case, leads to an increased amplitude near the origin, notably at the fringes.

CHAPTER 3

Atomic States and Interactions with Light

Atomic ion experiments implement lasers to cool the modes of the QHO and introduce a coherent coupling between the internal state of the atom and its motion. This coupling is what allows atoms to serve as precision sensors and as a platform for quantum computing. At the moment, however, there are still only a handful of atomic ions with closed cycling transitions for laser cooling, typically limited to the second column of the periodic table, the alkaline earth elements [htt]. The other standard laser-coolable ion is ytterbium. Through co-trapping with a laser-addressable ion, other atomic species, in addition to molecules, can be sympathetically cooled to their motional ground states as well. To facilitate these experiments, it is convenient to pair a molecule with a laser-coolable atom with comparable mass. In large part for this reason, calcium was chosen as the atom for our experiment to be based around.

3.1 Calcium States

$^{40}\text{Ca}^+$ is the workhorse ion. Historically, it has been studied for quantum computing applications, resulting in well-documented transitions and experimental procedures from loading, to state readout, to two-qubit operations [Roo00]. Additionally, since many experiments are developed around this species, the lasers required to drive its transitions are convenient for off-the-shelf purchasing. The lack of a nuclear spin leaves it susceptible to magnetic fields, but also limits the number of possible states that need to be addressed. Its branching ratio from the excited state is much higher for the $S \Leftrightarrow P$ decay than the $P \Leftrightarrow D$, leading to a higher photon collection rate than other species. Reactions with H_2 molecules are also quickly dissociated with the cooling laser, leading to lifetimes on the order of a week in our chamber [RCJ16].

$\lambda(\text{nm})$	$\tau(\text{ns})$	Γ (2 π MHz)
396.847	7.7(2)	20.7
729.147	$1.165(11) \times 10^9$	1.52×10^{-7}
866.214	94.3	1.69
854.209	101	1.58
393.366	7.4(3)	21.5
732.389	$1.196(11) \times 10^9$	1.47×10^{-7}
849.802	901	0.18

Table 3.1: $^{40}\text{Ca}^+$ natural lifetimes and spontaneous emission rates. Values are taken from [WM], [BS78], [AS05], [RA88].

And for our experiment, the medium-light mass of 40amu has an innately higher secular frequency and also pairs well with our molecule of choice, HCl^+ , due to their similar masses.

3.1.1 Field-Free Energies

As an aside, a brief discussion of the atomic Hamiltonians can help remind us of some of the mechanics at play, as well as help to motivate the later discussion on molecular ions. Starting in the field-free picture, atoms have their largest energy splittings given by the orbits of the valence electrons. For simplicity, most atoms are chosen such that there is a single valence electron, hence the prevalence of laser-cooled singly ionized alkaline earth ions. By solving the time-independent Schrödinger equation with a potential given by the Coulomb interaction between the electron and the nucleus, the atomic quantum numbers n , l , and m_l are introduced for the electronic orbital number, orbital angular momentum, and the projection of the orbital momentum onto the quantization axis. The next most notable effect comes from the spin-orbital coupling of the electron’s spin with its orbital momentum. Switching from the frame of a stationary nucleus, whereby the electron is moving, into the frame of the electron, will introduce an orbiting nucleus and a magnetic field produced by its motion. The

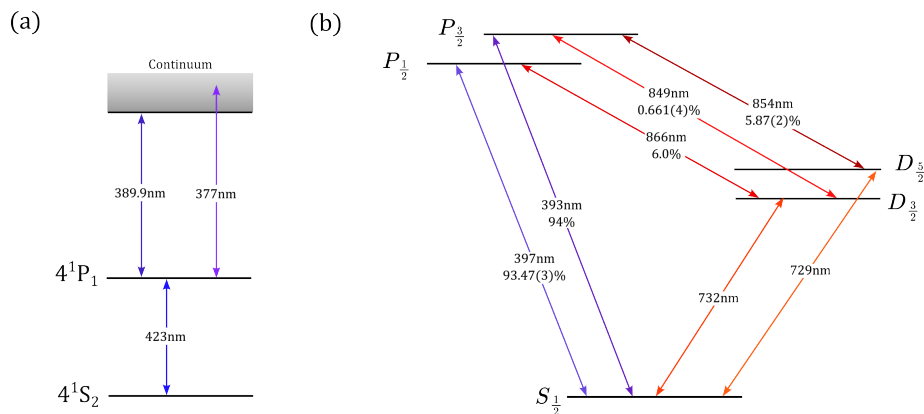


Figure 3.1: Calcium undergoes a two-step photoionization process, using a resonant transition from the ground 4^1S_2 to the excited 4^1P_1 state via 423nm light, followed by light at 377nm that excites to the continuum (a) [DS04]. Energy levels of Ca^+ (b). The branching ratios for P-state decays are shown in percentages underneath each wavelength [R 08]. The D-states will only decay to the S-state via an electronic quadrupole transition.

Hamiltonian for the spin-orbital coupling is

$$H_{so} = \frac{Ze^2\hbar^2}{2m_e\mu c^2} \frac{1}{r^3} \mathbf{L} \cdot \mathbf{S} \quad (3.1)$$

where $Z|e|$ is the nuclear charge, e is the electron's charge, μ is the reduced mass of the electron/nucleus system, m_e is the electron's mass, and r is the distance the electron is from the nucleus [Wei78]. The electronic spin is given by $\mathbf{S}$ and the orbital angular momentum is defined by $\mathbf{L}$. Since the values L_z and S_z do not commute with Eqn.3.1, their projections m_l and m_s are not good quantum numbers. Instead, the total angular momentum $\mathbf{J} = \mathbf{L} + \mathbf{S}$ is a good quantum number, such that the atomic state is given by

$$|\psi\rangle = |n, l, s, J, m_j\rangle = \sum_{m_l, m_s} \langle n, l, s, m_l, m_s | n, l, s, J, m_j \rangle |n, l, s, m_l, m_s\rangle \quad (3.2)$$

where the value $\langle n, l, s, m_l, m_s | n, l, s, J, m_j \rangle$ is the Clebsch-Gordan coefficient, which can be calculated through vector addition with the ladder operators $J_{\pm}$. While there is a closed form solution, it is typically quickest to find a table for the specific case at hand. Together with two other corrections (the relativistic effects and the Darwin term), the energy from the spin-orbital coupling produces the fine-structure energy levels of the atom in the absence of an external field (field-free terms). Although

it need not be considered for the isotope of calcium used in this experiment, the nuclear spin of the atomic nucleus will introduce additional splittings referred to as the hyperfine structure. Coming from the nuclear magnetic moment interaction with the electron's magnetic moment, the resulting Hamiltonian is given by

$$H_{hf} = A(\mathbf{J} \cdot \mathbf{I}) \quad (3.3)$$

where A is the first hyperfine constant and $\mathbf{I}$ is the nuclear angular momentum. When considering the hyperfine structure, m_J and m_I are not good quantum numbers, so the total angular momentum including nuclear spin $\mathbf{F} = \mathbf{J} + \mathbf{I}$ is introduced with projection m_F . An additional term that plays a larger role for heavier atoms is the contribution from a non-spherical nucleus

$$H_{nsn} = B(\mathbf{J} \cdot \mathbf{I}(\mathbf{J} \cdot \mathbf{I} + 1)),$$

where the primary interaction comes from the quadrupole component of the nucleus and B is the second hyperfine constant.

3.1.2 Zeeman Splitting

The atomic energy structure so far has been in a field-free picture that is unrealistic for most experiments. When exposed to a homogeneous static magnetic field, the energy levels of the atom will split. There are two regimes typically considered: a weak field introduces the Zeeman effect and a strong field results in the Paschen-Back effect. The strength of the field corresponds to how the energy changes relative to the energy from the spin-orbital coupling. A “weak” field generates a small splitting in energy for each m_J state compared to the $P_{\frac{3}{2}}$ and $P_{\frac{1}{2}}$ splitting, whereas a “strong” field prevents the different P states from being well-defined. For an atom with no nuclear spin like $^{40}\text{Ca}^+$, within the “weak” regime, the good quantum numbers are given by the state $|l, s, J, m_J\rangle$, and within the “strong” regime the state is no longer well defined by the quantum numbers l, s, J , or m_J but instead by m_s and m_l . For atoms with nuclear spin, the “weak” field state is given by $|F, m_F\rangle$ and the “strong” field state is given by $|m_J, m_I\rangle$.

In our experiment, the magnetic field is only several Gauss, which puts us in the

weak field regime. The magnetic moment of the atom is given by

$$\boldsymbol{\mu}_{atom} = -\frac{e\hbar}{2\mu}(\mathbf{L} + g_e\mathbf{S}) = -\boldsymbol{\mu}_L - \boldsymbol{\mu}_S \quad (3.4)$$

where $g_e = 2.0023$ is the Landé g-factor for the electron spin, resulting in a Hamiltonian given by

$$H_Z = -\boldsymbol{\mu}_{atom} \cdot \mathbf{B} = -\frac{e\hbar}{2\mu}(\mathbf{L} + g_e\mathbf{S}) \cdot \mathbf{B} = -\boldsymbol{\mu}_L \cdot \mathbf{B} - \boldsymbol{\mu}_S \cdot \mathbf{B} \quad (3.5)$$

with $\boldsymbol{\mu}_L$ and $\boldsymbol{\mu}_S$ represent the magnetic moments associated with the electron orbital and spin momenta, respectively. The energy for a given state will be $\langle n, l, s, J, m_j | H_Z | n, l, s, J, m_j \rangle$, and we will define the quantization axis by the magnetic field $\mathbf{B} = B_0$. To solve for $\langle n, l, s, J, m_j | (L_z + g_e S_z) | n, l, s, J, m_j \rangle$, we will utilize the Landé formula

$$\frac{\langle \alpha, j, m_j | \mathbf{T}_q^{(1)} | \alpha, j, m'_j \rangle}{\langle \alpha, j, m_j | \mathbf{U}_q^{(1)} | \alpha, j, m'_j \rangle} = \frac{\langle \alpha, j, m_j | \mathbf{T}^{(1)} \cdot \mathbf{U}^{(1)} | \alpha, j, m_j \rangle}{\langle \alpha, j, m_j | \mathbf{U}^{(1)} \cdot \mathbf{U}^{(1)} | \alpha, j, m_j \rangle} \quad (3.6)$$

which is a special case of the Wigner-Eckart formula, where $\mathbf{T}^{(1)}$ and $\mathbf{U}^{(1)}$ are irreducible rank one tensors. Using $\mathbf{T}^{(1)} = \mathbf{L} + g_e\mathbf{S}$ and $\mathbf{U}^{(1)} = \mathbf{J}$, Eqn. 3.6 can be rewritten as

$$\langle \alpha, j, m_j | L_z + g_e S_z | \alpha, j, m'_j \rangle = \frac{\langle \alpha, j, m_j | (\mathbf{L} + g_e\mathbf{S}) \cdot \mathbf{J} | \alpha, j, m_j \rangle}{\langle \alpha, j, m_j | \mathbf{J} \cdot \mathbf{J} | \alpha, j, m_j \rangle} \langle \alpha, j, m_j | J_z | \alpha, j, m'_j \rangle, \quad (3.7)$$

and with the following substitution [Wei78]

$$(\mathbf{L} + g_e\mathbf{S}) \cdot \mathbf{J} = (\mathbf{L} + \mathbf{S}) \cdot \mathbf{J} + (g_e - 1)\mathbf{S} \cdot \mathbf{J} = \mathbf{J}^2 + \frac{(g_e - 1)}{2}(\mathbf{J}^2 + \mathbf{S}^2 - \mathbf{L}^2) \quad (3.8)$$

we reveal that the presence of a weak magnetic field splits the energy states by the projection m_j

$$E_Z \approx \frac{e\hbar}{2m_e} g_J B_0 m_j. \quad (3.9)$$

The Landé g factor for the total angular momentum $\mathbf{J}$ is

$$g_J = 1 + (g_e - 1) \frac{j(j+1) + s(s+1) - l(l+1)}{2j(j+1)}. \quad (3.10)$$

3.2 Atom-Laser Interactions

In addition to static fields, the atom will experience oscillating fields, the most common form coming from an interaction with a laser. These interactions come in

several forms; this thesis will discuss dipole and quadrupole interactions. Dipole transitions typically possess strong couplings between the atomic states, resulting in short lifetimes given by Fermi's Golden Rule: spontaneous decay from an excited state is proportional to the difference in energy between the excited and ground state cubed. Short lifetimes prevent coherent operations between such states, but facilitate Doppler cooling using these transitions. A quadrupole laser allows for coupling between two dipole-forbidden states, which results in narrow linewidth transitions and longer state lifetimes, which can be used for coherent operations, most notably operations that couple the internal state of the atom to its motion within the trap.

3.2.1 Interaction Picture

Before describing the atom-laser interaction, it is important to outline the interaction picture of the Hamiltonian, where time-independent terms are removed [Ste07]. For a Hamiltonian that is made up of time independent and time dependent components

$$H = H_0 + H_1(t), \quad (3.11)$$

the time evolution operator $U = e^{-i\frac{H_0}{\hbar}t}$ is used to transform the time dependent component into the interaction picture

$$H_I = U^\dagger H_1 U. \quad (3.12)$$

For a trapped ion with addressable internal states, there will be two time-independent terms, one for the internal states of the ion (assumed here to be an atom but not limited to) and one for the phonon modes of the quantum harmonic oscillator

$$H_{atom} = \frac{\hbar\Delta}{2}\sigma_z \quad (3.13)$$

$$H_{QHO} = \hbar\omega(\hat{a}^\dagger\hat{a} + \frac{1}{2}) \quad (3.14)$$

where the separation in energy between the two spin states is given by Δ and the energy of a single phonon is ω_0 . It is clear that moving to the interaction frame of the atom will not affect the quantum harmonic oscillator, and vice versa. The atom-laser interaction is modeled by the Jaynes-Cummings Hamiltonian, so consider the time-dependent Hamiltonian to be the spin-motion coupled term from the quadrupole

laser Eqn. 3.34

$$H_1 = \hbar\eta\Omega(\sigma_+ + \sigma_-)(\hat{a}^\dagger + \hat{a})\sin(\omega_l t - \varphi_l). \quad (3.15)$$

The first step will involve transforming into the interaction picture of the atomic energy level. In this frame the operators $\sigma_\pm$ transform to $\sigma_\pm e^{\pm i\Delta t}$,

$$\frac{H_{I,atom}}{\hbar} = \omega\hat{a}^\dagger\hat{a} + \eta\Omega(\sigma_+e^{i\Delta t} + \sigma_-e^{-i\Delta t})(\hat{a}^\dagger + \hat{a})\sin(\omega_l t - \varphi_l). \quad (3.16)$$

The second interaction picture involves moving into the ion frame. In this case the operator $\hat{a}$ and $\hat{a}^\dagger$ transform into $\hat{a}e^{-i\omega_0 t}$ and $\hat{a}^\dagger e^{i\omega_0 t}$, respectively, to produce

$$\frac{H_I}{\hbar} = \eta\Omega(\sigma_+e^{i\Delta t} + \sigma_-e^{-i\Delta t})(\hat{a}^\dagger e^{i\omega_0 t} + \hat{a}e^{-i\omega_0 t})\sin(\omega_l t - \varphi_l). \quad (3.17)$$

Now that the Hamiltonian has been rewritten in the interaction picture, we can apply the necessary rotating wave approximations. The rotating wave approximation (RWA) removes terms in the Hamiltonian that are oscillating at high frequencies and only keeps the terms which are slowly varying.

3.2.2 Dipole Interaction

Most laser-based transitions are classified as dipole interactions, because the electric field of the laser is able to couple between two states via their transition dipole moment [Ste07]. The electron that takes part in the interaction will be a distance r_e from the atomic nucleus, representing the dipole as $\mathbf{d} = -er_e$. Calculation of the transition dipole moment is non-trivial, so it is typically left as $\langle g|\mathbf{d} \cdot \hat{\varepsilon}|e\rangle$ where $|g\rangle$ and $|e\rangle$ are the ground and excited states under consideration and $\hat{\varepsilon}$ is the unit vector of the applied electric field. The Hamiltonian governing this interaction is

$$H_{AF} = -\mathbf{d} \cdot \mathbf{E} \quad (3.18)$$

where we use the subscript AF to indicate this term is the atom-field component of the Hamiltonian. If we start by considering the atom as a two-level system as in Eqn. 3.13, and that the laser generates an electric field $\mathbf{E}(t) = E_0 \cos(\omega_l t)\hat{x}$, then the dipole is expressed as $\mathbf{d}\sigma_x$. By switching into the interaction picture of the atom, the atom-field Hamiltonian becomes

$$H_I = -\langle g|\hat{x} \cdot \mathbf{d}|e\rangle \frac{E_0}{2}(\sigma_-e^{i(\omega_l - \Delta)t} + \sigma_+e^{-i(\omega_l - \Delta)t}) = \frac{\hbar\Omega}{2}(\sigma_-e^{i\delta t} + \sigma_+e^{-i\delta t}) \quad (3.19)$$

which has a Rabi frequency $\Omega = \frac{\langle g|\hat{\mathbf{x}}\cdot\mathbf{d}|e\rangle E_0}{\hbar}$ and detuning $\delta = \omega_l - \Delta$. When on resonance, $\delta = 0$, and this becomes $H_I = -\frac{\hbar\Omega}{2}\sigma_x$, illustrating perfect Rabi flopping between $|g\rangle$ and $|e\rangle$ using the time evolution operator

$$U(t) = e^{-i\frac{\Omega t}{2}\sigma_x} = \cos\left(\frac{\Omega t}{2}\right)\mathbb{1} + i\sin\left(\frac{\Omega t}{2}\right)\sigma_x \quad (3.20)$$

or, choosing to represent this time evolution as a matrix

$$U(t) = \begin{pmatrix} \cos\left(\frac{\Omega t}{2}\right) & i\sin\left(\frac{\Omega t}{2}\right) \\ i\sin\left(\frac{\Omega t}{2}\right) & \cos\left(\frac{\Omega t}{2}\right) \end{pmatrix}. \quad (3.21)$$

3.2.3 Doppler Cooling

Transitions between $S \Leftrightarrow P$ and $P \Leftrightarrow D$ are dipole-allowed which follow the previous discussion. Driving $S_{\frac{1}{2}} \Leftrightarrow P_{\frac{1}{2}}$ allows for cooling of the Ca^+ ion through Doppler cooling. Assuming the atoms are moving into the laser of frequency ω_l with velocity v , then the detuning from the atomic transition is given by $\delta = \Delta - \omega_l + kv$ since there is a slight blue shift of the light in the atom's reference frame. A single-photon will produce a force on the atom from the radiative pressure [Foo05] given by the rate of momentum change for the single photon event

$$F_{rad} = p_\gamma R_{scatt}. \quad (3.22)$$

The scattering rate is determined by the lifetime of the excited state and steady-state population in that state $\rho_{ee} = \frac{1}{2} \frac{s}{1+s}$, where s is the cross section, also referred to as the saturation parameter. For a homogeneous field there will be a Lorentzian frequency dependence and the saturation parameter is

$$s = \frac{\Omega^2}{2(\delta^2 + (\frac{\Gamma}{2})^2)} \quad (3.23)$$

or also commonly denoted as

$$s = \frac{\frac{I}{I_{sat}}}{(1 + 4(\frac{\delta}{\Gamma})^2)} \quad (3.24)$$

using the laser intensity I and the saturation intensity

$$I_{sat} = \frac{c\epsilon_0\Gamma^2\hbar^2}{4|\langle g|\hat{\mathbf{e}}\cdot\mathbf{d}|e\rangle|^2}. \quad (3.25)$$

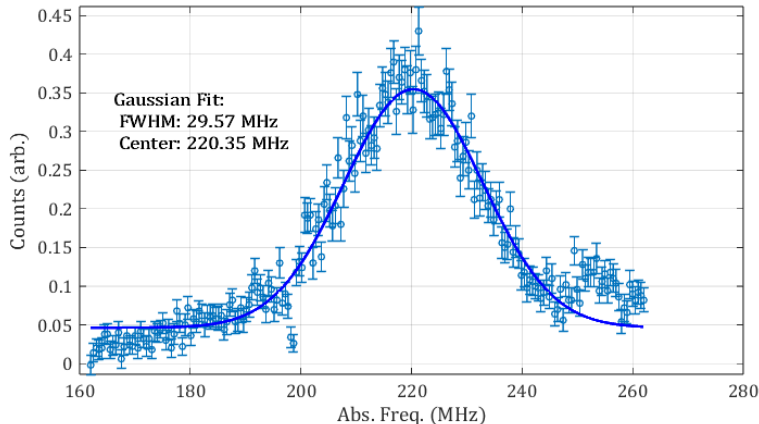


Figure 3.2: Example measurement of the 397nm linewidth. The horizontal axis is the frequency that the 397nm AOM is being driven at. To compensate for frequency dependence in the AOM performance, a calibration curve has been taken. The fit line is shown with a Gaussian to account for Zeeman broadening, although the natural lineshape of a single transition is a Lorentzian.

The force felt by the atom can thus be framed as

$$F_{rad} = \frac{\hbar k \Gamma}{2} \frac{s}{1+s} = \frac{\hbar k \Gamma}{2} \frac{\frac{\Omega^2}{2}}{\frac{\Omega^2}{2} + \delta^2 + (\frac{\Gamma}{2})^2} \quad (3.26)$$

resulting in an ion temperature of

$$T = \frac{\hbar \Gamma}{4k_B} \frac{1 + (\frac{2\delta}{\Gamma})^2}{\frac{2\delta}{\Gamma}}. \quad (3.27)$$

Cooling is optimized when the laser is detuned to $\frac{\Gamma}{2}$, such that the minimum Doppler temperature is $k_B T_D = \frac{\hbar \Gamma}{2}$. For Ca^+ , the Doppler temperature is about 0.5mK. To estimate the average phonon number, we assume that the temperature for a given mode can be described by $\frac{1}{2}k_B T = \hbar \omega_0 (n + \frac{1}{2})$, such that we expect to have $\bar{n} \approx 15$ for a single ion after being Doppler cooled.

When the laser intensity is greater than the saturation intensity, the linewidth of the transition will undergo power broadening given by $\Delta\omega_{FWHM} = \Gamma(1 + \frac{I}{I_{sat}})^{\frac{1}{2}}$. During laser-induced fluorescence of the Ca^+ , the laser is slightly red-detuned in order to ensure the ion does not undergo additional heating. The linewidth measured in Fig. 3.2 is broader than the natural linewidth of the transition and is fit with a Gaussian. From Eqn. 3.26 it should be clear that the linewidth should be a

Lorentzian. Since the magnetic field applied caused a Zeeman shift ($2\pi \cdot 1.4\text{MHz/G}$) between the two ground states given by Eqn. 3.9, there will be two Lorentzians with peak separations of $\sim 10\text{MHz}$. Due to the central limit theorem, two closely separated Lorentzian peaks can be approximated as a Gaussian, which motivated the choice of fit in Fig. 3.2. This behavior is referred to as Zeeman broadening, noting that it's not a true broadening effect.

3.2.4 AC Stark Shift

When the atom is off-resonantly coupled to an additional dipole transition it will experience a shift in its energies, referred to as either the AC Stark shift or the light shift. The shift comes from off-resonant coupling between two states $|g\rangle$ and $|e\rangle$, whose splitting is given by Δ , by a laser oscillating at ω_l with electric field amplitude of E ,

$$H_{eff} = -\frac{\hbar|\Omega|^2}{4\delta}(|g\rangle\langle g| - |e\rangle\langle e|). \quad (3.28)$$

A convenient calculation is possible using the effective Hamiltonian theory [JJ07]. The detuning from the atomic transition is given by $\delta = \omega_l - \Delta$ and the resonant Rabi rate between the two atomic states is given by $\frac{\Omega}{2} = \frac{\langle g|\hat{x}\cdot\mathbf{d}|e\rangle E}{\hbar}$. An alternative way of labeling the shift is as

$$\Delta E = \frac{3\pi c^2}{2\Delta^2} \frac{\Gamma}{\delta} I \quad (3.29)$$

where the laser's intensity is given by I and the scattering rate of the transition is

$$\Gamma = \frac{\Delta^3}{3\pi\epsilon_0\hbar c^3} |\langle g|\hat{x}\cdot\mathbf{d}|e\rangle|^2 \quad (3.30)$$

for a dipole driven transition (typically interpreted as Fermi's Golden Rule). This way can often be preferable experimentally, since the intensity of the laser can be measured with relative ease, and the scattering rate can be looked up for most common species (scattering rate is $2\pi \times$ the natural linewidth).

3.2.5 Quadrupole Interaction

Electric dipole (E1) interactions are single photon interactions between two different states. The cooling light ($S \Leftrightarrow P$) as well as the repumping light ($P \Leftrightarrow D$) are allowed by an electric dipole interaction, however transitions $S \Leftrightarrow D$ are not. These

transitions, however, are accessible via an electric quadrupole (E2) interaction, which requires a narrow linewidth laser.

For Ca^+ , we are interested in driving transitions between $S_{\frac{1}{2}} \Leftrightarrow D_{\frac{5}{2}}$ since accessing the metastable state enables motional control of the ion. Unlike the $D_{\frac{3}{2}}$ state, the $D_{\frac{5}{2}}$ state is not part of the Doppler cooling scheme for Ca^+ , and the $D_{\frac{5}{2}}$ state's additional energy levels make it preferable for shelving schemes needed for generating $n \neq 0$ Fock states. Interactions with this laser enable coupling of the spin of the atom, characterized by the electronic state, with the motion of the ion within the trap. Consider a traveling wave that is near-resonant with this transition given by some interaction strength Ω , frequency ω_l , and phase φ_l

$$\frac{H_{\text{laser}}}{\hbar} = \frac{\Omega\sigma_x}{2}(e^{i(k\hat{x}-\omega_l t+\varphi_l)} + e^{-i(k\hat{x}-\omega_l t+\varphi_l)}). \quad (3.31)$$

Ion location within this field is given by $\hat{x} = x_0(\hat{a}^\dagger + \hat{a})$ where $x_0 = \sqrt{\frac{\hbar}{2m\omega_0}}$. After separating out the position-dependent term, we can perform the Lamb-Dicke approximation ($\eta \ll 1$)

$$\begin{aligned} \frac{H}{\hbar} &= \frac{\Omega\sigma_x}{2}(e^{ikx_0(\hat{a}^\dagger + \hat{a})}e^{-i(\omega_l t - \varphi_l)} + e^{-ikx_0(\hat{a}^\dagger + \hat{a})}e^{i(\omega_l t - \varphi_l)}) \\ &= \frac{\Omega\sigma_x}{2}((1 + ikx_0(\hat{a}^\dagger + \hat{a}))e^{-i(\omega_l t - \varphi_l)} + (1 - ikx_0(\hat{a}^\dagger + \hat{a}))e^{i(\omega_l t - \varphi_l)}) \end{aligned} \quad (3.32)$$

where the Lamb-Dicke parameter is

$$\eta = kx_0 = k\sqrt{\frac{\hbar}{2m\omega_0}} \quad (3.33)$$

for a given mode ω_0 . We take only the first two terms in the Lamb Dicke approximation, and rewrite the term $k\hat{x}$ as $\eta(\hat{a} + \hat{a}^\dagger)$ by turning the position term into the position operator. Rewriting Eqn. 3.32 gives

$$\frac{H}{\hbar} = \Omega\sigma_x((\cos(\omega_l t - \varphi_l) + \eta(\hat{a} + \hat{a}^\dagger)\sin(\omega_l t - \varphi_l)) \quad (3.34)$$

which illustrates that one component of this interaction is not coupled to motion (the carrier) and one component is (the sideband).

3.2.6 Carrier Flopping

Before examining the spin-motional coupling from the laser, we'll consider the case of driving carrier transitions between $S_{\frac{1}{2}} \Leftrightarrow D_{\frac{5}{2}}$. Optical pumping allows us to prepare the $|S_{\frac{1}{2}}, m_j = -\frac{1}{2}\rangle$ state. The orientation of the magnetic field, laser k-vector,

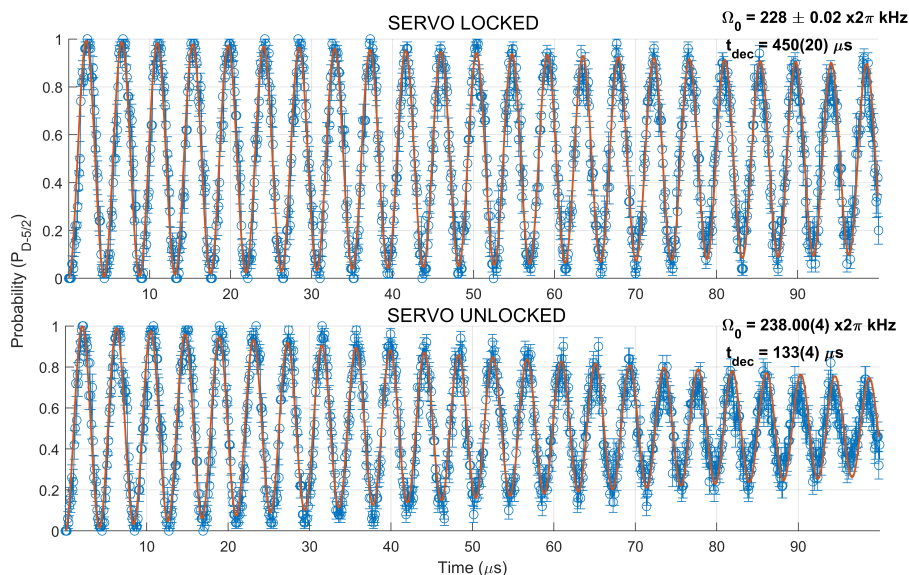


Figure 3.3: Two carrier Rabi flopping results are shown, the top with active laser stabilization and the bottom without. A single calcium ion is sideband cooled on each of its modes prior to the 729nm Rabi pulse. The servo locked test has a longer coherence time but a weaker Rabi rate, since the locking point must be held below the peak power.

and laser polarization determine the coupling strength of the Zeeman transitions for different Δm_j values. By orienting the laser perpendicular to the magnetic field, and its polarization orthogonal to both the laser k-vector and the magnetic field, transitions with $\Delta m_j = \pm 2$ are optimized, and $\Delta m_j = 0, \pm 1$ transitions are suppressed [Roo00]. After optical pumping, $|S_{\frac{1}{2}}, m_j = -\frac{1}{2}\rangle \Leftrightarrow |D_{\frac{5}{2}}, m_j = -\frac{5}{2}\rangle$ and $|S_{\frac{1}{2}}, m_j = -\frac{1}{2}\rangle \Leftrightarrow |D_{\frac{5}{2}}, m_j = -\frac{3}{2}\rangle$ are allowed, with the former being five times more easily excited by resonant light. As such, a two-level atomic system can be reasonably approximated by the states $|S_{\frac{1}{2}}, m_j = -\frac{1}{2}\rangle = |\downarrow\rangle$ and $|D_{\frac{5}{2}}, m_j = -\frac{5}{2}\rangle = |\uparrow\rangle$.

Near-resonant light described by the spin only component of Eqn. 3.34 results in an interaction Hamiltonian given by

$$\frac{H_I}{\hbar} = \Omega(\sigma_+ e^{i\Delta t} + \sigma_- e^{-i\Delta t}) \cos(\omega_l t - \varphi_l). \quad (3.35)$$

To implement the rotating wave approximation, consider the near resonant behavior of the laser at $\omega_l = \Delta + \delta$, where δ is some small detuning relative to the splitting in

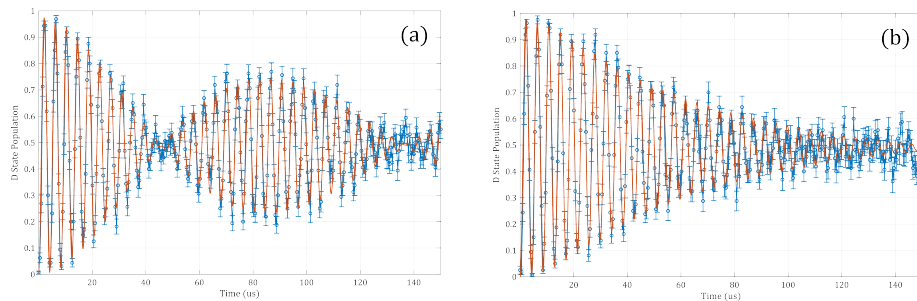


Figure 3.4: Two calcium ions are ground state cooled prior to driving carrier Rabi oscillations. Due to slight differences in the laser power on each ion, there will be a beat note that is seen in the ion fluorescence. In (a), this beat note is more pronounced since the laser is more misaligned. Alignment prior to (b), allows for a less obvious power difference between the two ions' Rabi rates. From the fits, shown in orange, the alignment in (a) has a 4.6% increase in Rabi rate on one ion, whereas in (b) it is only 1.3%. This data was taken with the servo unlocked.

energy between $|\downarrow\rangle$ and $|\uparrow\rangle$. We now have

$$\begin{aligned} \frac{H_I}{\hbar} &= \frac{\Omega}{2}(\sigma_+(e^{i(2\Delta+\delta)t} + e^{-i\delta t}) + \sigma_-(e^{-i(2\Delta+\delta)t} + e^{i\delta t})) \\ &\approx \frac{\Omega}{2}(\sigma_+e^{-i\delta t} + \sigma_-e^{i\delta t}) \end{aligned} \quad (3.36)$$

after introducing the rotating wave approximation. When the laser is on-resonance with the atomic transition, $\delta = 0$,

$$\frac{H_I}{\hbar} = \frac{\Omega}{2}\sigma_x. \quad (3.37)$$

The time evolution of this Hamiltonian will transfer population between the $|\downarrow\rangle$ and the $|\uparrow\rangle$ states given by Eqn. 3.20, which is measured by applying the laser light for different periods of time. As shown in Fig. 3.3, by cooling to the motional ground state of each mode, a single Ca^+ is able to undergo many periods of oscillation before it starts to decohere. Locking the intensity of the laser prior to it entering the chamber gives us a noticeable improvement in our coherence. When a second Ca^+ is present in the trap, the fluorescence can indicate a difference in laser intensity on the two ions via a beat note on the Rabi oscillation shown in Fig. 3.4.

3.2.7 Sideband Cooling

Spin-motional coupling introduced by the position operator in Eqn. 3.31 resulted in Eqn. 3.17 when operating in the Lamb-Dicke regime and dropping the carrier term. When the orientation of the laser and the modal contributions are considered, the time dependent Hamiltonian will be

$$\begin{aligned} \frac{H_I}{\hbar} = & \Omega(\sigma_+ + \sigma_-) \cos(\omega_l t - \psi_l) \\ & + \Omega \eta_m b_m^{(a)} \cos(\vartheta) (\sigma_+ + \sigma_-) (a_m^\dagger + a_m) \sin(\omega_l t - \psi_l) \end{aligned} \quad (3.38)$$

resulting in a interaction-picture Hamiltonian given by

$$\begin{aligned} \frac{H_I}{\hbar} = & \frac{\Omega}{2} (\sigma_+ e^{i\Delta t} + \sigma_- e^{-i\Delta t}) (e^{i(\omega_l t - \psi_l)} + e^{-i(\omega_l t - \psi_l)}) \\ & + \frac{i\Omega}{2} \eta_m b_m^{(a)} \cos(\vartheta) (\sigma_+ e^{i\Delta t} + \sigma_- e^{-i\Delta t}) (a_m^\dagger e^{i\omega_m t} + a_m e^{-i\omega_m t}) (e^{-i(\omega_l t - \psi_l)} - e^{i(\omega_l t - \psi_l)}) \end{aligned} \quad (3.39)$$

where the subscript m denotes the operation on mode m , $b_m^{(a)}$ is the normalized mode eigenvector component for atom a , and ϑ is the angle between the laser k-vector and the atom's motion $\hat{x}_m$. The mode component $b_m^{(a)}$ depends on the number of ions and their masses. For a single Ca^+ , the value will be 1. For two Ca^+ , the values have magnitude $\frac{1}{\sqrt{2}}$ with the COM mode having the same sign from being in-phase and the ROCKING mode having opposite signs from being out-of-phase. For one Ca^+ and one H^{35}Cl^+ , the b values are dependent on the trapping parameters. For simplicity, we absorb the orientation dependence into the Lamb-Dicke parameter η , assume a single atom is being trapped, and ignore the laser phase. Additionally, the laser is near resonance with a motional sideband of a single mode ($\omega_l = \Delta \pm \omega_0 + \delta$), such that under the rotating wave approximation only one mode is under consideration, which results in

$$\frac{H_{BSB}}{\hbar} \approx \frac{i\Omega}{2} \eta (\sigma_+ \hat{a}^\dagger e^{-i\delta t} - \sigma_- \hat{a} e^{i\delta t}). \quad (3.40)$$

and

$$\frac{H_{RSB}}{\hbar} \approx \frac{i\Omega}{2} \eta (\sigma_+ \hat{a} e^{-i\delta t} - \sigma_- \hat{a}^\dagger e^{i\delta t}). \quad (3.41)$$

when $\omega_l = \Delta \pm \omega_0 + \delta$, for the blue sideband and red sideband Hamiltonians respectively [CW95, FW89, RZR99]. The rotating wave approximation resulted in four

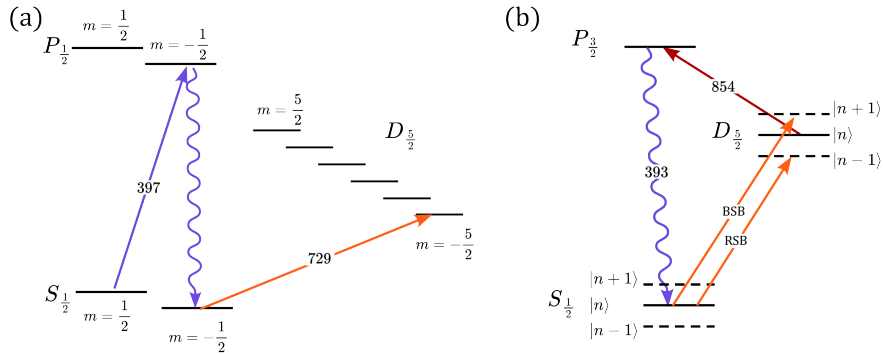


Figure 3.5: (a) Using spin-polarized 397nm light, the population from $m_j = +\frac{1}{2}$ can be optically pumped into the $m_j = -\frac{1}{2}$ state. Addressing this state with our 729nm laser dominantly transfers population into the $m_j = -\frac{5}{2}$ state. (b) Driving the motional sidebands of the $S_{\frac{1}{2}} \leftrightarrow D_{\frac{5}{2}}$ will add or subtract phonons from the given mode. A quench laser at 854nm is then used to remove population from the $D_{\frac{5}{2}}$ state back into the $P_{\frac{3}{2}}$ state, which rapidly undergoes spontaneous decay. After preparing the $|S_{\frac{1}{2}}, m = \frac{1}{2}\rangle$ state through optical pumping with spin-polarized 397nm light, coherent operations are generated using 729nm light to the $|D_{\frac{5}{2}}, m = \frac{5}{2}\rangle$ state. Sideband operations are carried out within this manifold by coupling to the red sideband (RSB) and blue sidebands (BSB) of the motional modes of the excited state. In the vacuum state only the BSB can be driven, and the RSB population remains in the ground state.

terms associated with the carrier vanishing, the slowest of which were $\frac{\Omega}{2}(\sigma_+ e^{-i(\omega_0 + \delta)t} + \sigma_- e^{i(\omega_0 + \delta)t})$, which are included in this discussion as a reminder that there will be some residual effect on sideband motion from the carrier, though it is significantly weaker.

3.2.8 Off-Resonance Effect

Off-resonance Rabi flopping between two states is characterized by

$$P = \frac{\Omega^2}{\Omega^2 + \delta^2} \sin^2\left(\frac{\sqrt{\Omega^2 + \delta^2}}{2}t\right) \quad (3.42)$$

where δ is the detuning and Ω is the Rabi rate of a given interaction [Ste07]. For a carrier transition, $\Omega = \Omega_0$ and for a sideband transition, $\Omega = \Omega_0\eta$. As an example, the typical Rabi rate of our carrier transition is $\Omega_0 = 2\pi \cdot 200\text{kHz}$ and $\eta = 0.1$. The

carrier term that was dropped in the rotating wave approximation of the sideband oscillations would have $\delta = 2\pi \cdot 1\text{MHz}$. The maximum probability of the carrier off-resonantly driving a transition on the sideband tone would then be 4%.

3.3 State Readout

Fluorescence emitted by the decay $P_{\frac{1}{2}} \Rightarrow S_{\frac{1}{2}}$, or the lack thereof, serves as the sole source of readout for information regarding the ions. When viewing Ca^+ as a simple three-level Λ -system, it may first seem like there isn't much information that can be gleaned from the fluorescence. However, the narrow line performance of the quadrupole laser enables single sideband addressing, which provides spin-motional coupling that encodes motional information into the electronic state of the Ca^+ ion.

3.3.1 Blue Sideband Rabi Flopping

Readout on the blue sideband of the ion is the typical method used to determine properties of a motional state. For a single Ca^+ , the probability of being in the $|S_{\frac{1}{2}}\rangle = |\downarrow\rangle$ state is

$$P_{\downarrow}^{BSB} = \frac{1}{2} \sum_{n=0}^{\infty} (1 + \cos(\Omega_{n+1,n}t)) P_n \quad (3.43)$$

when measuring on the blue sideband of a motional mode and

$$P_{\downarrow}^{RSB} = P_0 + \frac{1}{2} \sum_{n=1}^{\infty} (1 + \cos(\Omega_{n-1,n}t)) P_n \quad (3.44)$$

for the red sideband. The Rabi rates are

$$\begin{aligned} \Omega_{n,n} &= \Omega_0 \\ \Omega_{n+1,n} &= \eta\sqrt{n+1}\Omega_0 \\ \Omega_{n-1,n} &= \eta\sqrt{n}\Omega_0. \end{aligned} \quad (3.45)$$

Readout is most commonly performed on thermal, squeezed, or coherent states shown in Fig. 3.6. Each of these states has its own probability conversion. The thermal state has already been analyzed in section 2.2.4 as well as the squeezed state in section 2.2.5, and the same procedure to determine P_n for coherent states will be reiterated here. The probability distribution for a state with density operator ρ is

$$P_n = \langle n | \hat{\rho} | n \rangle, \quad (3.46)$$

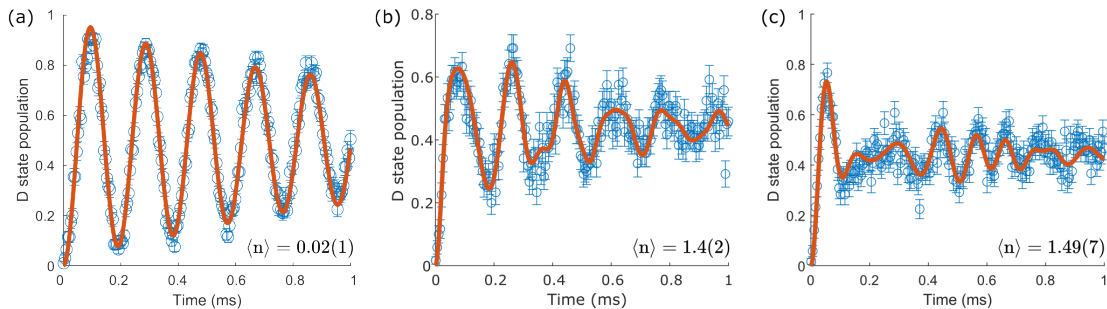


Figure 3.6: Blue sideband (BSB) Rabi oscillation for different motional states. The coherence time t_{dec} is roughly $1400 \mu\text{s}$, and the Rabi frequency (Ω_{00}) on resonance with the qubit transition is $2\pi \cdot 110 \text{ kHz}$. (a) BSB Rabi oscillations after sideband cooling to the motional ground state and fitted using a thermal state. (b) BSB Rabi oscillations for a squeezed state, which is prepared through parametric modulation of the trap potential at twice the secular frequency. (c) BSB Rabi oscillations for a coherent state produced using the QVSA interaction. Error bars represent one standard error.

which for a coherent state $\hat{\rho}_\alpha = |\alpha\rangle\langle\alpha|$ results in

$$P_n = \langle n | \hat{\rho}_\alpha | n \rangle = |\langle n | \alpha \rangle|^2 = e^{-|\alpha|^2} \frac{|\alpha|^{2n}}{n!}, \quad (3.47)$$

where $|\alpha|^2$ is the average phonon number for a coherent state.

To determine $\bar{n}$, $|\alpha|^2$, or $|\xi|^2$ we need to decompose the data into its different Fock state populations. Modifying Eqn.3.43 to include decoherence and imperfect flopping amplitude results in

$$P_{\downarrow}^{BSB} = \frac{A}{2} \sum_{n=0}^{n_{max}} p_n \left(1 + e^{-\gamma t \sqrt{n+1}} \cos(\Omega_{n+1,n} t) \right) \quad (3.48)$$

where p_n is the population in the n^{th} Fock state, up to the n_{max} Fock state. An initial calibration curve is performed on the vacuum state to determine the effective Rabi rate Eqn.3.45, the amplitude, and the decoherence rate for a given n_{max} . Performing a linear fit using Eqn.3.48 and the measured $P_{\downarrow}$ probability determines the probability of each Fock state within n_{max} . By decomposing the blue sideband fit into the different Fock state components [MMK96a, DH15], we can compare the resultant probabilities to the distribution for a given state such as Eqn.3.47.

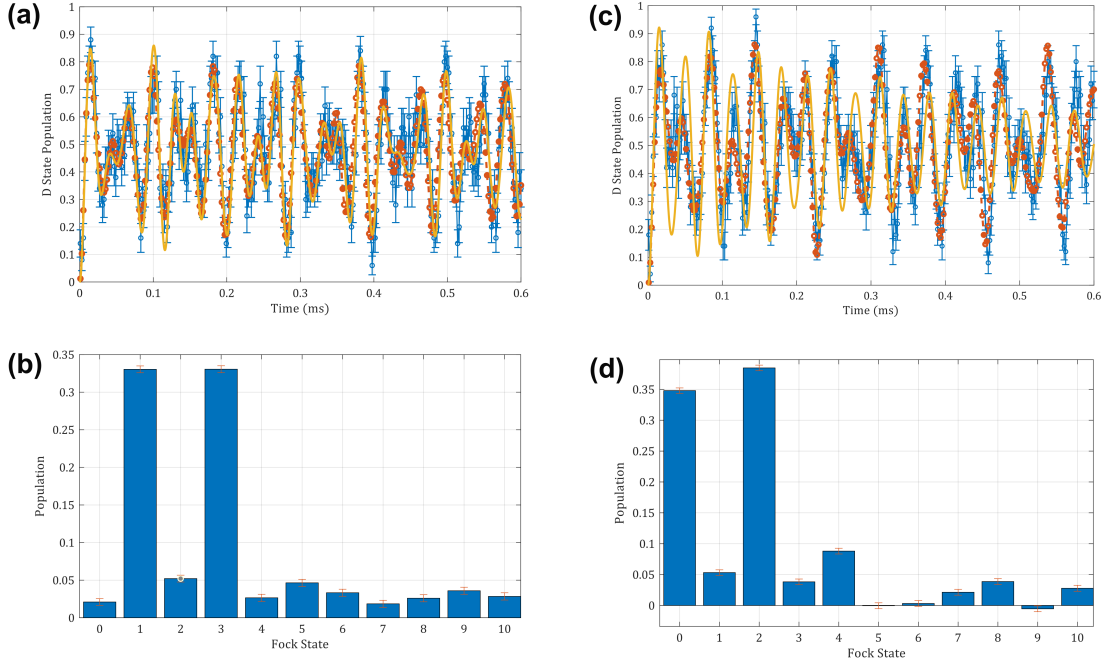


Figure 3.7: Blue sideband Rabi (BSB) and Fock State Decomposition (FSD) for odd (a)/(b) and even (c)/(d) CAT states. Using bichromatic light about the 729nm $S_{\frac{1}{2}} \Leftrightarrow D_{\frac{5}{2}}$ transition can generate a CAT state, which is read out using Fock state decomposition. In (a) and (c) the blue points are the RAW data, the orange are the FSD values, which utilized a calibration vacuum state BSB dataset, and the yellow is a fit of the FSD values. From the fit in (a), $|\alpha|^2 = 2.594$, $\Omega_{n+1,n} = 2\pi \cdot 35.22\text{kHz}$, $A=0.95$, and $t_{dec} = 2.525\text{ms}$. From the fit in (c), $|\alpha|^2 = 1.135$, $\Omega_{n+1,n} = 2\pi \cdot 43.07\text{kHz}$, $A=0.99$, and $t_{dec} = 0.7\text{ms}$. Visually, the fit in (a) appears better than in (c) which would explain some of the discrepancies in the fit solutions.

3.3.2 Sideband Ratio

Motional states are addressed using narrow linewidth 729nm light by tuning the light to be resonant with the motional sidebands of the atomic transition. When the laser is tuned to the bare transition, i.e. not to a secular or micromotion sideband, it is referred to as a carrier transition. Unless specified otherwise, a sideband transition in this work refers to the sidebands associated with the secular motion of the ion within the trap, not the sidebands that result from the micromotion of the ion within the trap. To read out motional information, the simplest method is to detect the probabilities of driving both a red and a blue sideband. The ratio of the probabilities enables us to convert to a phonon number, given an understanding of what the motional state will look like [LBM03, TKK00]. Consider an ion is cooled to its motional ground state, such that the ion is in $|S_{\frac{1}{2}}, n = 0\rangle$. If the quadrupole laser is tuned to the red sideband, it can only be absorbed if the ion is in state $|S_{\frac{1}{2}}, n \geq 1\rangle$, which would be unavailable for the ground state cooled ion. Tuning the laser to the blue sideband will add a phonon to the motional state, so it can always be driven, and for a ground state cooled ion, it will always be driven. The ratio for the ground state cooled ion is $\frac{P_{RSB}}{P_{BSB}} = 0$.

To determine the phonon number (or squeeze parameter ξ), the ratio of the two sidebands $\frac{P_{RSB}}{P_{BSB}}$ is compared to a numerically calculated function dependent on what the state is believed to be. The look-up function is generated by inserting the population distribution equations 2.57, 3.47, or 2.63 into equations 3.44 and 3.43. A ratio of sidebands is generated for a given phonon number in the thermal or coherent case, and $|\xi|^2$ in the squeezed case.

3.3.3 Rapid Adiabatic Passage

An alternative method for determining the average phonon number of a coherent state $|\alpha\rangle$ ($\bar{n} = |\alpha|^2$) is by using rapid adiabatic passage [Lec16, CLR10, NB01]. This measures motional overlap of the coherent state with a motional ground state $|n = 0\rangle$. To implement this scheme, the quadrupole laser is shaped and chirped, whereby the laser has both its intensity and frequency swept during the pulse duration. For our

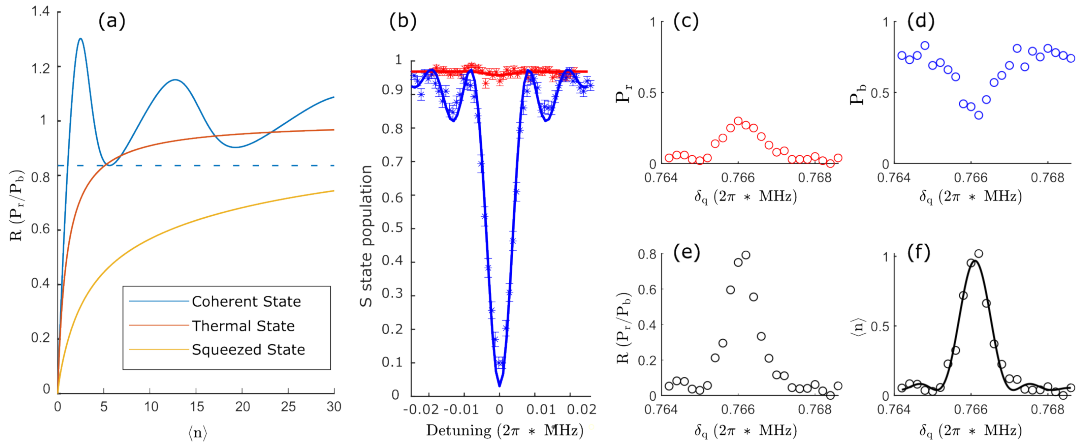


Figure 3.8: Sideband ratio measurement of $\langle n \rangle$. (a) Conversion of the sideband ratio R to a mean phonon number $\langle n \rangle$ for different motional states. The sidebands are interrogated for time $t_\pi = \pi/\Omega_{01}$, where Ω_{01} is the Rabi frequency for the $|S, n=0\rangle$ to $|D, n=1\rangle$ transition. To ensure injectivity as $R \rightarrow \langle n \rangle$, we enforce $R < 0.829$ for coherent states, depicted by the dashed line. (b) A typical red/blue sideband spectrum after ground state cooling. Assuming a thermal state, the spectra show cooling to $\langle n \rangle = 0.01 \pm 0.01$. Error bars represent one standard error. (c-f) Typical sideband spectra after a displacement (via the QVSA interaction) is applied and the swept through resonance. (c) and (d) are the red and blue sideband spectra, respectively. (e) shows the resulting sideband ratio. The ratio is converted to mean phonon number $\langle n \rangle$ in (f).

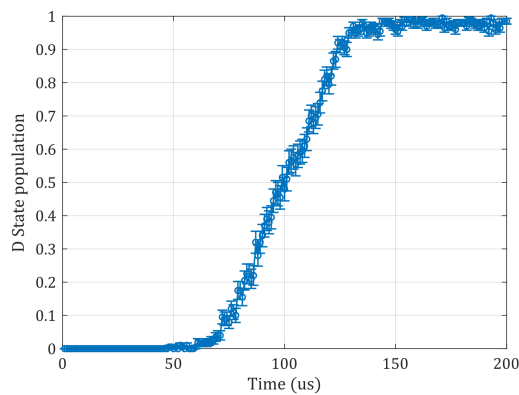


Figure 3.9: Applying a blue sideband RAP pulse on an ion prepared in the ground motional and ground electronic state transfers the entire population to the excited electronic state, while also adding a phonon into the QHO. This figure demonstrates the fidelity of the RAP procedure, transferring population with $> 98\%$.

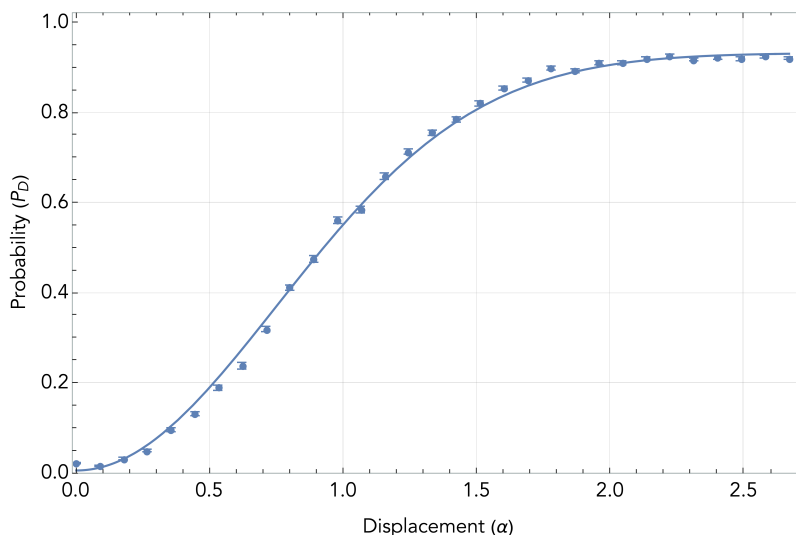


Figure 3.10: A motional overlap measurement of a coherent state can easily be implemented using the RAP protocol. Unlike the sideband ratio method, RAP is able to measure coherent states with $\bar{n} > 1$ but will still have an eventual limit around $\bar{n} \approx 4$.

experiment, this is achieved using a Blackman window given by

$$\Omega(t) = \frac{\Omega}{2} \left(0.84 - \cos\left(\pi \frac{t}{t_{slope}}\right) + 0.16 \cos\left(2\pi \frac{t}{t_{slope}}\right) \right) \quad (3.49)$$

The pulse frequency is swept through $\Delta - \omega_0$ for a given mode, in essence applying $\mathbb{I} \otimes |n=0\rangle\langle n=0| + \sigma_+ \hat{a}$. As a result, the initial state population gets transferred into the final state without leaving population within some intermediate state $|S_{\frac{1}{2}}, n \neq 0\rangle \rightarrow |D_{\frac{5}{2}}, n-1\rangle$. Laser-induced fluorescence via the 397nm light will projectively measure the $n=0$ overlap since all higher-phonon-number populations are moved from $S_{\frac{1}{2}}$ to the $D_{\frac{5}{2}}$ by

$$\langle g, n | \left(\mathbb{I} \otimes |n=0\rangle\langle n=0| + \sigma_+ \hat{a} \right) | g, \alpha \rangle \quad (3.50)$$

$$= \langle g, n | \left(e^{-\frac{|\alpha|^2}{2}} |g, n=0\rangle + e^{-\frac{|\alpha|^2}{2}} \sum_{n=1}^{\infty} \frac{\alpha^n}{\sqrt{(n-1)!}} |e, n-1\rangle \right) = e^{-\frac{|\alpha|^2}{2}}. \quad (3.51)$$

This behavior is similar to just driving the red sideband with a π -pulse but the chirping introduces a more robust measurement with respect to pulse area (frequency and time) [WHK07].

This adiabatic transfer requires a longer pulse than the equivalent sideband read-out method, typically $500 - 1000\mu\text{s}$ as opposed to $50 - 100\mu\text{s}$. However, since this

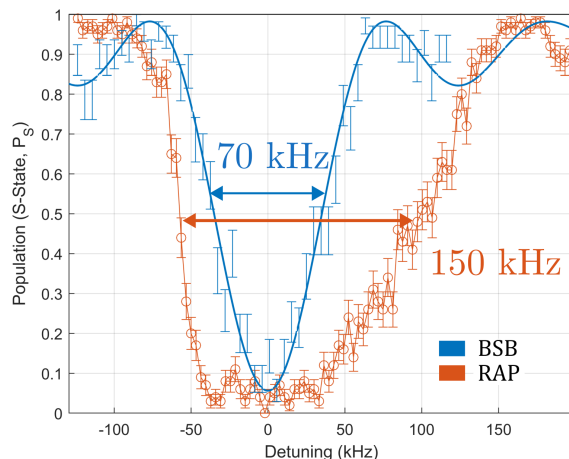


Figure 3.11: To demonstrate that the RAP procedure is more robust to frequency and timing errors compared to the sideband readout method, both readout methods are overlaid on this figure, BSB shown in blue and RAP shown in orange. A larger pulse duration shows the correct population transfer using RAP than using the BSB method.

method relies on a measurement of only the red sideband, it requires half as many measurements. Since the cooling of the ion takes up 5-10ms, this is more efficient for faster measurements.

3.3.4 Wigner Reconstruction

The Wigner probability distribution function is a convenient way to picture the state of a quantum harmonic oscillator, which is generated by its characteristic function $\chi(\beta)$, the mean displacement given by Eqn. 2.40. A procedure for mapping the state of the QHO onto the internal spin state of the ion is presented in [FH20] and outlined briefly here. The QHO state is entangled with the spin of Ca^+ via a pair of bichromatic tones tuned to the motional sidebands of a transition, so a fluorescence measurement will provide insight into the state overlap of the harmonic wavefunction. Fluorescence detection is equivalent to $\langle \psi | \sigma_Z | \psi \rangle$ where $|\psi\rangle$ is the state prior to the measurement. In order to scan the two-dimensional Wigner space, two operations are done prior to readout. A rotational operation is applied onto the spin state of

the atom using a carrier pulse given by

$$\hat{R}(\vartheta, \varphi) = \cos\left(\frac{\vartheta}{2}\right) \mathbb{I} - \imath \sin\left(\frac{\vartheta}{2}\right) \left(\cos(\varphi) \sigma_x + \sin(\varphi) \sigma_y \right) \quad (3.52)$$

where ϑ and φ are the rotation angles about the Cartesian axes in the Bloch basis. The phases are determined by the pulse duration and the laser phase. The second operation is a bichromatic displacement to create a state-dependent force given by $\hat{D}(\frac{\beta}{2} \sigma_x)$, where the strength of the displacement is $\beta = 2\eta\Omega t e^{\imath \frac{\Delta\varphi}{2}}$ with Ω and $\Delta\varphi$ determined by the amplitude and relative phases of the two laser tones. Prior to measurement, the state will be

$$|\psi\rangle = \frac{1}{\sqrt{2}} \left[\left(\cos\left(\frac{\vartheta}{2}\right) - \imath \sin\left(\frac{\vartheta}{2}\right) \right) \hat{D}\left(\frac{\beta}{2} \sigma_x\right) |\psi_0\rangle \otimes |+_x\rangle \right. \quad (3.53)$$

$$\left. \left(\cos\left(\frac{\vartheta}{2}\right) + \imath \sin\left(\frac{\vartheta}{2}\right) \right) \hat{D}\left(-\frac{\beta}{2} \sigma_x\right) |\psi_0\rangle \otimes |-_x\rangle \right] \quad (3.54)$$

using $\varphi = 0$ for maximum contrast, resulting in a fluorescence state measurement of

$$\langle\psi| \sigma_Z |\psi\rangle = \cos(\vartheta) \text{Re}[\chi(\beta)] + \sin(\vartheta) \text{Im}[\chi(\beta)]. \quad (3.55)$$

Sweeping the displacement β and the phase ϑ will allow for a two-dimensional mapping of the characteristic function, shown in (a) in Fig. 3.12, before being mapped onto the Wigner space using a discrete Fourier transform (b). For improved visual results, the RAW data is fit to the expected characteristic function prior to the transform; (c) in Fig. 3.12 is fitted using a coherent state distribution, resulting in the distribution shown in (d).

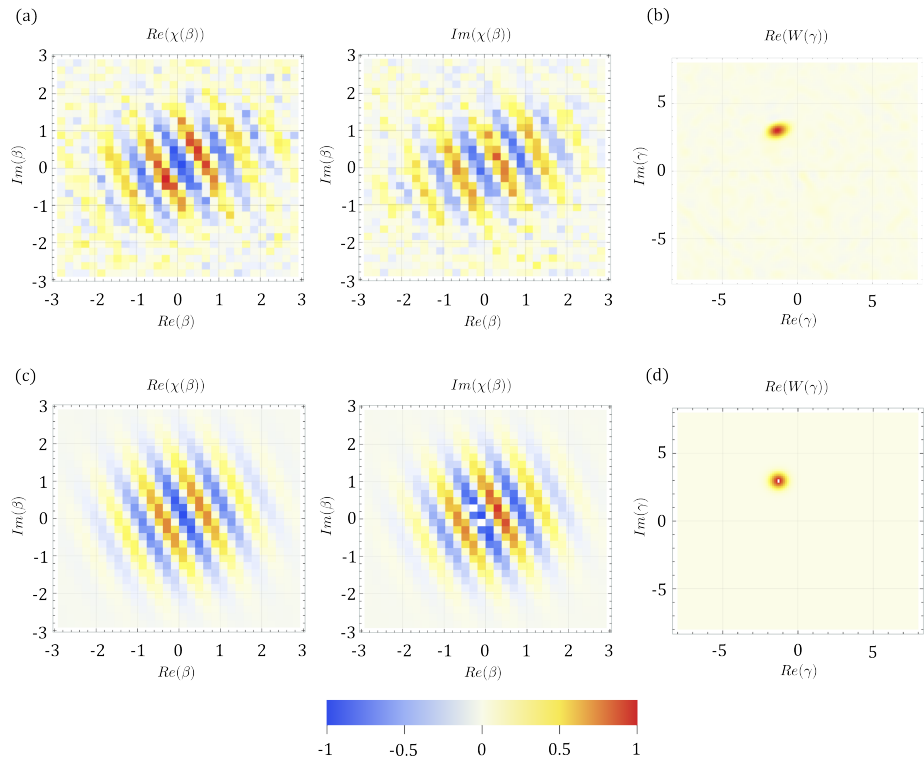


Figure 3.12: The top row (a) is the RAW data for the characteristic function of a coherent state taken during the experiment and the resultant Wigner distribution. The bottom row (b) is the fit results for the RAW data and the Wigner distribution that is generated from the results of the fits. Red coloration depicts $\langle \sigma_z \rangle = +1$ and blue depicts $\langle \sigma_z \rangle = -1$, where ± 1 denotes the $D_{\frac{5}{2}}$ or $S_{\frac{1}{2}}$ state, respectively.

CHAPTER 4

Diatomic Molecules

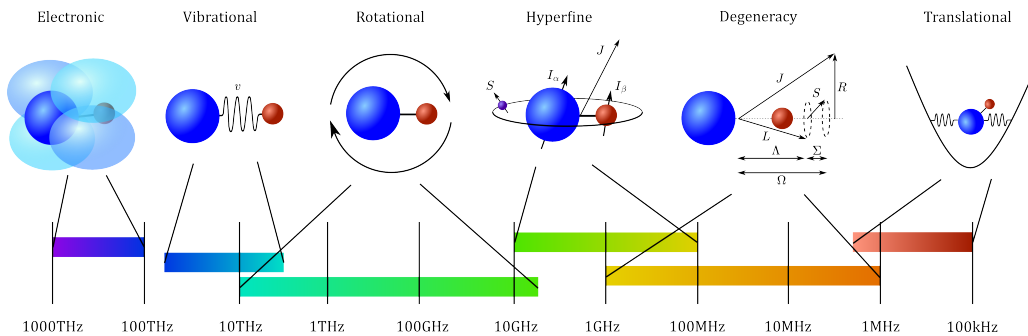


Figure 4.1: Molecules possess a plethora of different states, spanning regions of the electro-magnetic spectrum that are not normally filled with atomic transitions. These additional transitions come from changing ro-vibrational levels in addition to degenerate levels such as the Λ -doublets or Ω -doublets.

The shift from atoms to molecules is not for the faint of heart. Diatomic molecules are the simplest category of molecules, but already introduce many different possible states such as the ro-vibrational states. All the previously established states from atoms still persist, although new selection rules may open up pathways that would otherwise be forbidden. The Hamiltonian for a diatomic molecule takes the form

$$H_{total} = H_E + H_{vib} + H_{rot} + H_{SO} + H_{SR} + H_{HF} + H_{\Lambda}, \quad (4.1)$$

where H_E , H_{SO} , and H_{HF} are the components from the electronic, spin-orbit, and hyperfine terms of the Hamiltonian similar to those for atoms. The components H_{vib} , H_{rot} , H_{SR} , and H_{Λ} are unique to molecules and are the vibrational, rotational, spin-rotational, and Λ -doublet terms of the Hamiltonian, respectively.

Prior to defining the different cases, it will be instructive to have an organized list of the different angular momenta, see Tab.4.1. Calculations for the different Hamiltonian components as well as molecular properties follow primarily from [MC03],

but [AF05], [Her64], and [Wei78] have been excellent supplementary texts for studying molecules.

4.1 Electronic

Molecular notation for the electronic states is very similar to atom notation, with the exception that it is in Greek. In an atomic system, we use $^{2S+1}L_J$ to denote the spin (S), orbital momentum (L), and total angular momentum excluding nuclear spin (J). In a molecular system, we use $^{2S+1}\Lambda_{\Omega}$. Orbital momentum in atoms goes as S, P, D ...; orbital momentum in molecules goes as $\Sigma, \Pi, \Delta, \dots$, each step being one integer step above the last. The value J remains the total angular momentum excluding nuclear spin, but for molecules it will need to be calculated depending on the strength of the spin-orbital coupling to the internuclear axis. The different ways are separated into the different Hund's cases.

4.2 Vibrational

The potential energy well that binds the atoms together to form the molecule is deep enough that the motion of the atoms about some equilibrium position can be modeled by a harmonic oscillator of frequency ω_e . To correct for anharmonicities in the potential well, additional terms can be included in the Hamiltonian, resulting in

$$\frac{H_{vib}}{\hbar} = \omega_e \left(\nu + \frac{1}{2} \right) - \omega_e \chi_e \left(\nu + \frac{1}{2} \right)^2 + \omega_e y_e \left(\nu + \frac{1}{2} \right)^3 \dots \quad (4.2)$$

where χ_e and y_e are the first- and second-order corrections to the anharmonic oscillator. The value ν is the quantum number associated with the vibrations of the atoms within the molecule.

4.3 Rotational

Just like how two points define a line, two nuclei in a molecule determine an axis of rotation, the internuclear axis, which plays an important role in defining both the energy associated with rotations as well as the other good quantum numbers. The

Symbol	Meaning	Example
Electronic		
η	Electronic State	X, A, B, ...
Ro-Vibrational		
ν	Vibrational quantum number	0, 1, 2,...
R	Rotational quantum number	0, 1, 2,...
Spin-Orbit		
S	Spin	$0, \frac{1}{2}, 1, \frac{3}{2}, \dots$
Σ	Spin Projection onto Internuclear Axis	$-\mathbf{S}, -\mathbf{S}+1, \dots, \mathbf{S}-1, \mathbf{S}$
L	Orbital Angular Momentum	0, 1, 2, ...
Λ	Orbital Angular Projection onto Internuclear Axis	0, 1, 2,.. L
Ω	Total Angular Projection onto Internuclear Axis	$\Omega = \Lambda + \Sigma $
R	Nuclear Rotation	0, 1, 2,...
K	Case B Coupling	K = $\Lambda + \mathbf{R}$
J_a	Case C and E Coupling	J_a = L + S
N	Total Angular Momentum Excluding Spin	N = J – S
J	Total Angular Momentum	J = L + S + R
Nuclear Couplings		
I₁	Nuclear Spin of Atom 1	$0, \frac{1}{2}, 1, \frac{3}{2}, \dots$
I₂	Nuclear Spin of Atom 2	$0, \frac{1}{2}, 1, \frac{3}{2}, \dots$
F₁	Hyperfine	F₁ = J + I₁
F	Hyperfine	F = F₁ + I₂

Table 4.1: Symbols for different molecular quantum numbers and their defined or typical values. Different angular momenta for various Hund’s cases are included for completeness.

rotational quantum number $\mathbf{R}$ is introduced using the simplest model, two masses fixed at a constant length from one another, rotating about some point on this axis. Referred to as the rigid rotor or dumbbell model, this simple picture can lead to a Hamiltonian of the form

$$H_{rot} = B\mathbf{R}^2 \quad (4.3)$$

where B is the strength of this interaction. The total angular momentum excluding nuclear spin, $\mathbf{J}$, is assembled from the orbital angular momentum, the electronic spin angular momentum, and this rotational angular momentum ($\mathbf{J} = \mathbf{L} + \mathbf{S} + \mathbf{R}$). For some Hund's case molecules, a different rotational quantum number, $\mathbf{N}$, is better, such that $\mathbf{J} = \mathbf{N} + \mathbf{S}$. As a result, using $\mathbf{N}$ is more typical, for example switching to the effective Hamiltonian

$$H_{rot} = B(\mathbf{N}^2 - N_z^2). \quad (4.4)$$

For a Hund's case (a) molecule, like HCl^+ , this then takes the form

$$E_{rot} = B(J(J+1) + S(S+1) - 2\Omega\Sigma - \Lambda^2) \quad (4.5)$$

where Ω , Σ , and Λ are the projections of $\mathbf{J}$, $\mathbf{S}$, and $\mathbf{L}$ onto the internuclear axis, respectively. In general, the energy splitting between two different rotational states, R and R' , can be simplified to

$$\Delta E_{rot} = B(J_{R'}(J_{R'}+1) - J_R(J_R+1)) \quad (4.6)$$

assuming only the rotational quantum number was changed.

Often the rigid rotor model starts to fail, and higher order terms need to be introduced to compensate for the fact that the molecule is better described as a vibrating rotor. The energy for the rotation is now given by

$$H_{rot,\nu} = B_\nu\mathbf{R}^2 - D_\nu\mathbf{R}^4 + H_\nu\mathbf{R}^6 + \dots \quad (4.7)$$

where B_ν , D_ν , and H_ν are the rotational constants for the given vibrational state, ν . A good rule of thumb for considering rotational energies is that the greater the difference in mass, the greater the rotational constant. Given that the ground state for a Hund's case (a) molecule is typically a $J = \frac{3}{2}$ state, the energy splitting will be $\Delta E_{rot} \approx 5B_0$, whereas a Hund's case (b) will be $\Delta E_{rot} \approx 3B_0$. For example, H^{35}Cl^+ has $B_0 = 293\text{GHz}$ and the transition between the lowest two rotational states is 1.44THz .

4.3.1 Black Body Radiation

Ro-vibrational energy levels typically undergo transitions due to black body radiation (BBR) from the surrounding environment, since the intensity of a thermal mass will typically be emitting in this range of the spectrum. For example, the peak intensity of a room temperature (293K) object will be around 17.4THz, whereas a 4K object emits at 230GHz. Special attention is being paid here, since black body radiation will set a lifetime estimate on most of the lower energy splittings, since most do not have selection rules to prevent redistribution.

4.4 Spin-Orbit Coupling: Hund's Cases

Not too surprisingly, the Hamiltonian component for the spin-orbital coupling takes the form

$$H_{SO} = A\mathbf{L} \cdot \mathbf{S}, \quad (4.8)$$

where A is the spin-orbit constant. Each molecule will have its own spin-orbit constant, which plays a defining role in determining what the good quantum numbers are for the molecule. The different categories are given by the Hund's cases. The other component that is considered when classifying molecules into these cases is the rotation of the molecule. Together, the strength of the spin-orbit coupling and the coupling of the momentum to the internuclear axis allow us to categorize molecules. For the spin angular momentum $\mathbf{S}$, its projection is given by

$$M_S = -S, -S + 1, \dots, S - 1, S. \quad (4.9)$$

The projection onto the internuclear axis for the orbital angular momentum is given by

$$M_L = -L, -L + 1, \dots, L - 1, L. \quad (4.10)$$

If the direction of motion is inverted for the orbit of the electrons in a molecule, the projection M_L would experience a sign change but the overall energy would remain unchanged, representing a degeneracy. In contrast, if the spin of the electrons were to be inverted so would M_S AND the energy associated with the spin. As such, the quantum numbers that are used to describe the projection of the spin and orbital

Hund's Case	Coupling Strengths	Quantum Numbers
(a)	$A\Lambda \gg B\mathbf{J}$	$\eta, \Lambda, \mathbf{S}, \Sigma, \mathbf{J}, \Omega$
(b)	$A\Lambda \ll B\mathbf{J}$	$\eta, \Lambda, \mathbf{S}, \mathbf{N}, \mathbf{J}$
(c)	$A\Lambda \gg \Delta E_{el}$	$\eta, \mathbf{J}_a, \mathbf{J}, \Omega$
(d)	$B\mathbf{J} \gg \Delta E_{el}$	$\eta, \mathbf{L}, \mathbf{R}, \mathbf{N}, \mathbf{S}, \mathbf{J}$
(e)	$A\Lambda \gg B\mathbf{J} \gg \Delta E_{el}$	$\eta, \mathbf{J}_a, \mathbf{R}, \mathbf{J}$

Table 4.2: A is spin-orbit coupling strength, B is rotational coupling constant, and ΔE_{el} is the coupling to the internuclear axis. In case (a) and (b), the coupling to the internuclear axis is strong, whereas in (c), (d), and (e) it is weak. Case (a) molecules have strong spin-orbit coupling compared to the rotation, so both spin and orbital momentum have strong coupling to the internuclear axis. Case (b) molecules have a weak spin orbital coupling compared to the rotation. As such, the spin momentum is taken after the rotational momentum is considered. Case (c) molecules have a strong coupling between spin and orbital momenta but not a strong coupling to the nuclear axis, so they are added together before the rotational momentum is considered. Case (d) molecules have strong rotational coupling and weak spin-orbit coupling. Case (e) has strong spin-orbit coupling, but weak rotational and even weaker internuclear coupling.

angular momentum onto the internuclear axis are Σ and Λ , respectively, where

$$\Sigma = M_S = -S, -S + 1, \dots, S - 1, S \quad (4.11)$$

$$\Lambda = |M_L| = 0, 1, 2, \dots, L. \quad (4.12)$$

4.4.1 Hund's Case (a)

Molecules are categorized as Hund's case (a) if the spin orbital coupling is large compared to the coupling between the nuclear rotation and the electronic motion, and if the coupling of $\mathbf{L}$ (and by proxy $\mathbf{S}$) to the internuclear axis is also large. A large $\mathbf{L} \cdot \mathbf{S}$ coupling means that since $\mathbf{L}$ is strongly coupled to the internuclear axis, so should be $\mathbf{S}$. Because both are strongly coupled to the internuclear axis, their

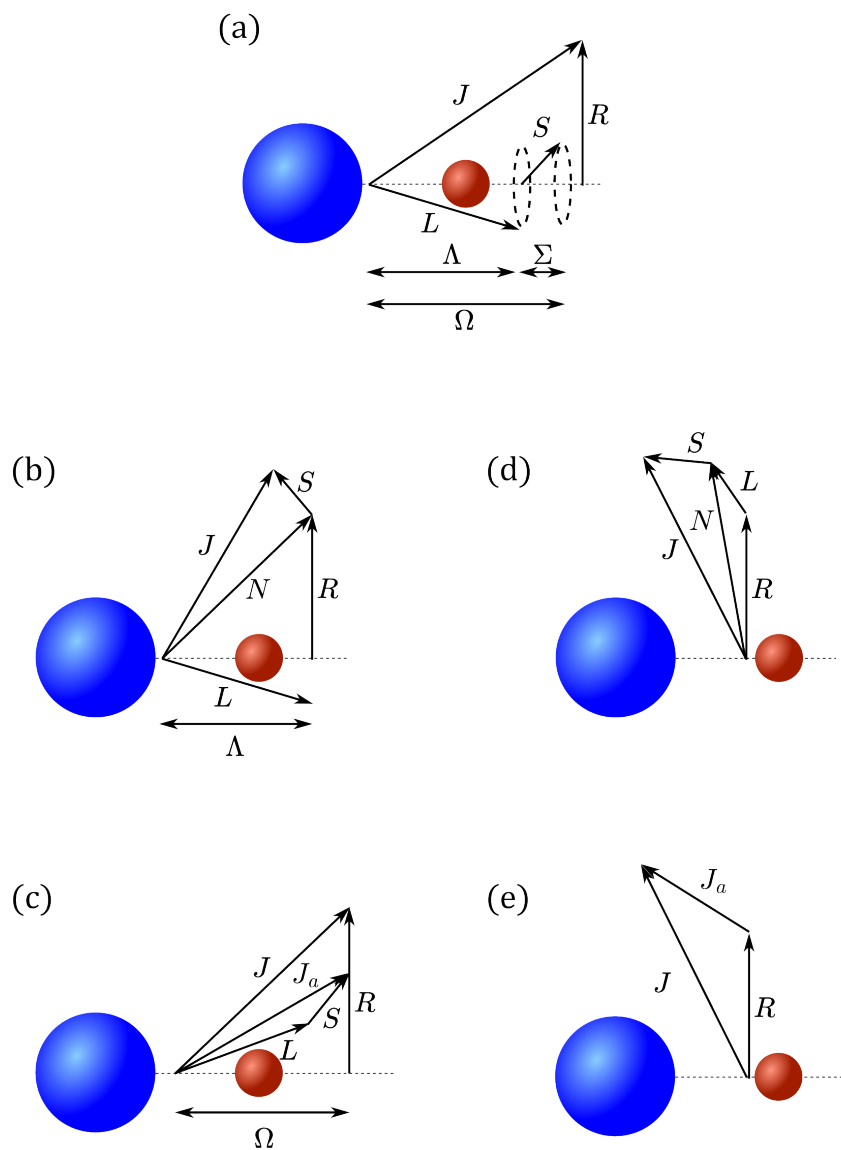


Figure 4.2: Diatomic molecules are categorized into different Hund's cases, determined by the strength of the spin-orbital interaction and the strength of the coupling to the internuclear axis. Case (a) and (b) are the most common cases.

projections onto it (Λ and Σ) are well defined quantum numbers, leading to the well-defined quantum number

$$\Omega = |\Lambda + \Sigma|. \quad (4.13)$$

Ω is the projection of $\mathbf{J}$ onto the internuclear axis. $\mathbf{J}$ is then defined by the vector sum of Ω and $\mathbf{R}$, the nuclear rotation quantum number. $\mathbf{R}$ is always taken to be perpendicular to the internuclear axis for a diatomic molecule.

A useful property that arises in case (a) molecules is the presence of multiplets. For a non-zero value of $\mathbf{L}$, there will be degeneracy with the projection Λ , as it can take either a positive or a negative value ($M_L = \pm\Lambda$). Similarly, the spin angular momentum $\mathbf{S}$ can also have degeneracy with its projection Σ . As a result there are multiple acceptable values of Ω . For example, HCl^+ in its ground electronic state ($\Lambda = 1, \mathbf{S} = \frac{1}{2}$) will have two possible values, $\Omega = |1 \pm \frac{1}{2}| = \frac{3}{2}$ and $\frac{1}{2}$. We write these two states as $^2\Pi_{\frac{3}{2}}$ and $^2\Pi_{\frac{1}{2}}$. Each of these states will still be degenerate in Λ , with an eventual doublet forming from the coupling to the Σ electronic state as well as to higher rotational states. Further discussion on these state will be provided, since these Λ -doublets are what we intend to address in HCl^+ .

4.4.2 Hund's Case (b)

The other most common category of molecules is case (b). Molecules in this category have a strong orbital coupling to the internuclear axis when $\Lambda \neq 0$. However, they have a weak spin-orbital coupling, such that the spin $\mathbf{S}$ is only weakly coupled to the internuclear axis. As a result, Σ is a bad quantum number. Instead, the quantum number $\mathbf{K}$ is introduced as

$$\mathbf{K} = \Lambda + \mathbf{R}, \quad (4.14)$$

where, as before, Λ is the projection of the orbital angular momentum. In case (b) molecules, this quantum number $\mathbf{K}$ is also the total angular momentum excluding spin ($\mathbf{N} = \mathbf{J} - \mathbf{S}$). $\mathbf{N} = \Lambda + \mathbf{R}$ does not hold for all cases however, as will be seen in case (d), hence the introduction of $\mathbf{K}$.

4.4.3 Hund's Case (c)

Hund's case (c) molecules are characterized by a strong spin-orbital coupling, but with a weak coupling of the orbital angular momentum to the internuclear axis. As a result, the vector sum of $\mathbf{L}$ and $\mathbf{S}$ is done before a projection to the internuclear axis. In this case, the new quantum number

$$\mathbf{J}_a = \mathbf{L} + \mathbf{S} \quad (4.15)$$

is introduced, with projection Ω onto the internuclear axis. The total angular momentum $\mathbf{J}$ is then given by $\mathbf{J} = \Omega + \mathbf{R}$.

4.4.4 Hund's Case (d)

Hund's case (d) molecules feature a strong coupling between the orbital angular momentum and the nuclear spin. The vector sum of these momenta results in $\mathcal{R} = \mathbf{L} + \mathbf{R}$. Spin is then coupled to this new term, producing $\mathbf{J}$. In molecules, projections onto the internuclear axis are no longer good quantum numbers.

4.4.5 Hund's Case (e)

As with case (d) molecules, quantum numbers associated with projections onto the nuclear axis are not good quantum numbers. In case (e) molecules, the spin-orbit coupling is so large that both $\mathbf{L}$ and $\mathbf{S}$ become bad quantum numbers, but their vector sum $\mathbf{J}_a$ remains a good value. The total angular momentum $\mathbf{J}$ is then the vector sum of $\mathbf{J}_a$ and $\mathbf{R}$.

4.5 Spin-Rotational Coupling

The electron's spin can also undergo a coupling to the rotation magnetic moment of the molecule, resulting in a Hamiltonian term that goes as

$$H_{SR} = \gamma \mathbf{R} \cdot \mathbf{S} = \gamma(\mathbf{J} \cdot \mathbf{S} - \mathbf{L} \cdot \mathbf{S} - \mathbf{S}^2), \quad (4.16)$$

with molecular constant γ .

4.6 Hyperfine

The hyperfine Hamiltonian can be expanded into the following

$$H_{HF} = H_{IL} + H_F + H_{dip} + H_Q \quad (4.17)$$

where H_{IL} is the orbital hyperfine component, H_F is the Fermi contact component, H_{dip} is the dipolar hyperfine component, and H_Q is the electric quadrupole coupling component. H_{IL} , H_F , and H_{dip} are all magnetic dipole hyperfine interactions. The diatomic nature of the molecule will introduce nuclear spins of both atomic nuclei, which will be denoted by the subscript, j . The orbital hyperfine Hamiltonian is

$$H_{IL} = \sum_j a_j \mathbf{T}^1(\mathbf{I}_j) \cdot \mathbf{T}^1(\mathbf{L}) \xrightarrow{q=0} \sum_j a_j I_{z,j} L_z, \quad (4.18)$$

which is the typical hyperfine interaction that exists, where a_j is a hyperfine coupling constant. The Fermi contact interaction comes from the electron spin and the nuclear spins' magnetic moments coupling to each other and takes the form

$$H_F = \sum_j b_{F,j} \mathbf{T}^1(\mathbf{S}) \cdot \mathbf{T}^1(\mathbf{I}_j), \quad (4.19)$$

where b_j is the hyperfine coupling constant associated with this interaction. The dipole-dipole coupling, also referred to as nuclear spin coupling,

$$\begin{aligned} H_{dip} &= - \sum_j \sqrt{10} g_s \mu_B g_j \mu_N \frac{\mu_0}{4\pi} \mathbf{T}^1(\mathbf{I}_j) \cdot \mathbf{T}^1(\mathbf{S}, \mathbf{C}^2) \\ &= \sum_j \frac{\sqrt{6}}{3} c_j \mathbf{T}_{q=0}^2(\mathbf{I}_j, \mathbf{S}) + \sum_{q=\pm 1} e^{-2iq\varphi} d_j \mathbf{T}_{2q}^2(\mathbf{I}_j, \mathbf{S}), \end{aligned} \quad (4.20)$$

has two hyperfine constants c_j and d_j which are the $q = 0$ and $q = \pm 1$ components of the rank 1 tensor inner product, respectively. The phase factor φ is the azimuthal angle of the electron orbital, and limits the non-zero matrix elements to interactions only between $|\eta, \Lambda = +1\rangle$ and $|\Lambda = -1\rangle$. The value g_j is the nuclear g factor for the j^{th} atom in the molecule.

While the magnetic dipole interactions are what typically first come to mind for the hyperfine energy levels, the electric quadrupole of the nuclei also plays an important role. The electric quadrupole hyperfine interaction

$$H_Q = -eT^2(\mathbf{Q}) \cdot T^2(\nabla \mathbf{E}) \quad (4.21)$$

will be of the form

$$H_Q = \frac{eQq_0(3I_z^2 - I^2) + eQq_2(I_x^2 - I_y^2)}{4I(2I - 1)}, \quad (4.22)$$

where eQq_0 and eQq_2 are the hyperfine constants.

4.7 Degeneracy

Previously, it was mentioned that for a Hund's case (a) molecule, a state with $\Lambda \neq 0$, the energies associated with a positive and a negative projection are degenerate. This degeneracy can be lifted by the spin-orbit and rotational Hamiltonians when there is mixing with other states, separated by $\Delta\Lambda = \pm 1$. The resultant splitting is referred to as the Λ -doublet. For HCl^+ , the ground state is $X^2\Pi$, and mixes with available $^2\Sigma$ and $^2\Delta$ states but has the dominant component coming from the first excited state, $A^2\Sigma$.

Mathematically, this mixing of states comes from a second order perturbation of several components of the rotational and spin-orbit Hamiltonian

$$\begin{aligned} H_{RSO} &= \mathbf{A}\mathbf{L} \cdot \mathbf{S} + B\mathbf{R}^2 = \mathbf{A}\mathbf{L} \cdot \mathbf{S} + B(\mathbf{J} - \mathbf{L} - \mathbf{S})^2 \\ &== \mathbf{A}\mathbf{L} \cdot \mathbf{S} + B(\mathbf{J}^2 + \mathbf{L}^2 + \mathbf{S}^2 - 2\mathbf{J} \cdot \mathbf{L} - 2\mathbf{J} \cdot \mathbf{S} + 2\mathbf{L} \cdot \mathbf{S}). \end{aligned} \quad (4.23)$$

To expand this Hamiltonian and indicate the relevant matrix components, we need to first switch to tensor notation where the relation

$$\mathbf{A} \cdot \mathbf{B} = T^1(\mathbf{A}) \cdot T^1(\mathbf{B}) = \sum_{q=-1}^{+1} (-1)^q T_q^1(\mathbf{A}) T_{-q}^1(\mathbf{B}) \quad (4.24)$$

is used. Before expanding the Hamiltonian, let's return to the two states that are getting mixed:

$$|\psi\rangle = |\Pi, \Lambda; S, \Sigma; J, \Omega, m_J\rangle \quad (4.25)$$

$$|\psi'\rangle = |\Sigma, \Lambda'; S, \Sigma'; J, \Omega', m_J'\rangle. \quad (4.26)$$

The total angular momentum $\mathbf{J}$ and the spin $\mathbf{S}$ will remain unchanged, but all other quantum numbers may change. The electron spin momentum is independent of the orbital state, so it is clear why it is held constant, and the total momentum is constant because, although the orbital momentum $\mathbf{L}$ will change by ∓ 1 the rotational

quantum number $\mathbf{R}$ will change as well by ± 1 . Expanding the Hamiltonian under these considerations results in

$$H_{RSO} = A \sum_{q=-1}^{+1} (-1)^q T_q^1(\mathbf{L}) T_{-q}^1(\mathbf{S}) + B \left[J(J+1 + S(S+1)) + \sum_{q=-1}^{+1} (-1)^q T_q^1(\mathbf{L}) T_{-q}^1(\mathbf{L}) \right. \\ \left. - 2 \sum_{q=-1}^{+1} T_q^1(\mathbf{J}) T_q^1(\mathbf{L}) - 2 \sum_{q=-1}^{+1} T_q^1(\mathbf{J}) T_q^1(\mathbf{S}) + 2 \sum_{q=-1}^{+1} (-1)^q T_q^1(\mathbf{L}) T_{-q}^1(\mathbf{S}) \right] \quad (4.27)$$

Not all of the components in Eqn. 4.27 will result in lifting the degeneracy of the Λ -doublet, only the non-zero matrix elements of a second order perturbation between the two states $|\psi\rangle$ and $|\psi'\rangle$ will. Discarding matrix elements that rely on remaining in the same electronic state simplifies the Hamiltonian to

$$\langle \psi | H_{RSO} | \psi' \rangle = - \langle \psi | \left((A + 2B) \sum_{q=\pm 1} (-1)^q T_q^1(\mathbf{L}) T_{-q}^1(\mathbf{S}) - 2B \sum_{q=\pm 1} T_q^1(\mathbf{J}) T_q^1(\mathbf{L}) \right. \\ \left. + B \sum_{q=\pm 1} (-1)^q T_q^1(\mathbf{L}) T_{-q}^1(\mathbf{L}) \right) | \psi' \rangle. \quad (4.28)$$

The first two matrix elements are non-zero only when $\Delta\Lambda = \pm 1$, which represent the interaction that can drive one parity state to another. The degeneracy is lifted with energy

$$E^{(2)}(J, S, \Lambda) = \frac{|\langle \psi | H_{RSO} | \psi' \rangle|^2}{E_{\Pi}^{(0)} - E_{\Sigma}^{(0)}} = \frac{|\langle \psi | H_{\Lambda D} | \psi' \rangle|}{E_{\Pi}^{(0)} - E_{\Sigma}^{(0)}} \quad (4.29)$$

where the Λ -doublet Hamiltonian is given by

$$H_{\Lambda D} = \sum_{q=\pm 1} e^{-2iq\varphi} (-q T_{2q}^2(\mathbf{J}, \mathbf{J}) + (p + 2q) T_{2q}^2(\mathbf{J}, \mathbf{S})) \quad (4.30)$$

or more simply

$$H_{\Lambda D} = -\frac{1}{2}q(J_+^2 e^{-2i\varphi} + J_-^2 e^{2i\varphi}) - \frac{1}{2}(p + 2q)(J_+ S_+ e^{-2i\varphi} + J_- S_- e^{2i\varphi}) \quad (4.31)$$

where φ is the same azimuthal phase that appears in the H_{dip} hyperfine term, and p and q are the molecular constants for the strength of the Λ splitting.

4.7.0.1 Parity

When a dipolar molecule is exposed to an oscillating electric field, an interaction can occur with the oscillating electric dipole moment of the molecule. These interactions

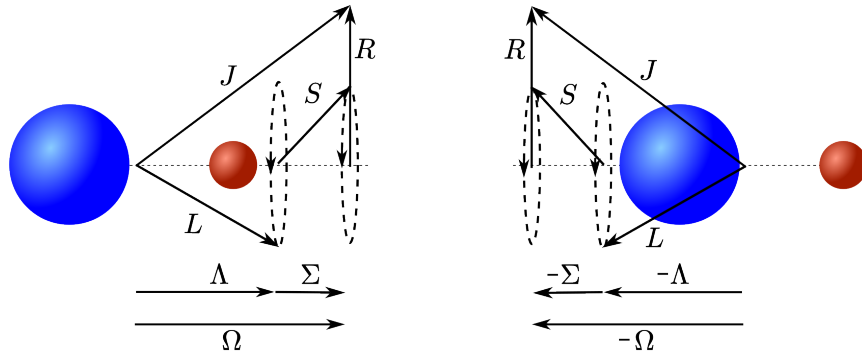


Figure 4.3: Illustration of the origin of the Λ -doublet: Λ and Σ can have different projections onto the internuclear axis.

are only allowed between states of opposite parity. Parity states exist between different rotational states, as well as from the Λ -doublets. The good quantum numbers for case (a) molecules were given in Table 4.2. Including the nuclear spins, the state of the molecule is given by

$$|\Psi_{\pm}\rangle = |\eta, \pm\Lambda; S, \pm\Sigma; J, \pm\Omega; I_1, F_1, I_2, F, m_F\rangle. \quad (4.32)$$

While these states form a good basis, they are not eigenstates of the parity operator. Being able to express the molecule's internal states as eigenstates of the parity operator offers us the ability to couple the internal states to an external electric field via the dipole of the molecule, given by $\hat{H} = -\mathbf{d} \cdot \mathbf{E}$. The parity states of the molecule can be expressed by

$$|+\rangle = \frac{1}{\sqrt{2}}(|\eta, \Lambda; S, \Sigma; J, \Omega; I_1, F_1, I_2, F, m_F\rangle + (-1)^{J-S} |\eta, -\Lambda; S, -\Sigma; J, -\Omega; I_1, F_1, I_2, F, m_F\rangle) \quad (4.33)$$

$$|-\rangle = \frac{1}{\sqrt{2}}(|\eta, \Lambda; S, \Sigma; J, \Omega; I_1, F_1, I_2, F, m_F\rangle - (-1)^{J-S} |\eta, -\Lambda; S, -\Sigma; J, -\Omega; I_1, F_1, I_2, F, m_F\rangle) \quad (4.34)$$

Note that the $|+\rangle$ and $|-\rangle$ states depend on the rotational state of the molecule through the quantum number J , in addition to the native $\pm$ sign. For simplicity, an alternative convention is often utilized, whereby the higher energy parity state

is denoted as $|f\rangle$ and the lower energy state by $|e\rangle$. Additional discussion on this notation is provided in [J 75].

When driving these parity transitions, standard selection rules must still be obeyed. For parity transitions between Λ -doublets, the molecule will remain in the ground ro-vibrational state, so the total angular momentum $\mathbf{J}$, its projection Ω , as well as Λ and Σ must all remain unchanged. Since the transition is driven using an electric dipole, it will not result in a change in nuclear spin for either atom (I_j), so the total angular momentum $\mathbf{F}$ follows the typical hyperfine selection rules $\Delta\mathbf{F} = 0, \pm 1$, $\Delta F \neq 0$ for $\mathbf{F} = 0$, and for its projections, $\Delta m_F = 0, \pm 1$.

4.8 Zeeman Splittings

The previous Hamiltonian terms have all been introduced in the absence of a field, whereas in reality the molecule will exist within some electric and magnetic field. Weak electric fields will shift molecular states, but arguably more important are the effects from the presence of the magnetic field, which will split the states. The Hamiltonian component representing the Zeeman effect is given by

$$H_Z = g_L\mu_B T^1(\mathbf{B}) \cdot T^1(\mathbf{L}) + g_S\mu_B T^1(\mathbf{B}) \cdot T^1(\mathbf{S}) - \sum_j g_j\mu_N T^1(\mathbf{B}) \cdot T^1(\mathbf{I}_j) - g_r\mu_B T^1(\mathbf{B}) \cdot T^1(\mathbf{R}) \quad (4.35)$$

where the first product is the orbital Zeeman effect, followed by the electron spin term, the nuclear hyperfine terms for each nucleus, and the rotational magnetic moment term, respectively. Each prefactor $g_i\mu_i$ indicates the relative strength of the interaction: $g_L\mu_B \approx \mu_B$, $g_S\mu_B \approx 2\mu_B$, $g_j\mu_N \ll \mu_B$, and $g_r\mu_B \ll \mu_B$. The nuclear magneton (μ_N) is more than 10^{-3} weaker than the Bohr magneton (μ_B), which reduces the nuclear component, even though the nuclear g-factor is more than an order larger than the electron g-factor for H^{35}Cl^+ ($g_H + g_{Cl} \approx 31.673$). For extremely sensitive spectroscopy measurements, the Zeeman terms for the nuclear and rotational terms should be considered, but for our experiment these terms can be dropped without consequence. The rotational term will be rewritten in terms of the other quantum numbers $\mathbf{R} = \mathbf{J} - \mathbf{L} - \mathbf{S}$. A rigorous derivation using the Wigner-Eckart method is provided in an appendix, but the results will be summarized here.

Matrix elements $\langle \Psi | H_Z | \Psi' \rangle$ are given by the following

$$\langle \Psi | T_p^1(\mathbf{B}) \cdot T_p^1(\mathbf{L}) | \Psi' \rangle = B_z \Lambda (-1)^{F-m_F+F'+2J+I+1-\Omega} \begin{Bmatrix} J & F & I \\ F' & J & 1 \end{Bmatrix} \begin{pmatrix} F & 1 & F' \\ -m_F & 0 & m_F \end{pmatrix} \begin{pmatrix} J & 1 & J \\ -\Omega & 0 & \Omega \end{pmatrix} \sqrt{(2F+1)(2F'+1)(2J+1)(2J+1)}, \quad (4.36)$$

$$\langle \Psi | T_p^1(\mathbf{B}) \cdot T_p^1(\mathbf{S}) | \Psi' \rangle = B_z (-1)^{F-m_F+F'+2J+I+1-\Omega+S-\Sigma} \sum_{q=-1}^{+1} \begin{Bmatrix} J & F & I \\ F' & J & 1 \end{Bmatrix} \begin{pmatrix} F & 1 & F' \\ -m_F & 0 & m_F \end{pmatrix} \begin{pmatrix} J & 1 & J \\ -\Omega & q & \Omega \end{pmatrix} \begin{pmatrix} S & 1 & S \\ -\Sigma & q & \Sigma \end{pmatrix} \sqrt{(2F+1)(2F'+1)(2J+1)(2J+1)(2S+1)(S+1)S}, \quad (4.37)$$

$$\langle \Psi | T_p^1(\mathbf{B}) \cdot T_p^1(\mathbf{J}) | \Psi' \rangle = B_z (-1)^{F-m_F+F'+J+I+1} \sum_{q=-1}^{+1} \begin{Bmatrix} J & F & I \\ F' & J & 1 \end{Bmatrix} \begin{pmatrix} F & 1 & F' \\ -m_F & 0 & m_F \end{pmatrix} \sqrt{(2F+1)(2F'+1)(2J+1)(J+1)J}, \quad (4.38)$$

and

$$\langle \Psi | T_p^1(\mathbf{B}) \cdot T_p^1(\mathbf{I}) | \Psi' \rangle = B_z (-1)^{F-m_F+F'+J+I+1} \sum_{q=-1}^{+1} \begin{Bmatrix} I & F & J \\ F' & I & 1 \end{Bmatrix} \begin{pmatrix} F & 1 & F' \\ -m_F & 0 & m_F \end{pmatrix} \sqrt{(2F+1)(2F'+1)(2I+1)(I+1)I}. \quad (4.39)$$

Alternatively, Eqn. 4.35 can be evaluated by doing a similar derivation as was done for Eqn. 3.10, producing a simplified energy shift of the form

$$\Delta E_Z = \mu_B g_F F_z B_z \quad (4.40)$$

where $F_z = m_F$ and the g-factor with the nuclear spin included is

$$g_F = g_J \frac{F(F+1) - I(I+1) + J(J+1)}{2F(F+1)} + g_N \frac{\mu_N}{\mu_B} \frac{F(F+1) + I(I+1) - J(J+1)}{2F(F+1)} \approx g_J \frac{F(F+1) - I(I+1) + J(J+1)}{2F(F+1)} \quad (4.41)$$

where we see the latter vanishes since the strength of $g_N\mu_N$ is much weaker than $g_J\mu_B$.

4.9 Translational Motion

The last regime for molecular ions is their translational energy, i.e. the energy associated with their motion within the trap. Ions, regardless of whether they are atoms or molecules, will have their translational behavior described by the dynamics of section 2.2. What separates the two has to do with what couplings each has between its spin and its motion within the trap. For our atom, Ca^+ , the interactions with the lasers allow us to achieve Doppler cooling, sideband cooling, state readout and some other techniques useful in quantum computing and sensing. For our molecule, HCl^+ , we will explore driving displacements using the internal parity states in a later chapter. Regardless, the shared motion of the two ions makes the chain as a whole susceptible to light that affects either of its constituents.

4.10 HCl^+ Molecular Structure

Several candidates are under consideration for implementing electric-field gradient gates, with the initial choice being HCl^+ . Rotational spectroscopy performed on this ion determined many of its constants, categorizing it as a Hund's case (a) molecule. In addition, a 2+1 resonantly enhanced multiphoton photoionization (REMPI) scheme has already been demonstrated in [MW01], ensuring that it can be loaded into a trap filled with a Ca^+ ion. Neutral HCl 's π_{nb} states are the highest energy states with electrons and are fully filled (closed shell). When ionized, an electron is removed from one of these states, resulting in the ground state of HCl^+ being $X^2\Pi$. When taking into account the spin-orbit coupling, the two states are $X^2\Pi_{\frac{3}{2}}$ and $X^2\Pi_{\frac{1}{2}}$, with the former being the ground state. The components of HCl^+ have nuclear spins of $\mathbf{I}_1 = \frac{3}{2}$ and $\mathbf{I}_2 = \frac{1}{2}$, for chlorine and hydrogen, respectively. Chlorine's nuclear spin will dominate the hyperfine interaction $\mathbf{F}_1 = 3, 2, 1, 0$, which can be used to get rough estimates of the A -doublets. By including the nuclear spin of the hydrogen, the F states split further.

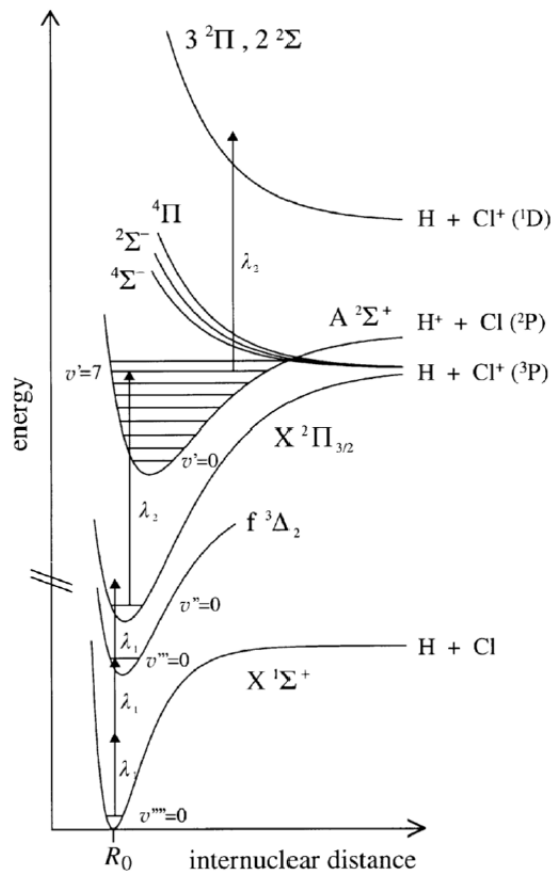


Figure 4.4: Taken from [MW01], HCl^+ possesses a 2+1 REMPI (resonance-enhanced multiphoton ionization) scheme via 243nm light (614990.8 GHz). This REMPI scheme allows for direct loading into an ion trap without reducing the trapping field.

The doublets are the result of mixing with the first excited $A^2\Sigma_{\frac{1}{2}}$ state, which has $\Lambda = 0$ but still can be described using the same quantum numbers as the $X^2\Pi$ states. As a result, the basis states for the relevant molecular states can be described within the parity basis as

$$\begin{aligned} |^2\Pi_{\frac{3}{2}}^{(\pm)}\rangle &= \frac{1}{\sqrt{2}}(|\eta, \Lambda = 1; S, \Sigma = \frac{1}{2}; R; J, \Omega = \frac{3}{2}; I_1 = \frac{3}{2}, I_2 = \frac{1}{2}, F_1, F, m_F\rangle \\ &\pm (-1)^{J-1/2} |\eta, \Lambda = -1; S, \Sigma = -\frac{1}{2}; R; J, \Omega = -\frac{3}{2}; I_1 = \frac{3}{2}, I_2 = \frac{1}{2}, F_1, F, m_F\rangle) \end{aligned} \quad (4.42)$$

$$\begin{aligned} |^2\Pi_{\frac{1}{2}}^{(\pm)}\rangle &= \frac{1}{\sqrt{2}}(|\eta, \Lambda = 1; S, \Sigma = -\frac{1}{2}; R; J, \Omega = \frac{1}{2}; I_1 = \frac{3}{2}, I_2 = \frac{1}{2}, F_1, F, m_F\rangle \\ &\pm (-1)^{J-1/2} |\eta, \Lambda = -1; S, \Sigma = \frac{1}{2}; R; J, \Omega = -\frac{1}{2}; I_1 = \frac{3}{2}, I_2 = \frac{1}{2}, F_1, F, m_F\rangle) \end{aligned} \quad (4.43)$$

$$\begin{aligned} |^2\Sigma_{\frac{1}{2}}^{(\pm)}\rangle &= \frac{1}{\sqrt{2}}(|\eta, \Lambda = 0; S, \Sigma = \frac{1}{2}; R; J, \Omega = \frac{1}{2}; I_1 = \frac{3}{2}, I_2 = \frac{1}{2}, F_1, F, m_F\rangle \\ &\pm (-1)^{J-1/2} |\eta, \Lambda = 0; S, \Sigma = -\frac{1}{2}; R; J, \Omega = -\frac{1}{2}; I_1 = \frac{3}{2}, I_2 = \frac{1}{2}, F_1, F, m_F\rangle) \end{aligned} \quad (4.44)$$

In order to generate an estimate for the hyperfine states of H^{35}Cl^+ , the measured molecular constants in Tab. 4.3 are input into PGOPHER for diagonalizing the matrix. Experimental measurements have not been made to separate the hyperfine constants between the two nucleus, but theory gives estimates of the constants for each nucleus shown in Tab. 4.4. The results generated the energy levels in the absence of a magnetic field. Introduction of this field was achieved by using equations 4.36, 4.37, 4.38, and 4.39, using the parity states given in 4.42. The Zeeman term for the hydrogen nucleus can be simplified to

$$\frac{H_H}{\hbar} = -g_H \mu_N m_H \quad (4.45)$$

where the subscript H denotes the nuclear spin for the hydrogen nuclei. Different possible hyperfine states for driving Λ -doublet transitions are shown in figures 4.5 and 4.6. The standard dipole selection rules allow for transitions between different parity states that are separated by $\Delta F = 0, \pm 1$ and $\Delta m_F = 0, \pm 1$. Transitions between same-parity states are forbidden for an electric dipole interaction, as are transitions between positive and negative hydrogen spin. Like normal hyperfine transitions in

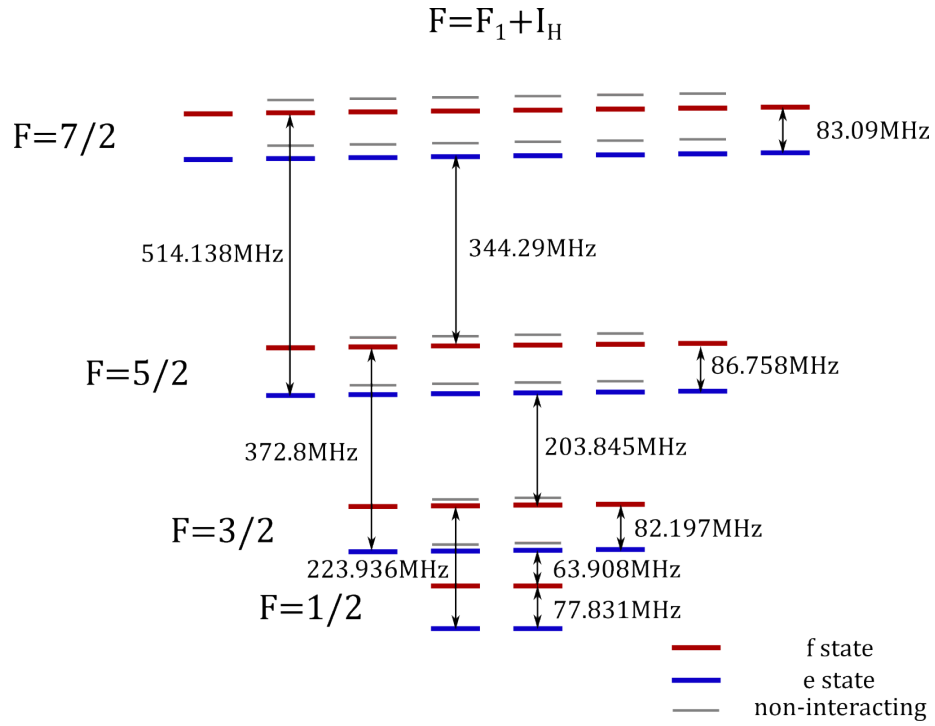


Figure 4.5: Hyperfine states with positive hydrogen contribution for $J = \frac{3}{2}$ ground ro-vibrational state. Estimated electric dipole transitions between different parity states. Gray states indicate the negative hydrogen contribution states, unaddressable with an electric dipole.

atoms, these interactions are allowed with a magnetic dipole. Zeeman splittings are excluded in these figures, but are estimated in order to accurately perform the spectroscopic search (see later chapter on EGGs).

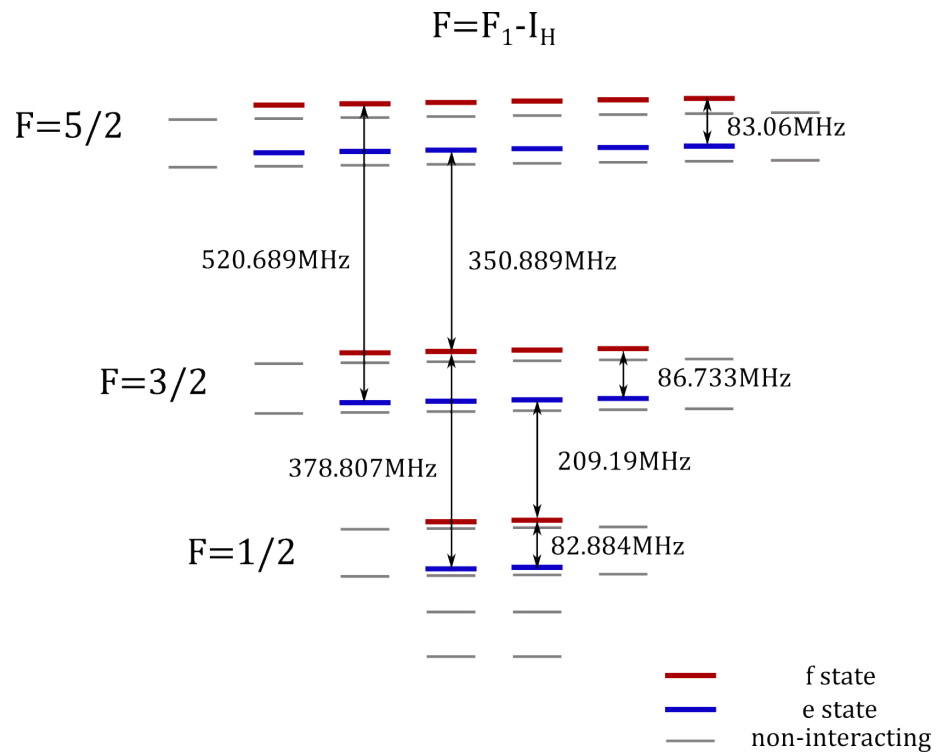


Figure 4.6: Hyperfine states with negative hydrogen contribution for $J = \frac{3}{2}$ ground ro-vibrational state. Estimated electric dipole transitions between different parity states. Gray states indicate the positive hydrogen contribution states, unaddressable with an electric dipole.

Constant	H ³⁵ Cl ⁺ Value (MHz)	Reference
Electronic		
T_e	28626.5 cm ⁻¹	[]
Ro-Vibrational		
ω_e	2673.69 cm ⁻¹	Webbook
B_0	293444.013(48)	[HP12]
D_0	16.39759(157)	[HP12]
$10^4 H_0$	4.468(126)	[HP12]
Spin-Orbit		
A	-19279138(38)	[HP12]
A_D	54.34(99)	[HP12]
Spin-Rotation		
γ	-9163(34)	[HP12]
Λ -Doubling		
p	18280.17(102)	[HP12]
p_D	-1.355(25)	[HP12]
q	-332.198(84)	[HP12]
$10^3 q_D$	41.05(478)	[HP12]

Table 4.3: Tabulated constants for H³⁵Cl⁺ used for estimating the internal molecular structure in PGOPHER, excluding the hyperfine values.

Constant	H ³⁵ Cl ⁺ Value (MHz)	Reference
Hyperfine (Experimental)		
h	357.04(60)	[HP12]
a	409.16(88)	[HP12]
b	169.6(34)	[HP12]
b_F	78.34(254)	[HP12]
c	-273.9(33)	[HP12]
d	523.12(281)	[HP12]
$10^3 d_D$	60.7(414)	[HP12]
eQq_0	-8.03(210)	[HP12]
eQq_2	-364.9(188)	[HP12]
Hyperfine (Theory)		
a_H	-15(1)	[BG06]
a_{Cl}	408(3)	[BG06]
$b_{F,H}$	-64(2)	[BG06]
$b_{F,Cl}$	85(15)	[BG06]
$\frac{c_H}{3}$	14(1)	[BG06]
$\frac{c_{Cl}}{3}$	-81(2)	[BG06]
d_H	-29(1)	[BG06]
d_{Cl}	489(1)	[BG06]

Table 4.4: Comparison of the experimental hyperfine constants for H³⁵Cl⁺ excluding the different nuclear contributions to theoretical constants for each nucleus.

CHAPTER 5

Experimental Setup

5.1 Cryogenics

The interactions of molecules with black body radiation served as the primary motivation for the design of a cryogenic ion trap experiment. The previous molecule experiment, MOTION [Pur19, Mil20], is a room temperature experiment, which posed a limitation towards future endeavors with molecular ions, such as implementing dipole-phonon quantum logic [CH20, MH00, QRY24]. Several considerations were taken into account when it came to the choice of cryogenic system [Bra16, Eki06]:

- High Cooling capacity
- Low Vibration
- Minimal Complications

The first item is the easiest to consider. The cryocoolers that are able to cool to the lowest temperatures are typically the complex and costly dilution fridges used in superconductor and semiconductor quantum experiments. These systems would cost all of the start up funding for this experiment, as well as impose strict limitations on optical access. For our experiments we want to cool as low as possible, $\sim 4\text{K}$, to limit black body radiation from the surrounding system and to freeze out hydrogen gas. A more standard coldhead, like a pulse tube or a Gifford-McMahon (GM) coldhead can accomplish these temperatures easily with typically $\sim \text{W}$ of cooling capacity.

The second point on this list is very important when it comes to ion systems because narrow waist lasers need to reliably address atoms without suffering from large intensity fluctuations. In a system with large vibrations, the ion trap (and as a result the ions) would vibrate through the laser, which is fixed to the table. A typical

pulse tube or GM coldhead would transfer these large oscillations to an ion trap if it were attached directly to the second stage of these devices. Unlike GM cryocoolers, pulse tubes do not have the cold stages attached to a motor, which offers significantly weaker vibrations but typically comes at a higher cost. To minimize vibrations, these coldheads are paired with either copper braids or with helium as an exchange gas. Both options will often reduce the cooling capacity significantly but still provide enough cooling capacity for a small enough load.

The last consideration may initially sound silly, but ends up playing a significant role. Different commercial systems on the market have different Achilles' heels. For example, systems that use helium exchange gas within a rubber bellows to transfer cooling from the stage to the coldplate will often have water leak in and cause damage inside the system. In our system, the Stinger, helium exchange gas is also used but instead is flowed through tubing to transfer cooling while minimizing vibrations. The challenge faced during my time has been the frequent clogging of the gas through these lines that forces us to perform a complete warmup over several days.

5.1.1 Stinger

The Stinger system is a closed-cycle cryogenic system designed by Coldedge Technologies, which uses high pressure helium exchange gas to cool the coldplate with minimal transfer of vibrations from the coldhead. A typical dissertation may leave it there, but the amount of time and effort put into this system warrants a deep dive into the system and how it operates. Nearly every component of this system has been disassembled by my hands, from the compressors that serve as the heart of the system to the coldhead itself, to the guts inside both the blue housing and within the the main chamber that lie underneath the coldplate.

To begin with, the Stinger system is made up of an SHI Model SRDE-418 GM cryocooler powered by an F-70 compressor. This coldhead sits in what we refer to as the blue housing, a vacuum system that encapsulates the stages of the coldhead and the tens of feet of copper tubing through which the helium gas is cooled. These lines end in the Stinger insert line, which is a 5' long bellows that has the supply and return lines going to the main vacuum chamber, where a custom insertion flange

prevents Helium from exiting to atmosphere and air from leaking into the system. At the tip of the insert line is a couple-foot-long capillary tube. The unique feature of the Stinger system is an additional step of Joule-Thompson cooling by the gas as it exits the tube and gets to the coldplate. After several windings of tubing on the 4K stage, there is a partial winding on the radiation shield, cooling it down to 20K. The gas is then pulled back through the return side of the Stinger insert line, no longer requiring a capillary. As this gas returns to the blue housing, it pre-cools the gas that is entering the blue housing and going around the first stage of the GM coldhead. In the beginning, the compressor that circulates the helium exchange gas was an HC-4E1 but later was swapped to an F-20 with a custom venturi device; regardless of which compressor is used, this component is called the recirculator. This transition was made since we were experiencing a significant rate of clogging, despite additional filters and adsorbers put in line. The venturi device allows the F-20 to operate above atmosphere, so air shouldn't enter the system. With the new compressor, the minimum temperature has been $\sim 6\text{K}$ although we believe that an additional throttling component could restore our prior performance since it reaches $\sim 4\text{K}$ during a cooldown with the coldtrap install; after removing the coldtrap the temperature increases.

The cooling capacity of the GM Cryocooler is quoted to be 1.8W at 4.2K, and the Stinger system reduces it to 0.9W at 4K. Our typical experiment operates between 4.5K and 6K at the coldplate. The pressure and flow of helium are the dominant parameter that determines our performance. In the beginning we had a thermal short between the supply input and the radiation shielding, which resulted in a much higher temperature, but was fixed after breaking vacuum and fixing this short.

5.1.1.1 Cooldown Performance

Similar to electronic systems, cryogenics follows similar calculations to determine how cold a system can get with a given heatload. The cooling capacity quoted by vendors will inevitably be reduced as more components are introduced between the trap and the coldplate, limited by thermal resistance and joint resistance. Thermal resistance is given by $R(T) = \frac{l}{A\lambda(T)}$ where l is the length of the component, A is the cross-

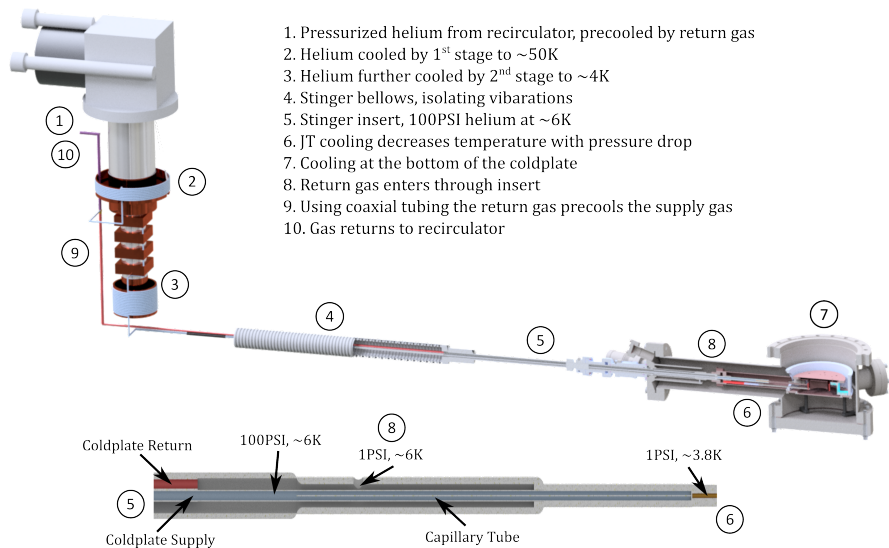


Figure 5.1: Illustration of Stinger operation. Not shown in this figure is the Stinger Manifold which regulates the flow of helium from the recirculator to the coldhead. Typical operation used 120psi of helium into the coldhead. 1) Helium enters through the inner line of coaxial tubing, the outer line filled with the return gas, precooling the supply before it gets to 2) and 3). The key feature of the Stinger is the bellows that mitigate vibrations 4). Helium exiting the Stinger insert at 5) undergoes Joules-Thomson cooling 6) by flowing through a narrow capillary tube. Helium flows over the 4K coldplate 7) and the 20K radiation shield before returning through the insert 8). As the gas returns to the recirculator 10) it first precools the entering gas.

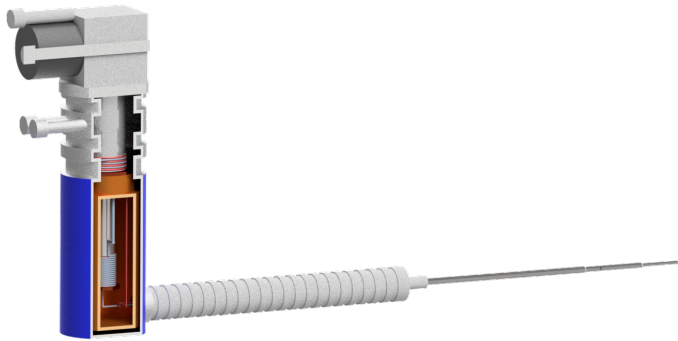


Figure 5.2: Illustration of the Stinger and its recognizable blue housing. The blue housing is the outer vacuum chamber that maintains a $10^{-3} - 10^{-5}$ mbar vacuum at room temperature. Material choices within the blue housing have relatively high outgassing and the connections on the housing are KF, so it is unable to achieve UHV. Within this chamber, there is a single layer of radiation shielding, cooling using the first stage of the coldhead. A temperature diode is attached to the supply line just before the bellows to measure the system's performance. This diode needs flow to effectively measure the temperature of the second stage; with a clog, this will eventually settle around 50K, due to the conduction through the supply line.

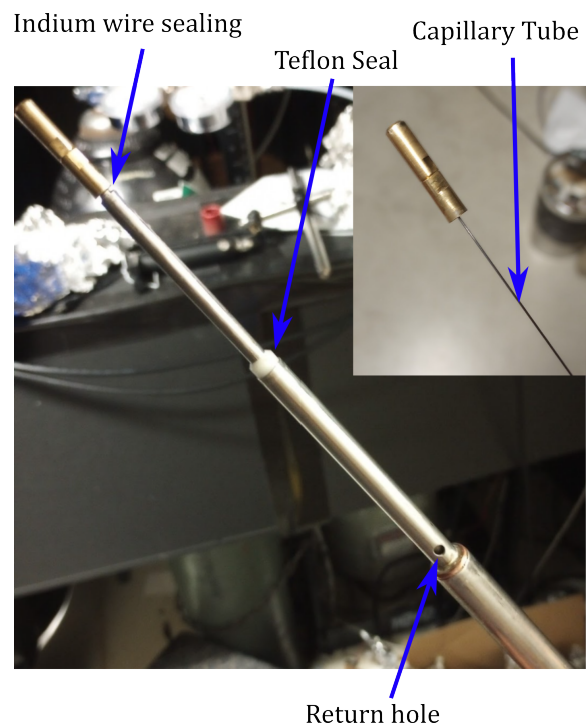


Figure 5.3: In order to achieve Joule-Thomson cooling, a 100 micron ID capillary tube is used. Different diameter tubes are available from Coldedge, with larger diameters offering greater cooling power at higher temperature. The capillary tube is attached to the Stinger insert using an indium seal. A Teflon seal ensures gas in the supply side is forced around the coldplate and does not return directly through the return hole to the recirculator.

sectional area of it, and $\lambda(T)$ is the temperature dependent thermal conductivity of the piece [Eki06]. Its inverse, thermal conductance, is often also cited for different materials. The heat transfer via conduction is given by

$$\dot{q}_{cond} = \frac{\Delta T}{R(T)} = \lambda(T)A \frac{\Delta T}{l}. \quad (5.1)$$

In order to achieve the lowest thermal resistance, cryogenic systems will implement Oxygen-Free High Conductivity (OFHC) copper, and use as few pieces as possible. The latter choice comes from the desire to limit joint resistance which is an increase in thermal resistance between two different surfaces. Joint resistance can be reduced by tightening the two surfaces with greater force, or by using a thin film of highly malleable metal, such as gold or indium. Imagine zooming into the two surfaces. If the surfaces are very rough there will be few points of contact where conductive cooling can occur; if those gaps are filled in, then the conductance will increase from greater contact area. Common thermal conductivity values and joint resistances can be found in Chapter 2 of [Eki06].

In addition to thermal resistances, consideration of different heating mechanisms must be made. The most obvious source of heating comes from power dissipation from electrical devices, namely the ion trap. Radiative heating between two surfaces, maybe a less obvious source, plays a dramatic role as well, following the Stefan-Boltzmann equation

$$\dot{q}_{rad} = \sigma EA(T_2^4 - T_1^4) \quad (5.2)$$

where σ is the Stefan-Boltzmann constant, E is a factor dependent on the emissivity of the two surfaces, A is the area of the surface, and T_1 and T_2 are the initial and final temperatures of the surface. The emissivity of a surface ε spans 0 to 1, where a perfect black body absorber will have an $\varepsilon \rightarrow 1$ and a perfect mirror will have $\varepsilon \rightarrow 0$. Values for parallel plates and coaxial systems can be found in Chapter 2 of [Eki06].

As systems become colder, they will experience thermal contraction. Different materials have different rates of thermal contraction, and in order to maintain the pressures needed to mitigate joint resistance, it is important to make sure that screws, spacers, and washers are chosen to avoid problems during thermal cycling. In cryogenics, we follow the ABC's (Chapter 3 of [Eki06]) which is an easy nomenclature

for Aluminum, Brass, Copper, Stainless steel; this represents common metals organized in highest to lowest rate of thermal contraction. Titanium is commonly used as a spacer due to its low contraction rate. In general, ceramics and crystals will possess low contraction rates, whereas plastics will contract by large amounts at cryogenic temperatures, with several exceptions, one being Polyimide (Kapton). To compensate for higher contraction rates, it is typical to include a spacer with low contraction rates, or use Belleville washers. A Belleville washer is shaped to spring out to maintain contact forces to mitigate the effects of thermal contraction.

As part of designing the cryogenic system, I also made estimates of how long a cooldown should take considering the Stinger cooling capacity and all the masses that need to be cooled. A rough calculated mass of each stage was generated by measuring the volumes of the different metal parts in AutoCAD. Assuming uniform density, the 4K stage and ion trap represented one mass load ($\sim 857g$) and the radiation shield represented the other $\sim 3010g$. The cooling capacity curve for the Stinger demonstrated the cooling power over temperature, and was split into six temperature regimes. Cooling power is the rate of heat transfer given by

$$\dot{q}(T) = \frac{\Delta q(T)}{\Delta t} \quad (5.3)$$

where $\Delta q(T)$ is the change in heat over a given temperature range, and Δt is the time it took to cover that temperature range. Within a given temperature regime, the specific heat capacity for each mass was calculated using values from the NIST database on cryogenic material properties. The time it takes to cool from the high end of a temperature regime to the low end is given by

$$\Delta t = m \frac{k(T) \Delta T}{\dot{q}(T)}, \quad (5.4)$$

where the total cooldown time is the sum of each region. In Fig. 5.4, the estimated cooldown times (shown in orange and red) are compared to an experimental run (shown in blue and purple) for the 4K and 40K shields, respectively. Since the Stinger's performance varies based off of the flow and pressure of the helium gas, there can be variations in performance compared to the test sheet. Additionally, a rough estimate of that total mass load was made; including more than just the shield and ion trap on the 4K stage would result in a slower estimate of the cooldown time, which may better match the experimental cooldown time.

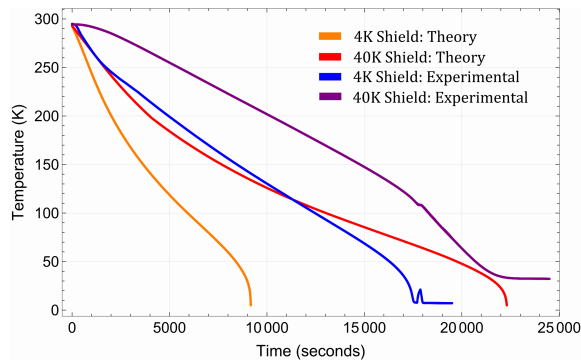


Figure 5.4: Prediction of the time it takes to cool from room temperature to 4K compared to an actual cooldown. The estimated cooldown times (shown in orange and red) are compared to an experimental run (shown in blue and purple) for the 4K and 40K shields, respectively.

5.1.1.2 Vibration Measurement

The Stinger system is quoted to limit vibrations to 10nm at the coldplate. One of the first tests done with our system was to measure the vibrations where our ion trap would sit. In place of a trap, an aluminum block was machined in an L shape, with mirrors epoxied using Stycast to the piece, such that one mirror was facing up and the other facing away from the Stinger insert. To measure the vibrations a simple interferometer was assembled. Vertical vibrations were measured using the 397nm laser, and horizontal vibrations were measured using the 729nm laser. Light out of the fiber was sent through a 50/50 beam splitter, to create two arms of the interferometer, which were recombined onto a photodiode. One arm would have the in vacuum mirror at its end and the other would be held constant. To ensure the second arm wouldn't move, lead bricks were used to weigh down the mezzanine breadboards. The horizontal interferometer was able to take better measurements because of the stability of the system: narrow linewidth laser with higher wavelength light, in addition to optics on shorter posts traveling shorter paths. Vibrations would cause the phase of one arm to change according to $\varphi = 2\pi \frac{x}{\lambda}$. Measuring the voltage from the photodiode, the interference of the two beam paths follows

$$V = V_1 + V_2 + 2\sqrt{V_1 V_2} \cos(\varphi(t)) \quad (5.5)$$

with the subscript indicating a given beam path. The phase can be decomposed into

$$\varphi(t) = a_0 + \sum_{n=1} \sin(\omega_n t) \quad (5.6)$$

where ω_n is some frequency that introduces a phase change. We can estimate the maximum vibration amplitude by assuming the vibration is caused by a single frequency generating a phase of

$$\varphi(t) = \frac{\varphi_{max} - \varphi_{min}}{2} \sin(\omega_1 t) + \frac{\varphi_{max} + \varphi_{min}}{2} \quad (5.7)$$

which will result in a maximum displacement of

$$x_{max} = \frac{\lambda}{2\pi} \frac{\varphi_{max} - \varphi_{min}}{2}, \quad (5.8)$$

choosing φ_{min} and φ_{max} to be the phases associated with the smallest and largest measured voltages when both arms are hitting the photodetector. In the ideal case, we get that $\varphi_{max} - \varphi_{min} = \pi$ and the maximum change in displacement measurable is $\frac{\lambda}{4}$. Variations in voltage from the photodetector were converted to variations in displacement, and a Fourier transform of the results indicated the spectrum of displacements. Integrating the spectrum from the highest frequency to the lowest indicates how the RMS displacements increase. Results from these measurements are shown in Fig. 5.5 with different conditions. The best performance was given when an additional support piece was added to fasten the Stinger insert to the optical table, and when bags of sand were placed at the Stinger bellows near the blue housing. Typical operation does not include the sand.

5.1.2 Limiting Black Body Radiation

Just by attaching an ion trap to a coldplate does not mean that the effect of black body radiation has been reduced to the extent needed. In order to reduce it properly, the ion trap needs to be well shielded. For this reason, cryogenic ion traps are put within several layers of thermal shielding. The radiation shield sits between the 4K stage and the room temperature vacuum chamber. In our experiment this stage is typically 20K. The 4K stage also offers additional shielding, surrounding the ion trap similarly to the radiation shielding. Both stages have some through holes for wiring and for vacuum conductance, but these holes were specifically designed to not

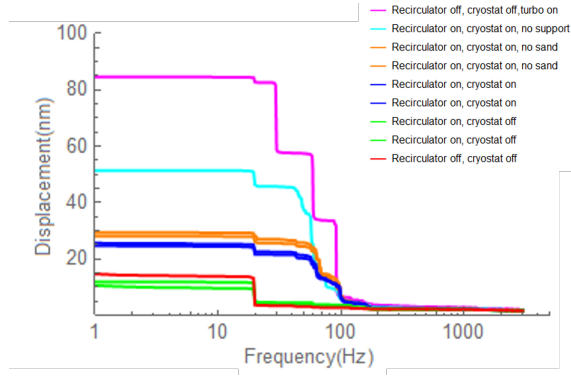


Figure 5.5: Vibrations were measured using 729nm cavity-locked light, sent in horizontally i.e. with the laser beam orientation. Different conditions were tested to mitigate the vibrations. Best performance involved adding an additional clamp to the Stinger insert and sandbags where the bellows attach to the blue housing. During typical performance, the sandbags have been removed; they were on loan and never replaced.

have direct line of sight from 300K to the ions. The sole exception is the hole for loading, since we load from an oven/from leaking in gas, not through ablation. For optical access, we have fused-silica windows from Laseroptiks, some uncoated and some AR-coated for 397nm. The horizontal windows are for the lasers to address the ions, and are 3 mm thick. The vertical window is for optical access and is 1 mm thick. Author comment: the thickness of the vertical window was chosen to limit optical aberrations, but in hindsight I would have tried using a thicker window due to Brewster's law. Measurements of the transmittance of these windows at 1.4THz were made in two other labs, the Musumeci Lab in Physics (our neighbors) and the Jarrahi Lab in Engineering, since both experiments could produce THz light and were gracious enough to let me stick my spare optics in their beamlines.

In both laboratories, I tested the AR-coated 1 mm thick window, the coated and uncoated 3mm windows, and a steel mesh that we have beneath the 4K imaging window to prevent charging of the window when ions are ejected from our trap. In Musumeci's lab, they operate a 0.6-0.8THz light source. Every component tested had about 80% transmittance; testing was done simply by comparing the power of the light with and without the components in line. In the Jarrahi lab, they operate a beam line that emits 0.15-2THz. Like before, we simply detected power without

Component	Transmittance
1 mm Window	43%
1 mm Window with mesh	28%
1 mm Window with double the mesh	20%
3 mm window	18%

Table 5.1: 1.4THz Transmittance

the components and then with them. The results from this test are shown in Table 5.1.

To determine how this may limit our molecular lifetime, the numerical aperture for each window to 300K was taken into consideration to estimate how frequently a transition to the higher rotational stage would occur. Based off of simulations performed by Hao, we estimate that HCl^+ , in a room temperature bath, will undergo a rotational transition will once every 6 seconds. Estimates of what we'd expect the effects of room temperature on our lifetime are shown in Table 5.2. The dominant limitation to our lifetime comes from the imaging window; it is the thinnest window and has a large numerical aperture. Further improvements to our system would come from replacing this with a thicker window and/or a lower transmittance mesh. Additional limitations to our lifetime may come from the BBR from a trap that is not at 4.5K. A temperature diode attached in a manner to replicate a trap blade suggests that the trap is at 20K, but because it is hard to accurately state what our trap's temperature is, the calculation is only shown for a 4.5K trap. Additionally, our 4K stage does not have any radiation absorbing material (RAM) which would ensure that any light entering would be absorbed and not reflected off the walls of the 4K stage.

5.2 Vacuum Chamber

The base of our vacuum chamber was fabricated as part of the Stinger system. It has an 8" CF flange on the top side, onto which we attach our 8" CF octagon. There are four 2.75" CF flanges on the sides, onto which we attach the following: a titanium

Component	NA	Emissivity	Lifetime
Vertical (1mm Window and mesh)	0.368	1	1707 s
All 7 Horizontal 3mm Windows	0.054	1	6156 s
Oven Without Window Holder	0.12	0.2	3142 s
Total			938 s
Oven With Window Holder	0.054	0.2	6981 s
Total With Window Holder			1163 s

Table 5.2: Estimated Lifetime from Black Body Radiation

sublimation pump (Ti:Sub), a Nextorr Z-200 NEG/Ion pump combination system, a residual gas analyzer (SRS RGA 200), and a tee that has the angle valve and an Agilent FRG-700 magnetron gauge [OH03, Wel01]. This base chamber also has a welded 4.5" CF nipple, the end of which has the Stinger insert adapter. The chamber is mounted to the table using two custom v-slot mounts and two post-mounted c-clamps. All but one of these supports clamp to a 2.75" nipple, the exception clamping onto the 4.5" nipple.

Connected to the top flange is a Kimball 8.0" extended spherical octagon chamber, which has eight 2.75" flanges and sixteen 1.33" flanges. Seven of the 2.75" flanges are used for viewports: an EUV fused silica with AR coating for 225-450nm viewport, a DUV fused silica viewport, two DUV sapphire viewports, and three Kodial viewports. The final 2.75" flange has the loading system attached to it. Most of the 1.33" flanges are unused, and those that are used have electronic feedthrough connections for the trapping voltages, for temperature diodes, and for cryogenic heaters. There are three inputs for the trapping voltages: one 4-pin feedthrough of DC (endcap and shimming), one 4-pin feedthrough for the EGGs voltages (two of the pins are unconnected), and one 2-pin feedthrough for the trapping RF voltage (one of the pins is unconnected). Three 4-pin feedthroughs were installed for silicon temperature diodes (DT-670A1): 2 pins for supplying a current and 2 pins for measuring the voltage. Readout of the diodes is done with a Lakeshore Model 336 temperature controller. This controller can also power two 50Ω heater cartridges; these heaters and most of the diodes are disconnected during standard operation as they introduce noise onto

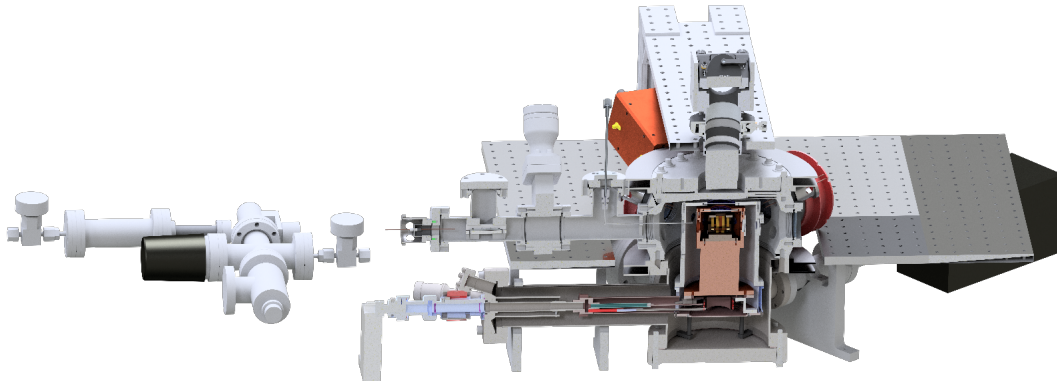


Figure 5.6: AutoCAD assembly of our entire vacuum system. Description of each component is given in the text.

the ion trap. Unfortunately, these heaters are prone to damage and several have failed during my work on the experiment. Running these at high power seems to lead to shorter lifetimes, and can also increase the amount of outgassing from them; we limit them to 25% max operation. The final flange, the other 8.0" flange, has a custom DUV fused silica viewport, with 2.69" view diameter, AR coating on both sides for 397nm, and $R < 0.25\%$ per surface at 397nm, purchased from MPF.

5.2.1 Additional Pumping

Our dominant pumping component is our cryogenic system [OH03, Wel01]. While this system was designed to handle a low temperature bake (156.6°C for the indium wires in our vacuum for cryogenic contact), we have only baked once in the beginning, prior to installing all the necessary components for molecular loading. We start by pumping our system with a turbo pump, reaching as low as 10^{-8} mbar, after activating the NEG and running the Ti:Sub. The performance of the Ti:Sub improves as the pressure decreases, but we've noticed that while running it does reduce our overall vacuum pressure, it outgasses Hydrogen. This means we don't run it when cold; it is run only when we do a full warmup and open the angle valve to our turbo. The Ti:Sub pump is installed within a 4.5" nipple since a larger surface area that the

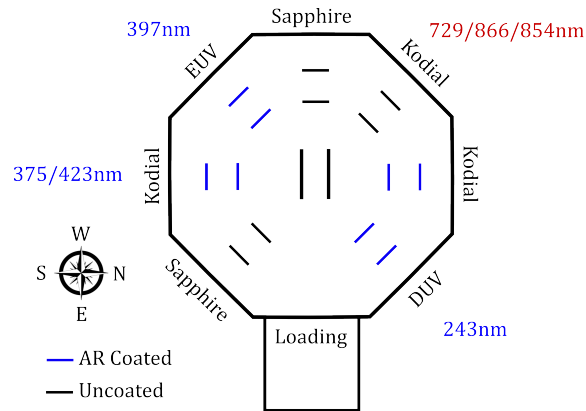


Figure 5.7: Illustration of the orientation of the cryogenic windows and the vacuum viewports. The blue coloring on the windows indicate that they are AR coated for 397nm light, otherwise they are uncoated. The two lines in the center of the chamber are there to indicate the axial orientation of the ion trap.

titanium can adhere to results in improved performance. A Nextorr Z-200 NEG/ion pump is installed to provide additional pumping, although the design of the combined system is not ideal, and the pumping of the ion pump component is weaker than in comparable chambers at UCLA. In this design the NEG sits in front of the ion pump, which means it does most of the pumping. The other UHV chambers in the AMO groups at UCLA use larger ion pumps to get higher pumping rates, however, this model Nextorr pump is small enough that it can fit on our vacuum system, whereas larger models would be too large to fit between the optical breadboards and the optical table without milling of the optical breadboards or replacement of the mounting supports, the latter of which could threaten beam stability due to shallow mounting holes.

5.2.2 Ion Loading

This experiment is designed to co-trap multiple species of ions: laser coolable Ca^+ ions and molecular ions, mainly HCl^+ . Calcium has a well-established photoionization scheme from a heated oven. A 423nm laser resonant with the S-P transition in neutral Ca is followed by light from a 377nm photon that takes the excited photon to the continuum, stripping it from the atom. Light with a shorter wavelength than 389.9nm is able to accomplish this second step [DS04]. Neutral calcium atoms are

emitted from an oven when a large enough current passes through it. The oven is a tube filled with calcium metal, packaged with an indium seal and shipped in argon gas. The lifetime in an unsealed container is quoted to be only 6 months.

The reactivity of calcium led to the choice of a gate valve between the calcium oven and the rest of the vacuum chamber. In the initial stages of this experiment, we would break vacuum several times a year, and it made more sense to close the gate valve to improve the longevity of our source. The trade off from adding this valve is the oven sits further from the trap than in most experiments, and loading Ca^+ ions takes on the order of 5-10 minutes at 3.25A. A larger current can be used for a faster rate of loading, but frequently loads more than one ion, requiring us to ramp the trap voltages to try to remove excess ions.

Our calcium oven sits in a homemade structure, entitled the tie-fighter holder, based off of its shape Fig. 5.9. The oven sits on a Macor insulator piece(s) that is between two steel pieces. The outer diameter for the steel pieces allows the whole structure to sit snugly within a 2.75" CF tee. A tee was chosen over a nipple, to allow for greater access during assembly. The Macor piece(s) broke during fabrication, but since the entire system worked well, it was never replaced.

The oven is a calcium vapor source from Alfa Vakuo. The source is quoted to be 99.5% pure ^{40}Ca , which is packaged within a stainless steel tube. During manufacturing, this tube is filled under argon gas before being sealed with indium. Once installed, running a large enough current breaks the indium seal, indicated by a spike in pressure. When using the RGA on our chamber, this shows a spike at 40amu from the argon gas being released. The oven contains 100mg of calcium to ensure the source does not need frequent replacement; a single oven was replaced before the gate valve was installed, with its replacement lasting through the end of this PhD.

The other component for loading is a gas distribution system for introducing HCl gas to our chamber. Due to its reactivity, several precautions were made to load this gas. The first is that it must be held in minute quantities within an inert gas. Initially the choice of balance gas was N_2 because it was an affordable option. While this gas worked when loading HCl^+ in the MOTION experiment, this experiment

has a much smaller ion electrode distance. As a result, when the photoionization light (243nm) was sent through, it would often clip on the electrodes. The UV light can then strip electrons off of the electrodes since the work function is less than the energy of the laser. These electrons, now introduced to a large voltage, have enough energy to ionize the N_2 balance gas. As a result, the next balance gas was chosen to be something that would be unstable within our trap if it was loaded. This means that gases heavier than 40amu were preferable, and specifically those that would not dissociate from the UV light or become doubly ionized. There were two choices we could come up with: Krypton and Xenon. Both of the gases are difficult to come by and were primarily exported by either Russia or Ukraine, and at this time a certain conflict had just started between the two nations. We were able to luck out and get a bottle of Krypton with 50ppm of HCl from Airgas specialty gas. This bottle cost us a pretty penny (\$10k): hence why we refer to it as our bloodgas.

To introduce this gas into our experiment, we implemented a leak system, limiting how much gas enters for longer vacuum lifetimes. The leak system is centered around a 2.75" CF cross. One arm of the cross is connected to a ball valve that then goes via a 1/4" VCR bellows to a VCR/CF adapter on the main chamber. On the CF side of this adapter is a 1/4" swagelok connector. A 1/8" copper tube is adapted to the swagelok and is bent to direct the flow of gas towards the ion trap. A steel disk was machined to sit within this tee to hold the copper tube so as to not impede the flow of calcium from the oven. Also attached to the cross is an angle valve to a turbo, constantly pumping the cross when not in use, a magnetron gauge to monitor the pressure in the cross, and a leak valve to a gas reservoir. The reservoir is a bakeable volume that holds the gas prior to leaking it in, made up mainly of a 2.75" nipple. Between it and the large bottle of gas is a VCR ball valve and bellows. The reservoir is a desirable step to limit contaminant gas that outgasses from the walls of the bellows between the large bottle and leak valve.

5.3 Ion Trap

Ion traps are designed with specific goals in mind. Previously experiments at UCLA used large rod traps which facilitated loading large numbers of ions, but were in-

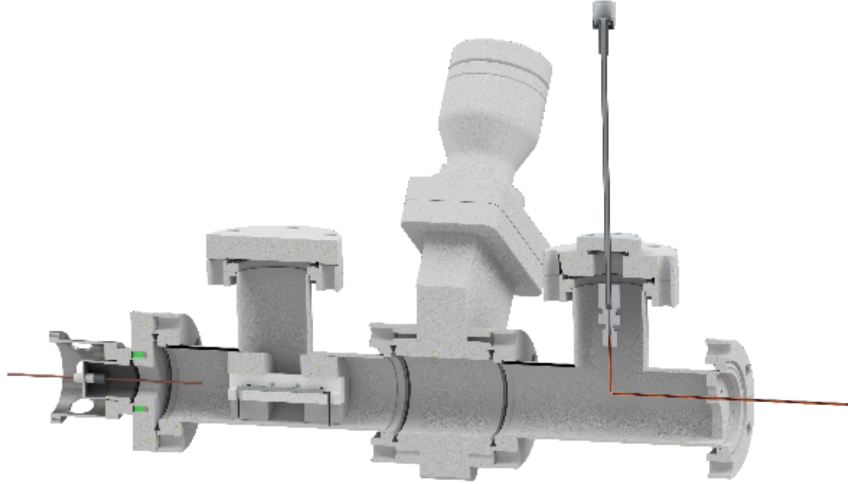


Figure 5.8: Attached to the octagon are two 2.75" tees in series. The tee closer to the trap has tubing that directs the gas for loading HCl^+ ; the other tee holds the calcium vapor source. Between the tees is a gate valve to prevent exposing the calcium source to atmosphere when the chamber is opened for upgrades.

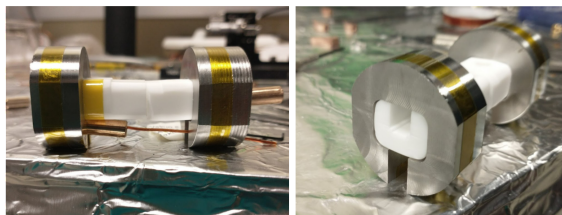


Figure 5.9: Images of the calcium oven in its holder. During the machining of the Macor holder, it snapped in two. Kapton tape was added to make sure the holder would not slip out of the steel block, which in turn had tape to keep it from slipping in the nipple. The notch in the steel block lets the oven electrode through without electrical contact.

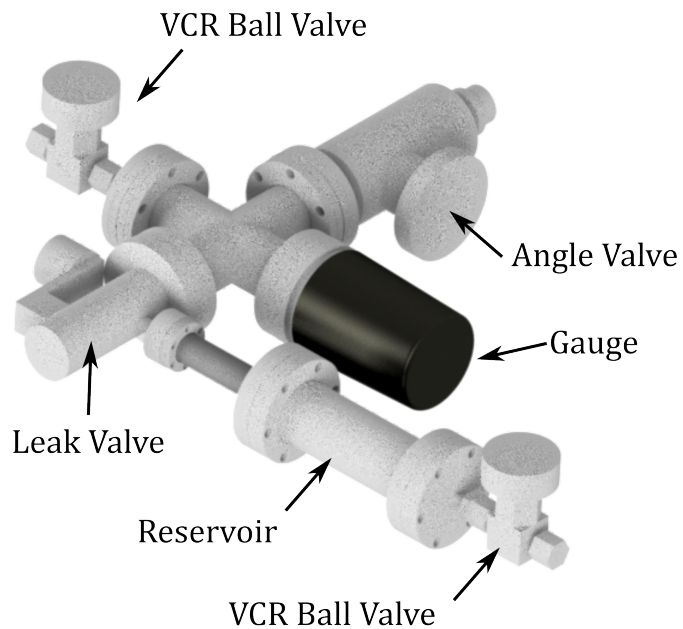


Figure 5.10: A separate vacuum system was assembled to leak HCl into the main chamber, which we refer to as the HCl cross. On the cross is a metal angle valve, a magnetron gauge, a VCR ball valve to the main chamber, and a leak valve to a reservoir of gas. On the other side of this reservoir is another ball valve that is connected to bellows that lead to the tank of gas. We baked this system using heater tape to 120°C to improve the vacuum prior to introducing gas into the reservoir. We have loaded several psi of gas into the reservoir. A turbo pump pumps the cross when not loading to maintain a low pressure (ball valve and leak valve closed).

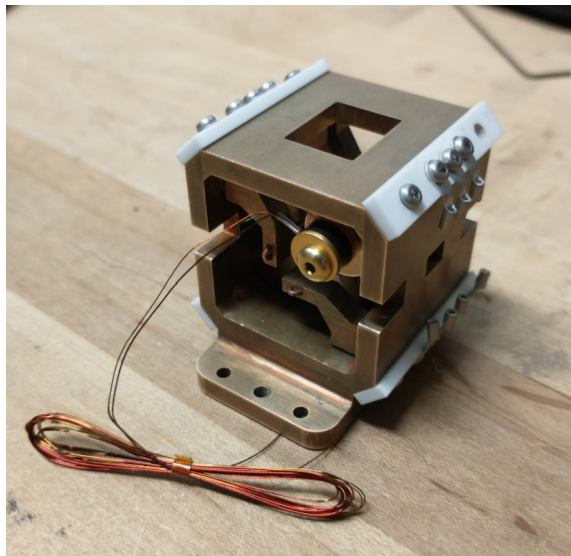


Figure 5.11: Test assembly of our ion trap prior to vacuum cleaning.

capable of implementing motional control of states for sideband cooling and state readout. This experiment, unlike those prior to it, requires these capabilities since it was conceived to be a trap for implementing the fundamentals of molecular quantum logic. Around the same time, a new barium ion experiment using a blade trap from the Monroe group was also being designed to provide high secular frequencies for two-qubit gates and the omg architecture [JWN17]. Unlike this trap, which would have been expensive to fabricate (the barium trap was an unused backup gifted to the experiment), our trap was designed to be cheap, easy to assemble, and implemented in a 4K cryogenic environment.

5.3.1 Trap Design

Inspiration for the design of this trap came from two sources. First, since there is no need to reinvent the wheel, we drew inspiration from all of the calcium experiments that came before us such as [Roo00, Gug17, Bra16, SCH20] and many more. While the theory outlined in section 2.1.1 suggests it is simple to pick geometries, practically it makes more sense to stay close to something that has already undergone rigorous testing. For the initial testing of trap designs, examples of traps from the Blatt group [Gug17, SCH20] were recreated in CAD and tested using SIMION to ensure the simulated results agreed with the published results. The second source of inspiration came from a rough idea that our staff scientist Dr. Christian Schneider

had; an idea about machining the trap blades when they were already in place to optimize the precision of the geometry. If a blade design was chosen for not only the RF but also the endcaps, then a wire EDM could create high precision on every electrode. Unfortunately, at the time of this trap's design and fabrication, the campus machine shop was understaffed and incapable of operating the machine envisioned for this project. The all-blade idea stuck around since simulations showed similar performance to the Blatt traps.

Typically, the most expensive component of a 3-D trap is the fabrication of the insulating frame. Due to the fact that our experiment operates at cryogenic temperatures, there were greater restrictions about what insulating materials could be chosen. Plastics would suffer from large changes from thermal contraction/expansion. Ceramics aren't as susceptible to these changes but are more difficult to machine and would have limited thermal conductance. Crystals, specifically sapphire, offer both low thermal contraction/expansion, and have high thermal conductance at low temperature but are significantly more expensive to grow and machine. The solution was to forsake an insulating frame, and instead make one from the same material as the electrodes ($\sim \$800$). Insulation is now achieved using simple pieces of alumina ($\sim \$100/\text{each}$) and sapphire disks for Polaris mounts ($\sim \$8$ for a two pack). If designed again, the alumina pieces would be better if machined instead from sapphire to make them less brittle, and if possible made as shoulder washers to improve axial assembly.

Next was the choice of materials for the electrodes and frame. There were three main considerations: machinability, thermal contraction, and thermal conduction. Typically, ion traps are made from steel or titanium; these materials are easy to machine. Both metals also have low thermal contraction. However, both are poor conductors and would have poor thermal conduction. Additionally, they are also relatively poor electrical conductors, which is why many ion traps made from them get these electrodes gold plated. Aluminum is easy to machine and has good thermal conduction, but it has a high level of thermal contraction and its oxide is an insulator, which would lead to large patch potentials. Copper has great thermal properties but is notoriously difficult to machine, which would prevent us from achieving the desired

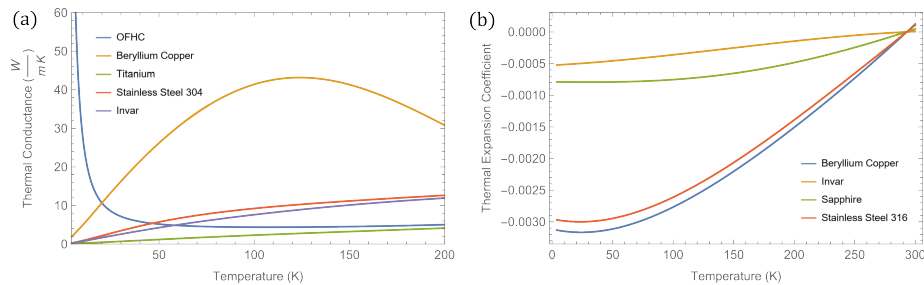


Figure 5.12: (a) Thermal conductance of various materials considered for the trap electrodes. (b) Thermal expansion/contraction of various materials for trap parts. Beryllium copper offers higher conductivity compared to most low expansion metals, with similar levels to stainless steel. Curves are taken from the NIST database of cryogenic materials.

level of precision. Instead, an alloy of copper was chosen. Beryllium copper offers the best options for the considerations outlined. Unlike copper, it can be easily machined to high precision (note that the dust from it can be dangerous to the lungs, so only experts should machine it). It has a similar level of thermal expansion as steel, and while it is not as good of a conductor as copper, it is significantly better than many alternatives. Lastly, while it does form a thin insulating oxide layer, this happens much slower than aluminum. Fig. 5.12 shows the thermal conductance and expansion of some of the considered materials (data taken from the NIST database).

5.3.2 Trap Parameters

The dimension of our trap drew their influence most heavily from [Gug17], which was designed for co-trapping Ca^+ and Al^+ . Since our experiment is designed to co-trap Ca^+ and a lighter mass species, HCl^+ this design seemed the most appropriate. However, the similarities end with ion electrode distances. Our trap has a frame made from a single piece of beryllium copper with an interior that is 27mm by 27mm with the corners cut to enable the blades to be mounted with a single screw each. Imaging is done vertically, so a 12mm by 15mm cut was made to enable optical access from above. This was chosen to allow for a numerical aperture of 0.368, which is larger than the NA of the imaging lens so that the trap did not limit the collection of light. To give optical access for the lasers being sent in horizontally, different

cutouts were made. For the purely radial light, these are 4mm by 6mm; for light with an axial component these are 5mm by 7.5mm. In all cuts, the second length is the dimension along the axial direction. The frame is 34mm long for the electrodes, with an additional 8.4mm on either side for mounting to the 4K shield. The frame is mounted to the 4K stage using 6 vented 2-56 screws. Each corner of the frame is machined flat and has 5 through holes for the electrodes to be screwed through.

The blade electrodes are designed to have a radial ion-electrode distance (r_0) of $550\mu\text{m}$, and an axial ion-electrode distance (z_0) of 2.2mm. These blades are machined using a wire EDM to allow for high precision and are mounted to the frame using a single, vented M2 screw. Between the blades and the frame are 0.5mm thick sapphire disks from Thorlabs. These disks are sold as components for Polaris mounts, but were chosen for their affordability and batch consistency. Each batch is machined from a sapphire sheet, so presumably all disks from a given sheet have the same thickness. Each was measured with a micrometer, and while this did not appear to be the case, the disks were sorted by their thickness prior to assembly. On the outside of the frame, a 2mm thick alumina piece serves as an insulating washer for each electrode on a corner. Three blades are mounted about the middle of the frame, and on the outer holes, the compensation electrodes are mounted. A pair of these are used to hold a piece of copper wire that provides either vertical or horizontal compensation. The wire is made into a U so that there is still optical access to the ions and each part of the wire is roughly 4.6mm from the trap center. Lastly, a piece of beryllium copper was machined to attach a thermal diode to replicate a trap electrode.

Further modifications have been designed for a future version of this trap. Electrodes are machined with venting holes and additional insets for sapphire disks to constrain the axial separation better. The frame has trapped holes for mounting PCB boards or magnetic coil holders. Beryllium copper rods were purchased to replace the copper wire. Lastly, each metal should be gold plated. This would have several benefits including better electrical conductivity at RF and microwave frequencies and a higher work function than beryllium copper for less trap charging during photoionization of our molecules. The biggest benefit would hopefully come

from improved thermal conductance between each piece of our trap. Since each blade is held using a single screw, it is likely that there is a large joint resistance at each interface [Eki06]. Gold plating would likely reduce the resistance significantly.

5.3.3 Trap Assembly

The trap was specifically designed to be straightforwardly assembled by hand, shown in Fig. 5.13. After being machined, cleaning was done using several stages of sonication in different solutions. Each electrode and sapphire disk was then measured using a micrometer and paired together. The goal was to pair them together to get the most similar ion electrode distance. Since the RF electrodes are the closest to the ion, they are the ones that require the most scrupulous assembly. To do so we set up an imaging system using an $f=200\text{mm}$ objective lens, a 50mm focusing lens, and a camera. Calibration was done first using a negative resolution test target. Next, the ion blades were screwed onto the frame without any spacers and an image was taken. Afterward the spacers were chosen to compensate and get the blades as uniform as possible. The sapphire pads spanned from $483\mu\text{m}$ to $528\mu\text{m}$ but most were between $493\mu\text{m}$ and $520\mu\text{m}$. To assemble the ion trap I needed to use a pair of tweezers, a screwdriver, and the necessary hex wrench. The first step, and one of the more difficult ones, is to start by putting in the RF blades. The blades are held in place with a single M2 screw, which wedges the spacers between the blade and the frame. On the outside of the frame, we have an alumina spacer and a M2 ring terminal. The alumina spacer serves to break electrical contact between the blade and the frame, as well as to make sure the trap doesn't loosen when cooled. Note that the height of the spacers was NOT chosen to make sure the thermal contraction of the screw matched that of the frame/spacer/terminal. Instead it was chosen so that we can confidently tighten the screws and not break the spacer. The thermal contraction rates are small, so it is unlikely that the lack of thermal matching will be an issue. A thin tube of alumina bisque was used between the RF screws and the frame to ensure that there are no shorts occurring beneath the spacer. After each of the RF blades have been tightened into place, the trap is placed on a 3-D stage to position it into focus of our imaging set up. A few combinations of pad pairs

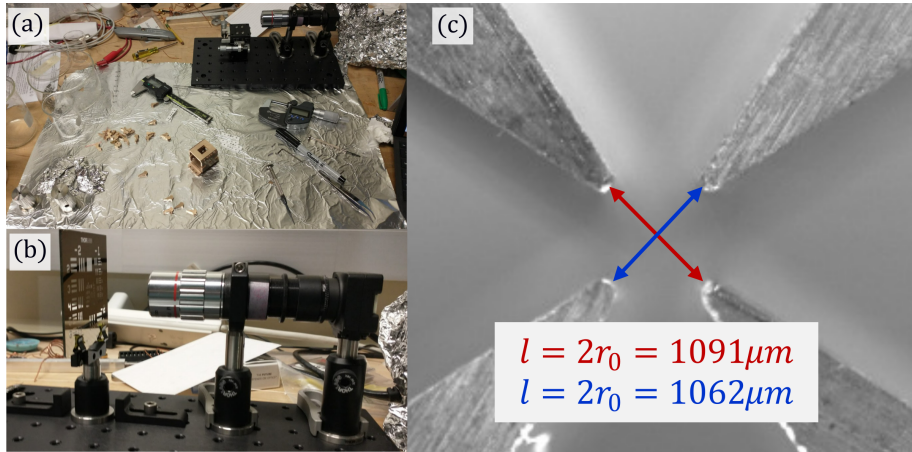


Figure 5.13: Assembly of the ion trap. After cleaning, the parts for the trap are laid on a new clean sheet of UHV foil (a). In addition to the parts, cleaned calipers, micrometer, screwdriver, hex wrench, and tweezers were prepared for assembly. In order to ensure that the blades are aligned properly, an imaging system (b) was used and calibrated with a negative resolution target. The trap blades are imaged (c) and the distance between electrodes is recorded. Based on this measurement, we predict there is 3% difference in angle between the blue electrode pair and the red electrode pair.

and blades were tried to get the blades well enough aligned. After the RF blades are in place, a similar but less rigorous procedure is done for the endcap blades, the compensation electrodes, and the temperature diode.

A single endcap electrode is made up of 4 blades. The initial plan was to have each blade in an electrode powered by its own DC supply. However, this plan was never implemented and instead all 4 electrodes are shorted to one another in vacuum, along the trap's frame. The pair of electrodes used for generating the trapping field (the RF electrodes) are also shorted together as close as possible in vacuum to limit phase mismatch. For similar reasons the wires chosen were carefully measured to try to limit any possible length mismatch. The other radial electrodes are what we refer to as the EGGs electrodes, for their purpose is to generate the electric fields for our EGGs scheme. These electrodes are not shorted together in vacuum to allow us to have phase control on them, which will become clearer later on as to why.

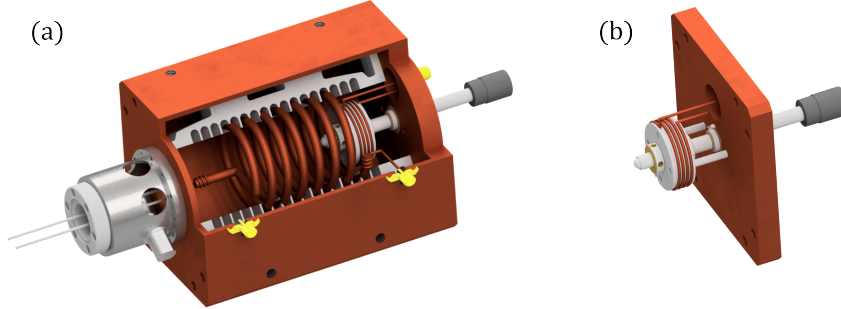


Figure 5.14: A helical resonator consists of a conductive enclosure referred to as the can, a main coil that largely determines the inductance, and an antenna coil that couples the input voltage onto the main coil. The full assembly with a quarter section view (a) shows the different components within the can including the coils and the coil holders, as well as the SMA feedthroughs for the coil and capacitive pickoff. A closer look (b) at the mounting of the antenna coil, shows that it can be translated using a micrometer rod.

5.3.4 RF Resonator

An important component for the ion trap is the resonator [Hen12]. It serves two purposes: the first is as a transformer and the second is as a narrow bandpass filter. The trapping voltage on the electrodes should be as stable and low-noise as possible, so the signal going into a resonator ideally is as low as possible. The more amplification needed, typically the more noise is present on the output. For this reason, the transformer aspect of the resonator is desired, converting more of the input power into a high voltage. Noise suppression will also be increased with a better resonator, so any undesired tones are filtered before they reach the ion. In either case, the way we succeed is with a higher quality factor.

Every ion trap will have some capacitance, typically somewhere between 1-100pF, as well as some resistance (depending highly on material choice). A helical resonator or can introduces some additional capacitance and resistance, which are typically smaller than the trap itself. It will also introduce a large inductance, which creates the RLC circuit. The first choice for the helical resonator is what number of

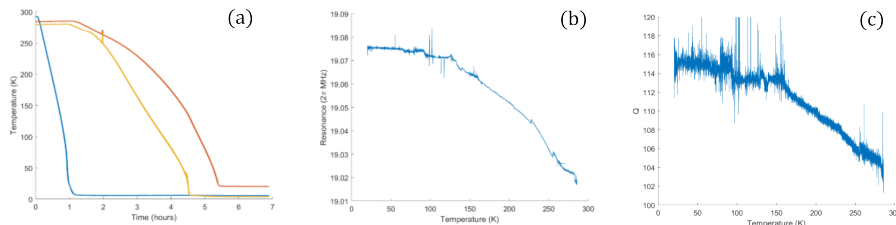


Figure 5.15: Resonator Performance as a Function of Temperature. (a) shows the performance of the cooldown in question. During the cooldown, the resonator was connected to a network analyzer, measuring the filter function near resonance. As the trap reached cryogenic temperatures its resonant frequency (b) changed from geometric changes and the quality factor (c) improved due to lower electrical resistance at cryogenic temperatures. The spikes we see in (b) and (c) are spurs from the network analyzer itself.

coils creates the designed resonant frequency

$$\Omega_{rf} = \frac{1}{\sqrt{LC}}. \quad (5.9)$$

The capacitance here is for the trap and resonator but is dominated by our trap. We have 36pF from our trap and 4pF from our can. The quality factor of the resonator is given by

$$Q = \frac{1}{R} \sqrt{\frac{L}{C}} \quad (5.10)$$

where R is the total resistance of both the can and the trap. An important consideration when talking about someone's can is whether it is loaded or unloaded. A loaded can includes the trap and an unloaded can does not. Unloaded cans will often boast very large quality factors (several hundreds), but this is misleading because their loaded value can easily drop by more than 50% (to Q 100). In order to determine the actual voltage on the electrodes we use

$$V_{rms} = \frac{V_{peak}}{\sqrt{2}} \approx \kappa \sqrt{PQ}, \quad (5.11)$$

where we introduce $\kappa = (\frac{L}{C})^{\frac{1}{4}}$. Improving the quality factor of a resonator allows us to reduce the power by the same amount, while keeping the voltage the same.

Two different resonators were used in this experiment. The first was used in the beginning when we first started trapping. It was built following the [Hen12]

paper and, being made from various plumbing parts we had lying around, it was not particularly stable. It required a special touch to align the antenna to the correct location within the can. It was used to create a $2\pi \times 21\text{MHz}$ resonance, that allowed us to quickly trap, but we formed concerns about its stability for future experiments.

To create a more stable resonator, we got an modified design for the “new” barium can. The postdoc at the time, Dr. Matthew Boguslawski had received the designs in question from a member of the Monroe group, although unfortunately I don’t know who to thank/cite. These can designs involved machined pieces that are assembled with mounting screws, an antenna on a micron translation stage, 3-D printed coil supports, and SMA connections for a DC input. All but the latter allowed this system to have significantly better performance and repeatability. The latter had no effect on the stability and was designed to allow for shimming voltages to be applied when a bifilar system is used like in the barium experiment. Careful consideration must be taken when using this connection because the resistance of what ever it is connected to will be added to the resonator and can tank the quality factor is caution is not taken. In our experiment, we use a single coil and use a grounding cap to short to the wall of the resonator.

The quality factor of the resonator is most typically measured using the network analyzer. An alternative way is to look at the ring down of a voltage applied to it. This is not a common way, but was tested as well to confirm the standard measurement. The voltage will decay as

$$V(t) = V_{peak} e^{-\frac{\Omega_{rf} t}{2Q}} \cos\left(\Omega_{rf} t \sqrt{1 - \frac{1}{4Q^2}}\right), \quad (5.12)$$

and fitting the measurement resulted in a quality factor of 130 at $2\pi \times 19\text{MHz}$. These results were in good agreement with the results from the network analyzer measurement.

Not shown in Fig. 5.14, is a pair of capacitors that serve to pick off a small fraction of the voltage without introducing a large capacitance to the system, following [K 16], in order to actively stabilize the trapping voltage. Around 1% of the trapping voltage is picked off using a capacitive divider built from a 0.2pF and 20pF capacitor, before going into a temperature-compensating rectifier, replicated for us by Peter Yu based off of [K 16]. The signal coming from the rectifier then went through a several-meter-

long SMA cable to a Newport servobox for locking. We eventually found that this cable would pick up a substantial amount of noise, leading us to lock our resonator using a voltage variable attenuator and the pickoff from a coupler just before our RF amplifier.

A final addition to our helical resonator is a TEC mounted to a side of the can with a heatsink. This TEC is controlled using a Peter box, reading the temperature of a thermistor epoxied into an aluminum block with tapped threading that is screwed directly into the can. Due to the large thermal mass, it is challenging to temperature stabilize the resonator, so this system is not typically in use.

5.3.5 DC Control

To produce the DC voltages used on this trap, we had our electronics lab build us a high voltage DC box referred to as AMO8. This box was originally designed for the HUNTER experiment [CL21]. Its input is 1kV from a Bertan 315B high voltage linear power supply. This was the most stable high voltage supply we could find, which is important for a highly stable trap. The AOM8 box is designed to produce 28 outputs; we only use 5 of these. The endcap DC voltages are the highest at about 200 and 350V, followed by the shimming (50-70V), and the A-ramp voltage (2V) that is applied to both of the EGGs electrodes. The smallest step size is 0.1V.

5.3.5.1 Endcaps

As mentioned earlier, there are 2 endcaps each made up of 4 blades shorted together in vacuum. The blades were chosen to be shorted together so as to require less troubleshooting should something go wrong during the initial loading and testing of the trap. Since the trap was never replaced or modified after loading began, there was never an opportunity to try loading with them individually controlled. The additional channels mentioned for AOM8 were desired should we switch to individual control. This manner of control is similar to what is used in the barium blade trap for shimming.

5.3.5.2 Shimming

In our experiment, we perform shimming using an additional pair of electrodes. One electrode is under the trap and the other is on the north side of our trap, to allow for vertical and horizontal control respectively. Shimming is an important step in ion trapping since it is used to reduce excess micromotion [BMB98]. Stray voltages can fluctuate as charging and discharging of the trap electrodes can move the ion out of the trap null. Several methods for shimming exist. The simplest method involves changing the trap depth by reducing/increasing the trapping voltage. A strongly confining voltage will produce a tighter potential well; a larger excess voltage is required to push the ion off of the trap null. Alternate between a strong and a weak trap depth, until the ion's position on the camera does not change on the camera. In our trap this method works only for horizontal shimming, and vertical shimming is measured using the ion fluorescence. When it is well shimmed the fluorescence will increase. This method is typically only used in the initial testing stage of the ion trapping to get a rough idea of what voltages are needed for shimming.

A more rigorous method for shimming is done using Rabi oscillations on the carrier and on a micromotional sideband [Gug17]. The electric field generated by the quadrupole laser will be given by

$$E(t) = E_0 e^{-i\omega_l t} e^{ikx} = E_0 e^{-i\omega_l t} e^{ikx_o(1 - \frac{q}{2} \cos(\Omega_{rf}t)) \cos(\omega_0 t)} \quad (5.13)$$

where we use Eqn. 2.22 for the ion's position within the trap. Changing to the reference frame of the ion's harmonic motion allows the $\cos(\omega_0 t)$ term to be dropped so that the electric field is described by $E_0 e^{-i\omega_l t} e^{ikx_o} e^{ikx_o \frac{q}{2} \sin(\Omega_{rf}t - \frac{\pi}{2})}$. The last exponential in terms of the Bessel function of the first kind takes the form $e^{ikx_o \frac{q}{2} \sin(\Omega_{rf}t - \frac{\pi}{2})} = \sum_{n=-\infty}^{\infty} J_n(\beta) e^{in(\Omega_{rf}t - \frac{\pi}{2})}$, where $\beta = kx_o \frac{q}{2}$ is referred to as the modulation index. β is typically relatively weak ($\ll 1$) which allows the ratio of the carrier ($n=0$) and first order micromotion sideband ($n=1$) to be approximated to

$$\frac{J_0(\beta)}{J_1(\beta)} = \frac{\Omega_{carrier}}{\Omega_{MMSB}} \approx \frac{2}{\beta} \quad (5.14)$$

where $\Omega_{carrier}$ and Ω_{MMSB} are the Rabi ratios for the carrier oscillation and the first order micromotion sideband such as in Fig. 5.16. The ion's displacement from RF

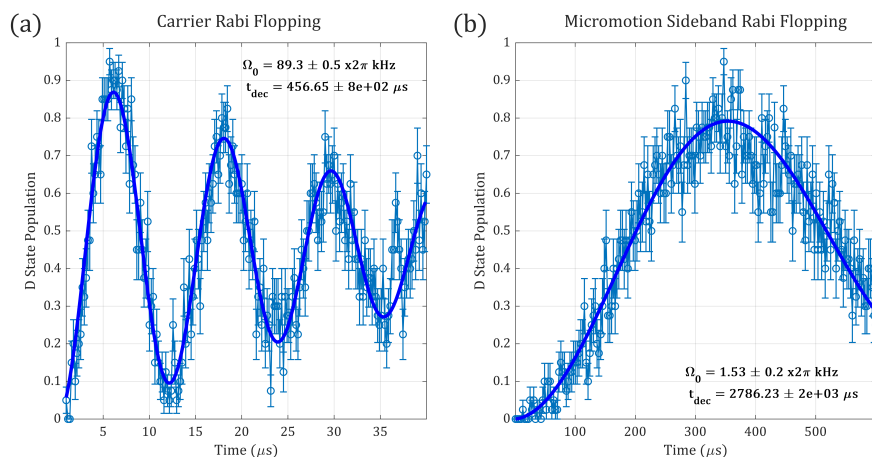


Figure 5.16: Example of the (a) carrier and (b) micromotion sideband (MMSB) Rabi oscillations. The ratio of these Rabi rates is used to estimate how well shimmed we are. Better shimming will increase the carrier rate and reduce the MMSB. Since the electric fields from all three shimming directions are needed to generate an electric field along the 729nm beampath, this method is infrequently used in our experiment.

trap null is described by $x_0 = \frac{2\beta}{qk}$. Since the ion can be described by its harmonic motion ($\ddot{x} = -\omega_o^2 x$), the Coulomb force to displace the ion by x_0 is given by

$$F = eE = m\ddot{x} = -m\omega_o^2 \frac{2\beta}{qk}. \quad (5.15)$$

Thus a measurement of the ratio of the carrier and micromotion sideband Rabi oscillations indicates how far off of the trap null the ion is, as well as what applied electric field will be required to move it back towards the null. While this method does provide a method to improve our shimming, it is challenging to implement because there is a single quadrupole laser and this method works by shimming along the laser k-vector; the shimming electrodes each have a projection onto this vector, complicating the measurement. To successfully implement this method, there should be three beampaths, each one moving along the direction of the field produced by the shimming electrodes.

The method for radial shimming that replaced this Rabi ratio technique is done using parametric modulation on the trap electrodes taken from [D 21a]. When the parametric drive is resonant with the secular motion of the ion, the ion will be excited. Displacements further from the trap null will increase the amplitude of the motion. Minimizing the displacement will reduce the signal amplitude. Scanning the

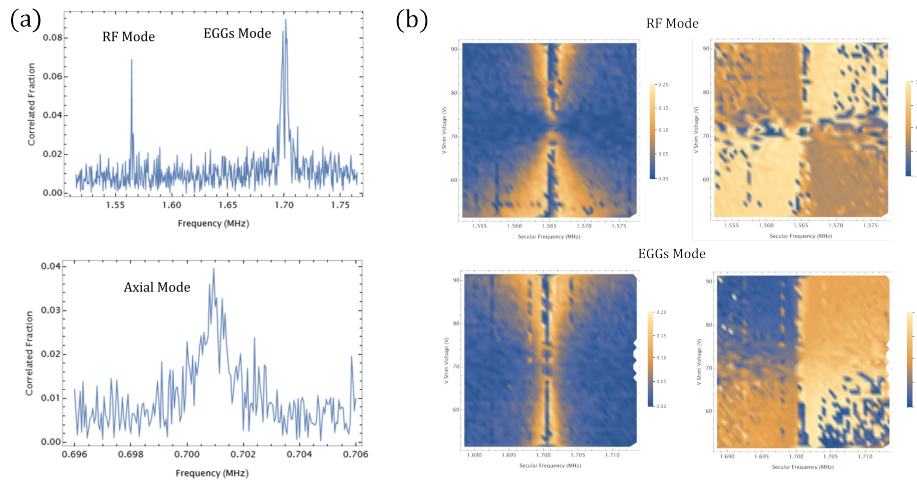


Figure 5.17: Shimming using a parametric sweep involves sweeping through the mode frequencies and one of the radial shim voltages, while keeping the other shim voltage constant. We typically sweep the vertical shimming electrode (V_{shim}). The color in the 2-D plot indicates the intensity of the correlated fraction; blue indicates a smaller fraction and yellow indicates a larger fraction. Typically, a smaller fraction means less heating, however, if the heating is large enough it can become saturated and decrease. Hence the blue patches in the center of the yellow. Each mode will have a preferred V_{shim} voltage, indicated by the horizontal dashed green line, representing the minimal micromotion. The horizontal shimming voltage is adjusted until both modes result in the same V_{shim} voltage. Experimentally, we find that one volt increase on the horizontal electrode will increase the compensated vertical voltage by approximately one volt on the EGGs mode, and will decrease it by approximately one on the RF mode.

shimming voltage and the frequency will produce a 2D map of the trap null for an individual mode shown in Fig. 5.17. By scanning over both radial modes the proper shimming voltages can be measured. In practice both the vertical and horizontal shimming electrodes need to be scanned together, either manually setting one of the shimming voltages and sweeping the other, or by using an automated sequence that iterates the sweeps until the two modes are at their minima. A weak projection onto the axial mode, limits practical application of this method to the two radial modes.

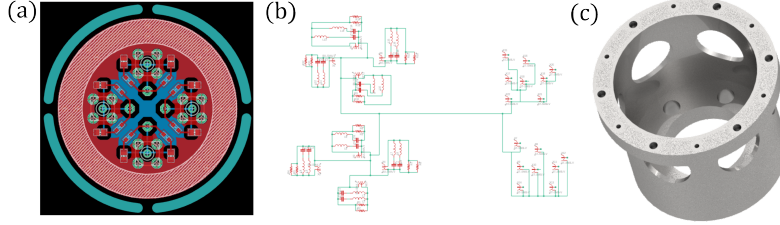


Figure 5.18: For the DC and EGGs feedthroughs custom PCB's were designed. (a) The PCB has 4 SMA inputs that go directly to a solderable electrical contacts that connects to the feedthroughs. Optional pads were included for filtering. Example circuit diagram used to create the PCB (b). Each PCB is mounted directly to a metal mount that is connected directly to the vacuum chamber by sitting over a 1.33" electrical feedthrough (c). Electrical contact is made between the chamber and the grounding plane on the PCB.

5.3.5.3 Filter Boards

I designed a PCB for simple filtering that attaches to the electrode feedthroughs and a PCB vacuum mount (Fig. 5.18). The vacuum mount is a design made by Dr. Schneider for some of the previous Hudson experiments [Pur19, Mil20]. The PCB has a grounding plane that is in physical contact to the vacuum chamber when connected to the mount. It has pins that connect to the feedthrough wiring, and converts that line to an SMA coaxial connection. Along that wire are pads for either a single component to ground or two components in series to ground; each option is available twice. We were unsure what filtering we may want/need and this choice was made for flexibility. We have switched between three different boards depending on the goals of the experiment at the time. The simplest board is unfiltered, which allows us to apply tickle voltages to our DC electrodes. Next is a simple filter board with a single 7nF capacitor to ground on each input pin, for reducing any RF noise on these electrodes. The last one, was a more complex filter made by Peter Yu. Five capacitors (combined to 1.1nF) to ground per electrode are attached by removing some of the insulating layer and a resistor was added in series between the input and output. The values were chosen either for the endcaps or for the shimming electrodes, to filter the endcaps more than the shimming electrodes ($1k\Omega$ for shimming, $1M\Omega$ for endcaps).

5.3.5.4 Loading

Depending on a given experiment, we have used slightly different DC voltages for operation. Typically these are hundreds of volts on the endcaps. Early on, we would load with the same voltages that we operate the experiments at, but eventually saw our loading rate decrease. What previously took 5 minutes started taking 20. An observation we made was that loading improved if the endcap voltages were reduced to about 20V even without any shimming voltages. This has become our standard method of loading a calcium ion: reduce the endcaps and turn off the shimming voltages.

5.3.6 Heating Rates/Coherence

Coherence is categorized for both the spin of the ions as well as the state of the quantum harmonic oscillator. Ion coherence will be described as spin flip errors ($\Gamma_{sf}\sigma_x$) or dephasing errors ($\Gamma_{sd}\sigma_z$), where Γ is the decoherence rate for a given loss channel [RZR99]. Quantum harmonic oscillators experience either heating/cooling errors ($\Gamma_h\hat{a}^\dagger/\Gamma_c\hat{a}$) or dephasing errors ($\Gamma_d\hat{a}^\dagger\hat{a}$) [TMK00,MKT00]. Spin flip errors are determined by the lifetime of the excited state, for Ca^+ 's $D_{\frac{5}{2}}$ state this is about 1.05 seconds ($\Gamma = 2\pi \cdot 1.52\text{MHz}$) from the spontaneous decay of the state back to the $S_{\frac{1}{2}}$ state. The hyperfine states will have much shorter spontaneous emission rates since they are 10's MHz separated, and Fermi's golden rule means they will be long lived compared to the 100's THz optical decay. A dephasing error is measured using a Ramsey protocol, transferring the spin to the $|+\rangle = \frac{1}{\sqrt{2}}(|\downarrow\rangle + |\uparrow\rangle)$, waiting some time τ , before doing another pulse to restore the spin to the initial state ($|\downarrow\rangle$ or $|\uparrow\rangle$). Fluctuations in the magnetic field cause the energy splitting of these states to change. In the Bloch sphere picture, a change in the energy between the two states produces a phase change because the applied field is now off-resonant show in Fig. 5.19. Without implementing a spin echo procedure, the spin dephasing time is 3.6(2)ms, which is extended to 24.6(1.6)ms with the introduction of two concatenated XY-8 dynamic decoupling sequences [ZD12,CH22]. For the molecular ion, the population would be in a given state and would have a long spontaneous decay lifetime, and won't experience any dephasing effect because rotations about the z-axis when on

Spin Dephasing

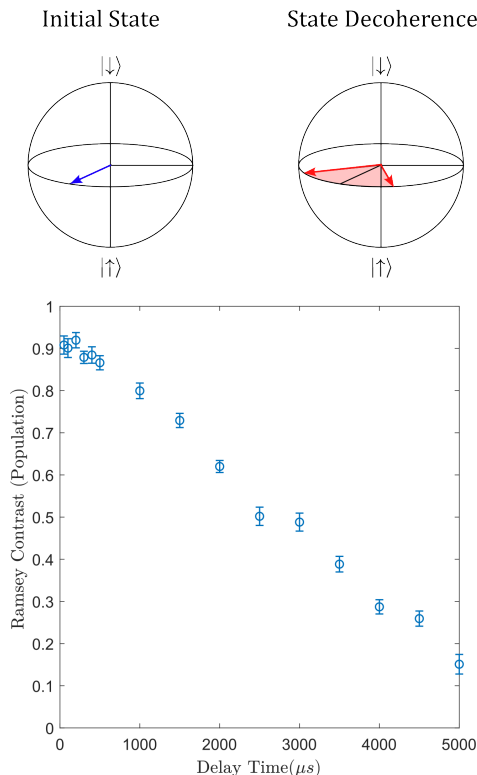


Figure 5.19: Spin dephasing can be visualized on the Bloch sphere as a blurring out of a state from rotations about the z-axis. By performing a Ramsey experiment and increasing the delay time between the initial $\frac{\pi}{2}$ pulse and a subsequent $\frac{\pi}{2}$ pulse that returns it to the initial state, we are able to measure a loss in contrast between Ramsey fringes that result from spin decoherence.

the axis won't lead to any dephasing.

For the quantum harmonic oscillator, heating effects occur on each mode, increasing the phonon number (for other systems like superconducting qubits, cooling is this loss channel rather than heating). Heating rates are measured by cooling a single Ca^+ into its ground motional state $|n=0\rangle$, and waiting some time before reading out each of its sidebands. The ratio of sidebands is then converted into a phonon number, the result of which are shown in Fig. 5.20 for the RF mode. Higher heating rates occur on the radial modes because the trapping voltage for radial confinement comes from a high voltage source with a smaller distance between the ion and the trapping electrode. As such the electric field the ion experiences is much higher. The axial confinement is provided by stable DC electrodes, and has a weaker projection onto the ions. DC has smaller fluctuation ($200 \pm 0.01\text{V}$) compared to the RF ($200 \pm 0.1\text{V}$). The lowest heating rates we achieved were $\dot{n}_{EGGS} = 0.98(8)$ quanta per second, $\dot{n}_{RF} = 0.72(9)$ quanta per second, and $\dot{n}_{Axial} = 0.39(21)$ quanta per second. To determine the dephasing of the quantum harmonic oscillator, after preparing the ion in the ground motional state ($|n=0\rangle$) we displace it by some amount α , wait some time τ before antidisplacing it with $-\alpha$. Instabilities in the secular motion

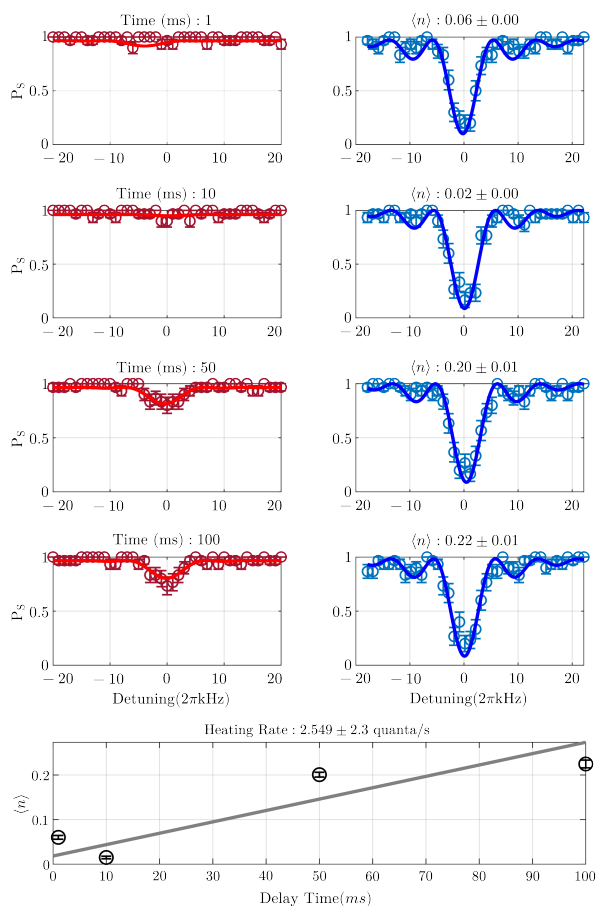


Figure 5.20: The motional analogy to a spin flip, motional heating of the modes leads to decoherence of the prepared state. Shown is the heating rate measurement performed on the RF mode of a single ion crystal. A measurement of phonon number is done using the sideband ratio method, assuming a thermal state distribution. Lower heating rates are measured on the axial mode since the DC supplies are able to create a potential with less noise than the RF trapping.

Motional Dephasing

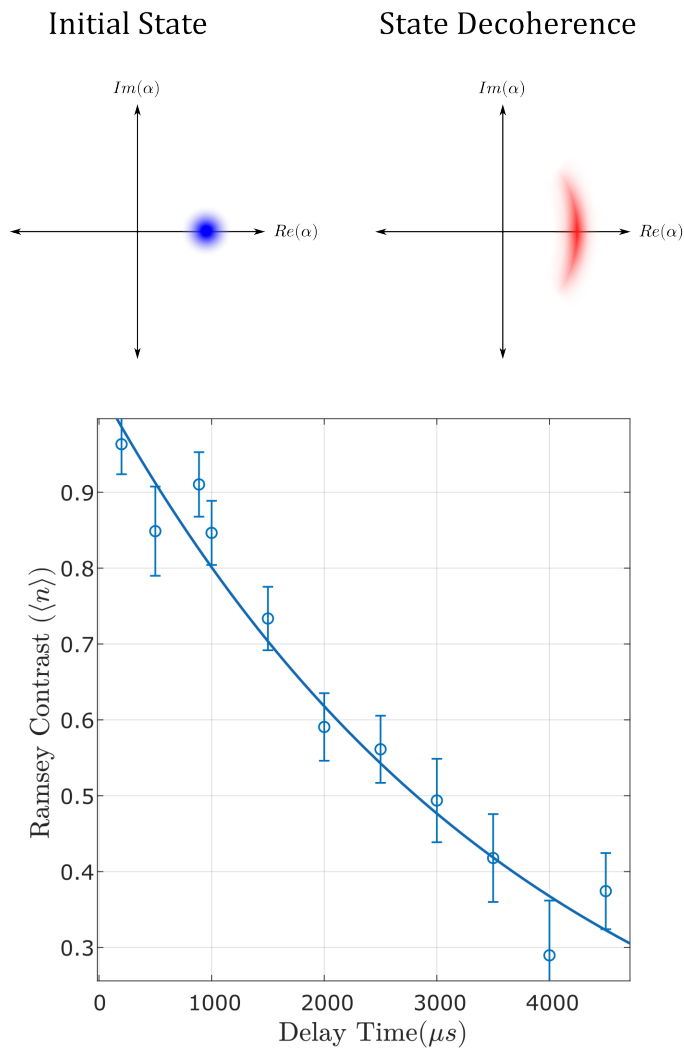


Figure 5.21: Each mode will experience motional dephasing, analogous to the spin dephasing. Motional dephasing can be visualized as the state blurring out in phase space from an unstable secular frequency. It will sometimes drift red and sometimes drift blue of the true frequency, rotating in phase space. To measure the loss of contrast, a Ramsey like experiment is done, displacing the ion, waiting some time, and then antidisplacing it back to the motional ground state. Shown is data taken for the EGGs mode; the RF mode is comparable and the Axial is significantly longer.

of the oscillator result in the decoherence and the ion does not return to the initial state. A loss of contrast between the peaks and troughs of the Ramsey, such as shown in Fig. 5.21 indicates the displaced state has decohered. Without implementing a spin-echo technique, the lowest T_2 times we achieved were $1084(78)\mu\text{s}$ for the EGGs mode, $717(52)\mu\text{s}$ for the RF mode, and $150(27)\text{ms}$ for the axial mode. Using a spin-echo sequence, the motional dephasing on the radial modes can be reduced by five times, for example the T_2 time for the RF mode was $3.94(51)\text{ms}$.

5.4 Lasers

A running joke about this experiment is that for a laser-free qubit, it has an awful lot of lasers. So while yes, the electric-field gradient scheme for molecular qubits would be laser-free, it still requires fine control over the atomic ion for cooling, motional state preparation, and readout. Lasers are used for photoionization, cooling (Doppler and sideband), and for spin-motional coupling of the Ca^+ with the phonon modes of the trap. Most of the lasers used are external cavity diode lasers (ECDL) from Toptica. The exceptions are a free-running diode for photoionization of calcium and the dye laser system (Quanta Ray + Sirah) for photoionization of HCl.

On our 5' by 10' optical table sits our chamber, with a surrounding mezzanine onto which light is fiber coupled, as well as photoionization lasers for calcium and the calcium Doppler cooling laser and the repump lasers. The quadrupole laser sits on two other tables and is fiber coupled to the mezzanine. One of these tables has the dye laser and its pump on it; light from the dye laser is sent to the chamber through free space optics.

5.4.1 Ca^+ Photoionization Lasers

Calcium ions are ionized using a two step ionization scheme [DS04]. The first step at 423nm resonantly excites neutral calcium from the ground state to the excited state, $4^1S_2 \Leftrightarrow 4^1P_1$. The next step requires a photon with a shorter wavelength than 390nm to excite to the continuum, removing an electron and ionizing the atom. In our experiment we use a Toptica DL Pro to generate the resonant 423nm light. The light for the second step is generated using a free running diode around 377nm from Nichia (NDU4116). Because this is not a resonant step, the diode does not require fine control and is not temperature stabilized. During loading we typically use $\sim 1\text{mW}$ of 423nm light and $\sim 0.75\text{mW}$ of 377nm light. The lasers have large beam diameters of several hundred microns. We wanted to have large beams to minimize the need for a rigorous alignment; we align the lasers roughly to the center of the ion trap using the camera and minimizing clipping of light on the blades.

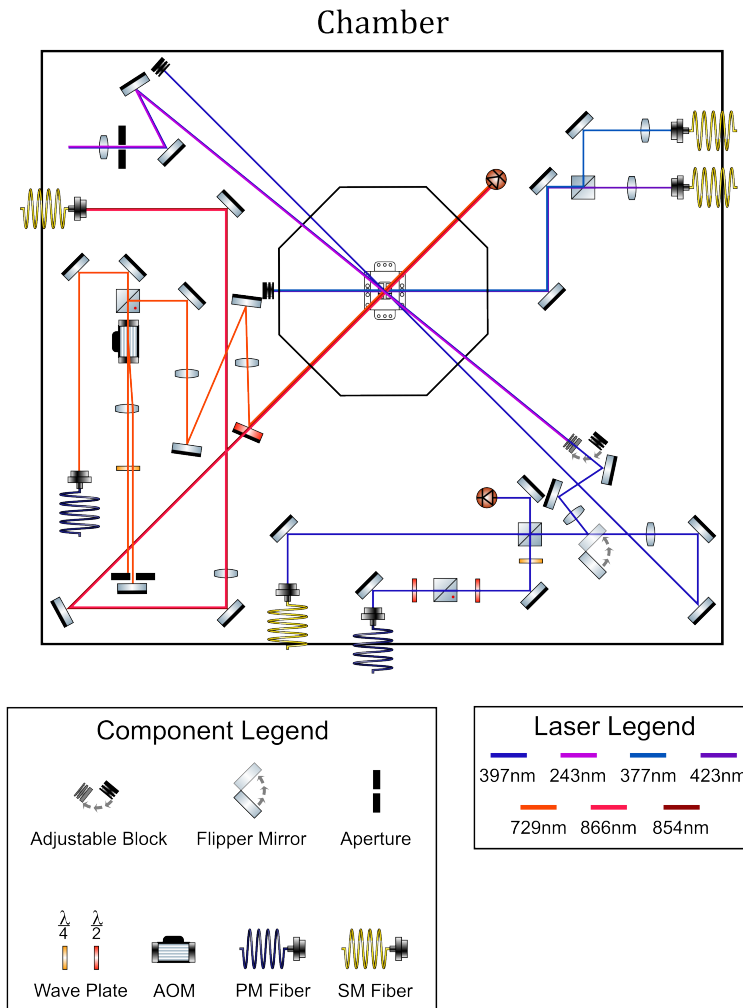


Figure 5.22: In order to minimize the post height of the laser optics going into the vacuum chamber, a mezzanine of optical breadboards is elevated above the optical table. All lasers are fiber coupled onto the breadboards, with the exception of the 243nm light, which is sent, free-space, from a nearby optical table that houses the dye laser. Both 397nm beampaths are overlapped on the mezzanine, with polarization optics on the spin-polarization beampath to ensure the correct polarization of light. A manual flipper mirror is used to overlap 243nm light with a 397nm path that is hitting the ion. The Ca^+ photoionization lasers are overlapped on the table. Prior to overlapping with the 866/854nm light, the 729nm light goes through a double-pass AOM setup.

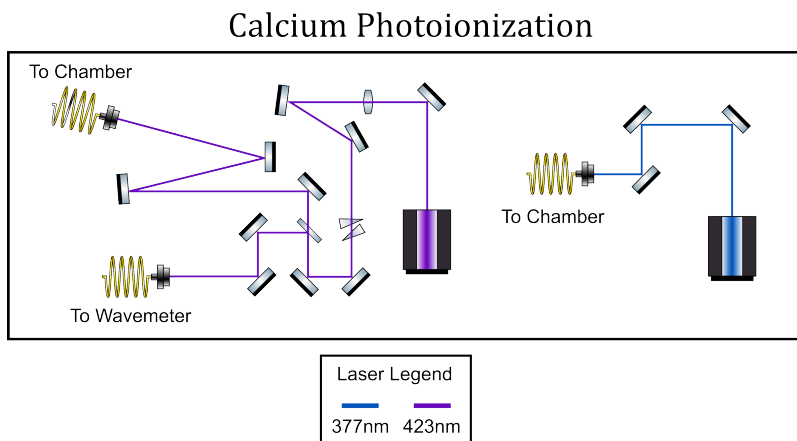


Figure 5.23: Two lasers are required to photoionize calcium, a 423nm ECDL laser and a free-running 377nm diode. The former is a resonant transition in neutral calcium, requiring a channel on the wavemeter, whereas the latter takes it to the continuum and does not require frequency monitoring.

5.4.2 397nm Doppler Cooling Laser

The Doppler cooling transition for Ca^+ drives population from the $S_{\frac{1}{2}} \Leftrightarrow P_{\frac{1}{2}}$. For optical pumping to one of the Zeeman states of Ca^+ , a second circularly polarized beam is required. A Toptica DL Pro with integrated 30dB isolator is used to generate narrow linewidth, stable light but requires a series of cylindrical lenses to collimate and balance beam waists shortly after leaving the laserhead. A fused silica window removes a small fraction of the light to reflect it into a fiber going to the wavemeter. Following the window is a half waveplate and PBS to adjust the power going to the main beam path and to the spin polarization beam bath. The transmitted light goes to the spin polarization path, where a double-pass AOM setup enables fast switching and frequency tuning without changing beam drifting. The double-pass setup involves a PBS before the AOM to linearly polarize the light, a lens centered to the $k=0$ order light that will collimate the $k=1$ order light, regardless of the frequency (different frequencies change the angle of the $k=1$ beam out of the AOM). A quarter waveplate rotates the polarization, before an iris that removes the $k=0$ order light, and then again after it is reflected by a mirror. As this light goes back through the AOM along the initial beampath, it experiences a second kick, overlapping the (total) $k=2$ beam with the input. Due to the rotations in polarization the light picked up, it

now will be reflected by the PBS. The light then goes to a PM single mode fiber, with a waveplate in front to optimize the coupling efficiency. The main 397nm light goes through a nearly identical process to get fiber coupled to the table, the exceptions being the light does not involve a PM fiber, and that the shutter that blocks the $k=0$ light on the first pass of the AOM is electronically adjustable (Thorlabs ELL15). This aperture is opened to allow the $k=0$ light to reach the ions which, because of how we oriented our AOM, will be red detuned by $\sim 200\text{MHz}$. This red detuned light is convenient for loading and rescuing the ions because it can quickly crystallize hot ions.

Both beampaths take the light onto the chamber mezzanine, and are overlapped onto an unpolarized beamsplitter. Prior to the beamsplitter, the spin polarization light goes through a half waveplate and PBS to fix the polarization to linear, after which another half waveplate and quarter waveplate are used to set the necessary polarization for optical pumping. The overlapped beams are sent through a focusing lens before going into the chamber. A separate beampath, accessible by flipping up a mirror, is used for aligning the photoionization laser for HCl^+ . This beampath confirms the light will be hitting the center of the trap. The beam waist for these lasers is $\sim 60\mu\text{m}$, and the main beam is typically $\sim 400\mu\text{W}$ with the spin polarization beam having $\sim 200\mu\text{W}$ at full power. The main beam typically has $26\mu\text{W}$ for Doppler cooling and $\sim 270\mu\text{W}$ for readout. For optical pumping, the spin polarization beam has $\sim 20\mu\text{W}$. When the aperture is open, the rescue light has typically $\sim 1\text{mW}$.

5.4.3 866nm and 854nm Repump Lasers

Population in the D states of Ca^+ is removed by two lasers, $D_{\frac{3}{2}} \rightarrow P_{\frac{1}{2}}$ with 866nm and $D_{\frac{5}{2}} \rightarrow P_{\frac{3}{2}}$ with 854nm. 866nm light is the main repump laser used for optical cycling during Doppler cooling of Ca^+ . While the 397nm ECDL can accidentally pump population to the $P_{\frac{3}{2}}$ on occasion, the main reason behind the 854nm laser is for removing population that was shelved into the $D_{\frac{5}{2}}$ state using the coherent 729nm laser. Both lasers are Toptica DL Pro's with integrated 30dB optical isolators and about 75mW of output power directly out of the laserhead. The diodes used in these lasers can be tuned from 840-875nm. A DFB Pro was also purchased for

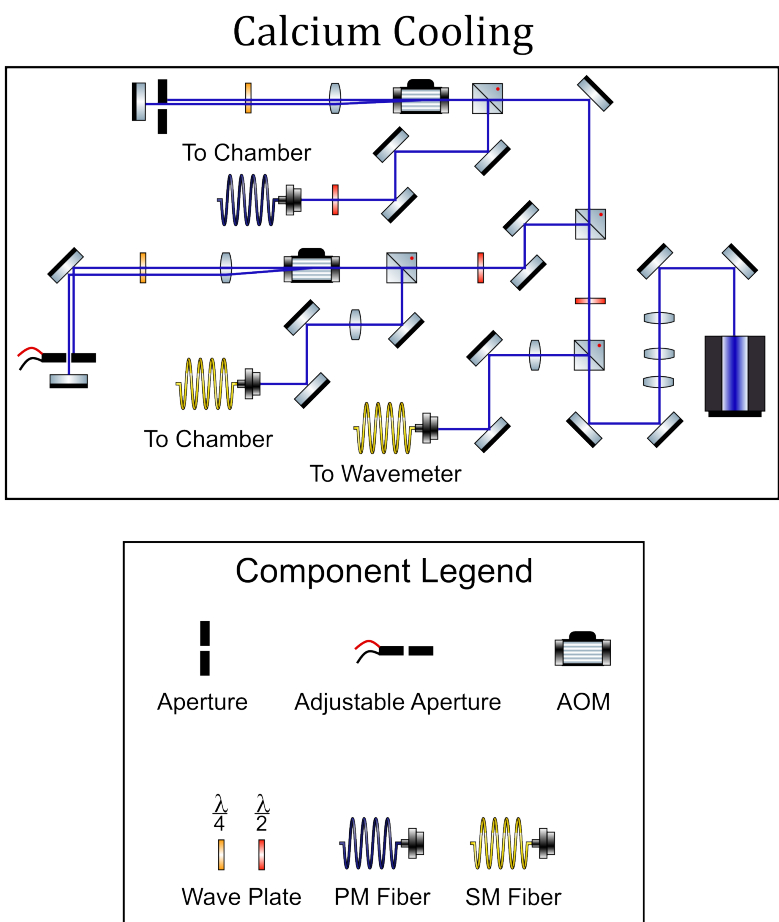


Figure 5.24: Components used within the 397nm laser system. See text for a description of components.

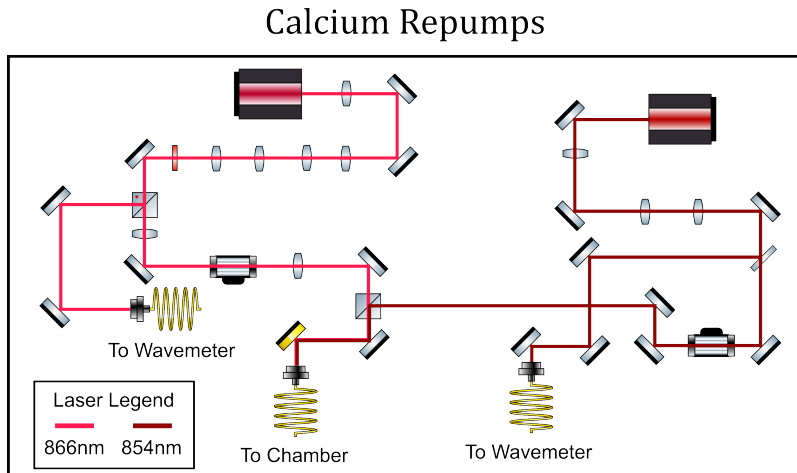


Figure 5.25: Components used within the 866nm and 854nm laser system. See text for a description of components.

854nm, allowing one of the DL Pro's to be available for generating 849nm light for the $D_{\frac{3}{2}} \rightarrow P_{\frac{3}{2}}$ transition; however we sent the laser back for repairs and ended up keeping the DL Pro as our permanent option.

The light emitted from the lasers needs to be collimated, and the elliptical nature required us to use cylindrical lenses to compensate for the imbalance. After collimation, a portion of the light is picked off to wavemeter channels. The 866nm laser used a half waveplate and a PBS to adjust how much is sent to the wavemeter fiber; the 854 uses a glass window to pick off a small portion instead. The light that is transmitted from both lasers each go through their own respective AOMs, which serve as fast switches for our pulse sequences. Since both lasers are close in frequency, we overlap them into a single SM fiber that takes the light to our chamber. On the chamber mezzanine, the light exits an adjustable collimator and goes through a single $f=300\text{mm}$ lens (LA1484-B) to focus onto the ions with a beam waist of $\sim 100\mu\text{m}$. We send about $\sim 180\mu\text{W}$ of 866nm light and $\sim 30\mu\text{W}$ of 854nm light into our vacuum chamber.

5.4.4 729nm Cavities

The metastable $D_{\frac{5}{2}}$ in Ca^+ is accessible coherently using narrow linewidth 729nm light to drive the $S_{\frac{1}{2}} \leftrightarrow D_{\frac{5}{2}}$. A high finesse cavity system ($\mathcal{F}=200000$, FSR=1.5GHz) from Stable Lasers Systems is used to narrow the linewidth of a Toptica DL Pro (nar-

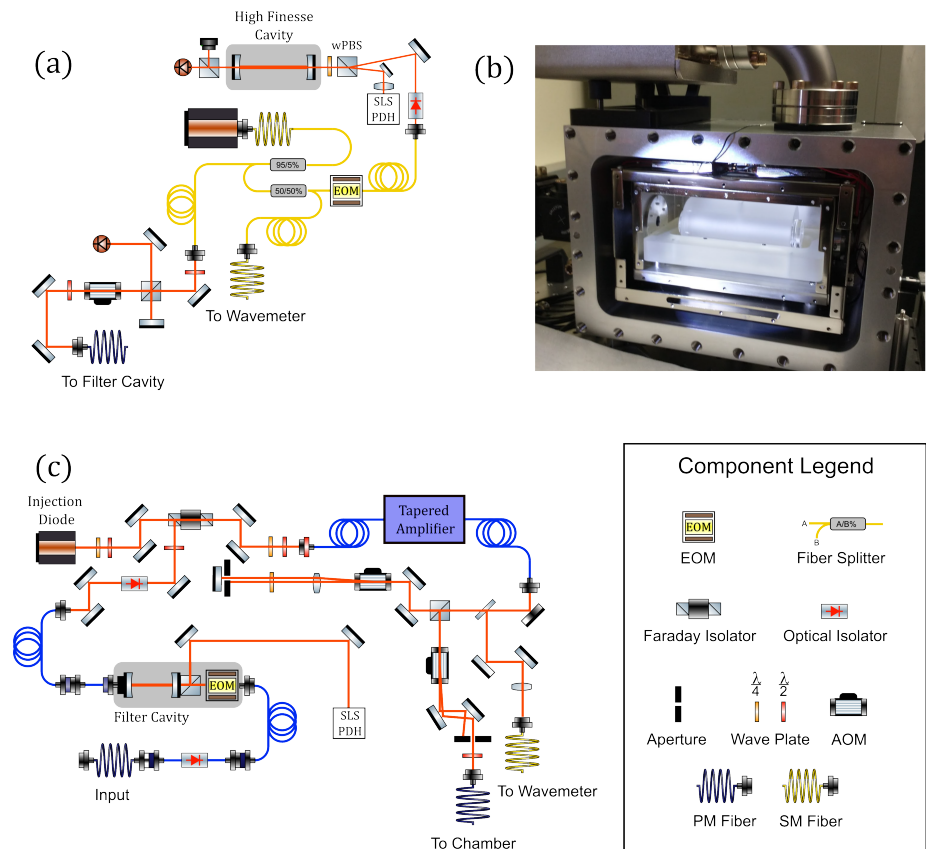


Figure 5.26: Since 729nm light plays an important role in our experiment it is one of the more complex setups. A system by Stable Laser Systems (SLS) (a) featuring a high finesse cavity (b) was originally purchased for coherent operations with the metastable state. An eventual addition was made by Josh Rabinowitz, featuring a filter cavity and tapered amplifier, purchased from SLS and Toptica, respectively. The filter cavity reduced the servobumps that are commonplace in cavity systems, allowing us to operate at higher powers. Additions after the TA are another doublepass to eventually replace the one on the mezzanine, and a singlepass for implementing bichromatic operations,

row linewidth option) to below 1Hz. This cavity shown in Fig. 5.26, is a single fused silica crystal mounted within two layers of radiation shielding inside of a vacuum chamber pumped by a Gamma ion pump (8.2E-8 mbar). The heaters on the outer radiation shield are used to stabilize the temperature of the cavity to 33.51°C. Light from the Toptica laser is fiber coupled using a Fiberdock to a 95/5% fiber beamsplitter, with the majority of the light going to a fiber noise cancellation system before being sent to the filter cavity. The fiber noise cancellation is meant to reduce the noise that is picked up through a fiber, using light reflected back through the output of the fiber after it hits a flat surface, but was intended for the system prior to the filter cavity installation. The other 5% of the laser light gets split again, half to the wavemeter output and half to the cavity for locking. For locking, the 2.5% goes through a fiber EOM for phase modulation after which it is collimated and directed into the cavity. Optical isolation results in horizontally polarized light being directed into the cavity through the Wollaston polarizing beam splitter, after which a quarter waveplate sits, so that the cavity reflected light is not vertically polarized going back through the wPBS, directing it onto the PDH detector. Cavity performance is monitored via the transmitted light sent onto a photodiode and camera, the former for locking and the latter for confirmation of the correct cavity mode. To cover a wide frequency range, the SLS system is designed to lock to the carrier, first order sidebands, and even second order sidebands ($J_0, J_{\pm 1}, J_{\pm 2}$) fringes. Typically, locking to the carrier is the easiest, and sometimes may be a necessary step prior to locking to a sideband if the system is having trouble immediately locking to a sideband.

A common problem with locking to a cavity is that the light from the laserhead develops servobumps [Pru14]. Light transmitted through a cavity does not possess this noise shown in Fig. 5.27, but it is too weak to use for any application ($< 4nW$). Amplification of the laser light would also result in amplification of this noise, so its reduction was prioritized. A custom filter cavity system was purchased from Stable Lasers Systems to suppress the servobumps, which has a linewidth of 176kHz with a finesse of ~ 11300 . Light from the main cavity box, goes through a 30dB fiber isolator before going into the filter cavity (without this isolator the high finesse cavity would become unlocked). This cavity also reduces the carrier light by more than an order of magnitude, requiring additional amplification before seeding a tapered amplifier. The

output of the filter cavity goes to another patchcable, which facilitates the alignment to the injection diode. Out of this cable, there is $\sim 200\mu W$, which goes through a 30dB optical isolator into a 60dB Faraday isolator. A half waveplate optimizes the reflection in the Faraday isolator, which sends the light into an injection diode. This diode is housed in a temperature-controlled mount ($20.5^\circ C$) and controlled with a stable current controlled (97mA) designed by Peter Yu. The light from the filter cavity seeds this diode, outputting 11mW of usable light that is sent into a Toptica Tapered Amplifier (TA).

Coming out of the TA is $\sim 150mW$, a small portion of which is picked off by a window and sent to the wavemeter. This signal is used to monitor the lock on the filter cavity as well as the seeding of the injection diode. The rest of the light is sent into a doublepass AOM setup, the output of which is a single pass AOM for bichromatic light operations. The light is then coupled into a high power PM fiber onto the mezzanine, whereby it goes through another doublepass AOM setup before being focused down and overlapped onto the repump light. At full power, there is $\sim 50mW$ of 729nm light out of the fiber, and $\sim 15mW$ that enters the chamber. The beam waist is focused down to $60\mu m$.

5.4.5 Dye Laser

The loading of HCl^+ requires a 2+1 resonantly enhanced multiphoton ionization (REMPI) process using 243nm light, which is generated using a dye laser system combining a Spectra Physics Quanta-Ray and Sirah. The Quanta-Ray is a pulse Nd:YAG laser that frequency triples 1064nm light to produce 355nm light for pumping the dye laser. Typical operation produces around 170mJ of 355nm light. The light is then sent into the first section of the Sirah, the dye laser. A beam splitter separates the input light: half directly goes to the oscillator dye cell to produce the seed light, and the other half is further split for pumping the preamp and the amplifier dye cell. The oscillator emits light that goes to a cavity grating that produces the seed light for the rest of the process, which is sent back through the dye cell at a different height. The dye cell at this height is now referred to as the preamp, which is pumped by other 355nm light, and begins lasing when the seed light enters

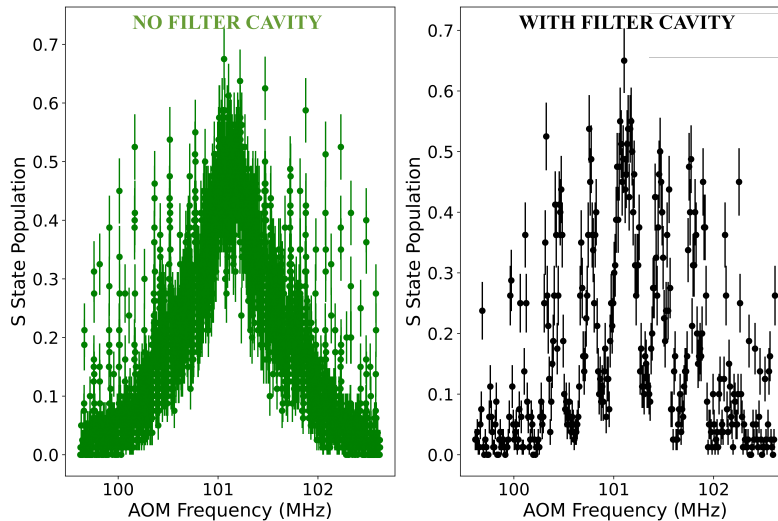


Figure 5.27: Demonstrating the need for the filter cavity to remove servobumps, high power 729nm light was applied to the ion with a wide scan of the AOM frequency to include multiple motional sidebands. Shown in the left figure, we see that the sidebands are not resolvable at high power without the filter cavity. Once the filter cavity was integrated into the system, the high power laser scan is now able to resolve the motional sidebands.

it. Preamplified light then exits the dye cell and goes through the amplifier dye cell, which has also been pumped with 355nm light. With fresh dye cells, we can generate around 20mJ of 486nm light. Each dye cell has a different concentration of dye to optimize the generation of 486nm; dye cells need to have the dyes replaced after a couple months of daily usage as it begins to degrade and the power drops. The 486nm light then goes through a doubling crystal to generate the 243nm light. A dichroic mirror picks off the 486nm light and sends it to a wavemeter via a multimode fiber. The 243nm light is then sent out of the Sirah enclosure and towards the chamber. Since the dye laser is on a different optical table, it needs several mirrors to level the heights, as well as a pair of lenses to reduce the beam waist and collimate the light.

5.4.6 Laser Pulse Sequences

Control of the pulse sequences is done using ARTIQ's DDS and TTL outputs. For each AOM, a DDS signal is applied which introduces the first order diffracted beam, and a TTL pulse goes to a switch that powers the AOM RF amplifiers. While

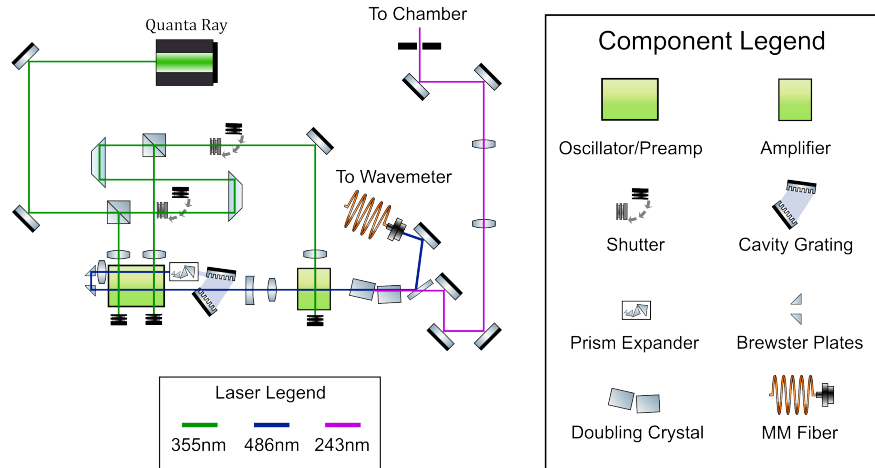


Figure 5.28: Dye laser system from Spectra Physics/Sirah for generating pulse light at 243nm for photoionization of HCl^+ . See text or Sirah manual for additional description of optical components.

the TTL may seem redundant, switching off the amp prevents stray signals from being amplified and applied to the AOM; we have seen these as an experimental necessity. Additional RF pulse sequences either require DDS channels or phaser channels (and TTL's for switching amps). The cooling sequence will be discussed here for illustrative purposes.

Cooling the ion chain is required on nearly every experiment. Initial cooling is achieved using Doppler cooling with 397nm, 866nm, and 854nm light. Doppler cooling time is optimized based off of several carrier Rabi oscillations. Sideband cooling is next applied, interleaved with spin polarized 397nm light to optically pump into the correct Zeeman state. Cooling all modes in the ion chain results in the best performance, since spectator mode heating is reduced. Each mode is addressed for a percentage of the pulse duration, with the 854nm quench beam adjusted to compensate for the different Lamb-Dicke parameters of each mode. For example, we typically apply sideband cooling for 5-10ms depending on whether we are addressing a single or multiple ions, with every $300\mu\text{s}$ a spin-polarization pulse being applied for $20\mu\text{s}$ to pump all the $S_{\frac{1}{2}}$ population into $m_J = -\frac{1}{2}$. Typically we use 11 cycles of sideband cooling during the pulse duration. Assuming we want to operate on the Axial mode with a single Ca^+ , the first 30% of a single sideband cooling cycle is spent cooling the EGGS mode, the next 30% is spent on the RF mode, and finally

40% on the Axial mode. After all the cycles have been completed, we apply a polishing pulse, which is a shorter, lower power pulse on solely the mode of interest. High power cooling can leave some residual phonon number from accidentally driving carrier oscillations; the low power pulse can clear out the remaining phonon number without driving the carrier. We typically only use $150\mu s$ for the polish pulse.

5.5 Imaging

To image the ions, a reentrant vacuum viewport from MPF is used to increase the maximum available numerical aperture (NA). The viewport is DUV grade fused silica with AR coating for 397nm light on both surfaces ($R < 0.25\%$). The reentrant viewport goes 1.277" into the chamber, since the working distance, the distance between the ion and the lens system, is 55.7mm. A custom objective lens from Silloptics (S6ASS2241) is used to reduce chromatic and spherical aberrations at 397nm and 729nm, while providing a moderate NA (0.3). Imaging was also achieved using a 2" achromat lens from Thorlabs (AC508-075-A); the custom lens system is intended for having the two different wavelengths of light focused the same, whereas the achromat will have different focusing at different wavelengths. The objective lens is mounted onto an adjustable lens tube for coarse z alignments, before using retaining rings to lock its position. It is then connected to an XY translator (CXY2A) for fine adjustment in the other two axes; coarse adjustment is done with positioning the lens system on the breadboard. The entire system is on a 1-D translation stage to allow for fine adjustment of the z-axis. Two different designs were implemented in this experiment. The initial design has the XY translation stage mounted on one side to cage mounting brackets (LCP33B) that are bolted to a custom image block with six 8 screws. Also attached to the block is the z-axis translation stage and 45° mount (H45E2) for the 2" broadband dielectric elliptical mirror (BBE2-E02), that sends the light through the rest of the imaging system. This original system was replaced to improve the stability of the imaging and to provide a finer degree of control for alignment.

In the second rendition, the XY translation stage is mounted on all 4 sides using the cage mount rods, as well as with a 2" lens tube, which connects to a right-angle

kinematic 2" mirror case mount with smooth bores for the cage rods (KCB2C). On either side of the mount, it is connected to a custom block with a set of cage brackets in addition to the tapped holes on the mirror mount. Both custom mounts are then connected using a third custom piece which connects to the z-axis translation stage. This stage is then mounted onto an additional 2-D translation stage for a greater range of x and y tunability. This stage is mounted to four 1" posts, each bolted to the breadboard using optic forks. Additional stability is provided by 2" lens tubes that are mounted to the other side of the adjustable 2" mirror mount. A slip ring (SM2RC) connects to the tube and is constrained to the breadboard using an adjustable post and fork (not shown in Fig. 5.29). Switching to the new system reduced the ion drift we would see on the camera from roughly 20 pixels a day to roughly 4 pixels a day (1 pixel $\approx 1\mu m$).

The light is collimated as it exits the objective lens, and needs to be focused onto the camera. The focusing lens, referred to as the tube lens, plays an important role in the magnification of the ion on the camera based off of

$$M = \frac{f_t}{f_o} \quad (5.16)$$

where f_t is the focal length of the tube lens and f_o is the focal length of the objective lens (note that the focal length of the objective is different than the working distance). The focal length of the objective lens is 66.9mm, and we used two different tube lenses. For rough alignment in the beginning a 2" achromat (ACT508-200-A) with focal length of 200mm was used, which allowed us to image the entirety of the trap. This was used in the beginning when we initially started loading ions to ensure that we could optimize the loading and trapping parameters so that we could confidently have a cloud of ions as we switched to the higher focal length lens. Since then, the tube lens (ACT508-1000-A) that we use has a focal length of 1000mm, which was the longest available 2" achromat that we could find sold off the shelf. The tube lens is mounted in another XY translation mount (CXY2A), which uses a lens tube and slip ring to mount it onto the breadboard. The XY translator is designed for cage systems, so two of the four holes have cage rods that connect it to the 45° mirror mount that the objective lens is connected to. The other two cage holes have rods that connect to the 45° kinematic mount (KCB2) that holds a circular E02 coated

dielectric mirror, the first mirror in a periscope that takes the light from the imaging breadboard onto the optics table to the camera and PMT. On the other side of the periscope is another 45° kinematic mount with the same mirror, which is constrained to the other mount in the periscope using a cage rod and using 2" lens tubes. The lens tubes use a lens tube flexure sleeve coupler (SM2CPL10) to unconstrain them with ease. To limit stray light, the mirrors have black out tape on their backs. The circular mirrors will have a slightly reduced NA compared to an elliptical mirror, but since they are after the tube lens, the light has already started being focused down, so that the smaller NA of the mirrors does not reduce the light collection.

As the light exits the periscope, it has about 16" of travel left until it is at the focus. About 5.5" in, a 1" flipper mirror is introduced to allow for quick and easy alternating between the camera and the PMT. When the mirror is down, the light travels straight. The camera has lens tube to prevent background light from entering and has a 400nm bandpass filter (Edmund Optics 65-193) with 10nm FWHM to only allow the 397nm light from the ions to reach the camera. When the mirror is up, the light gets reflected towards the PMT. After the flipper mirror, the light goes through a pair of kinematic cage mirror mounts connected with lens tube to prevent extra light from entering. After the aligning mirrors are two variable slits (VA100CP) that serve as spatial shutters to reduce the counts from the laser light scattering off the trap blades. Then there is a 400nm bandpass filter (FBH400-10) for the light going to the PMT. The PMT (H11890-210) is mounted to a lens tube using a custom 3D printed piece that limits any light from leaking in. All the components after the periscope, except for the camera itself, are enclosed in a box made with blackout board.

The camera used in this experiment is an Andor iXon3, a camera inherited from the previous velocity map imaging experiment in the Hudson lab. It is mounted on a 1D translation stage to move it into the focus of the ion light, with rough alignment done simply by moving the camera until the trap center was imaged. If used as a photon counter, the camera generates photoelectrons given by

$$Photoelectrons = (Counts - Background) \frac{Sensitivity}{EMGain} = Photons \times QE \quad (5.17)$$

where the sensitivity of our camera is 22.68, the EM gain is 200 at 100ms, and the

quantum efficiency (QE) at 397nm is 55%. We can use this estimate to compare to the photon collection of the PMT, however in this experiment the camera's sole purpose is to monitor the ion loading and position when a chain is loaded. All the data for processing is taken using a Hamamatsu photomultiplier tube (PMT) photon counting head (H11890-210).

Several campaigns have been made to optimize the imaging alignment. The initial alignment was done with the first imaging system by aligning the ions onto the camera, and about 45k counts/second per ion were collected. This attempt was done to confirm ions were being trapped and was suitable for accomplishing that goal. However, we estimate that with our given NA, transmission losses, and quantum efficiency, that we should be able to get higher. Once we switched to the second imaging system, we also tried to improve the fluorescence through the periscope, by using an iris after different mirrors to optimize them one step at a time using the PMT. The counts increased to between 50k and 90k per second on the camera per ion, although the higher count rate resulted in higher coma aberrations, which were filtered by the spatial shutter before the PMT. Our typical rate is about 55k counts/second per ion.

5.6 Magnetic Field

A stable magnetic field is important to avoid a drifting Zeeman shift in the internal states of our ions [Roo00]. Magnetic fields are produced either through two arrays of permanent magnets, or with a pair of magnetic field coils set up close to the Helmholtz configuration and powered by a low noise DC supply. The former was chosen in our experiment for two dominant reasons: to generate a large enough field, a several amp current would need to be supplied, and a magnetic field generated by a pair of solenoids is susceptible to any noise on the source of the current going through them. A set of permanent magnets seemed to be a simpler and cheaper option.

The permanent magnetic field is generated by using 16 Samarium Cobalt magnets that are inserted into a 3-D printed PLA holder. These magnets, their positioning,

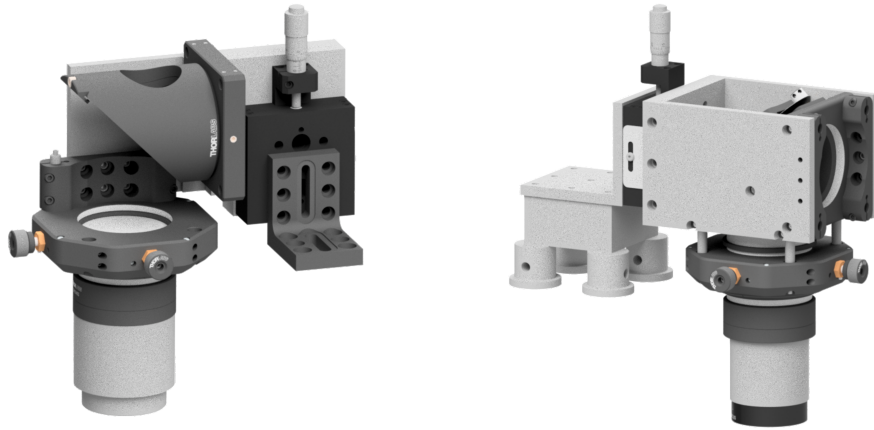


Figure 5.29: Different imaging systems used during this PhD. The first version (a) has the objective lens adjustable horizontally using an XY translator for a 60mm Cage system, and vertically using a single translation stage. All parts are mounted onto a block that I machined. An upgraded version (b) was redesigned to have greater stability, less drift, and additional tunability. By being symmetric, having a larger base, and cage mounting tubes as additional support, this design saw ion drift on the camera reduced dramatically (20 pixel drift a day to 4 pixel drift a day; 1 pixel $\approx 1\mu m$). The additional adjustable mirror facilitated alignment and enabled improved fluorescence collection.

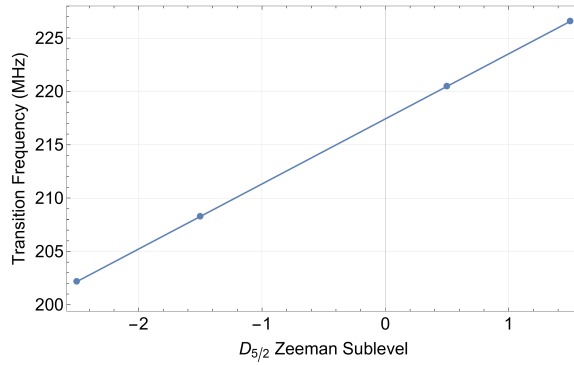


Figure 5.30: Rough measurements of the magnetic field were made before they were attached to the vacuum chamber using a gaussmeter. However to get an accurate measurement at the ions, we use the ions. Starting in the $S_{\frac{1}{2}}$ state, we scanned the 729nm AOM to locate the addressable $D_{\frac{5}{2}}$ states. A linear fit determined the magnetic field to be 3.6485G.

and the quantity were chosen in such a way as to generate ~ 3.5 G. Measuring with the atomic states of a calcium ion, we see that we generate a 3.6485G field at the center of our trap from Fig. 5.30. The holder is designed to fit snug over a 2.75" CF viewport (with bolts) but for extra stability, four holes every 90° were tapped into the piece for 4-40 screws. 3-D printing was chosen as a cost effective and non-magnetic option, but tapped threading in plastic is not the most robust. Screwed onto the permanent magnet holder is another 3-D printed holder, this time for magnetic coils for adjusting an additional field for either compensation or for tuning to a higher or lower strength field than the permanent magnets provide. These coils have never been used and have a 50Ω load connecting both ends of the wiring.

For compensation of stray fields in the other directions we implemented similar pairs of coils. As before, the holders are made from 3-D printed PLA. For the vertical coils, the dimensions were too large to print a single coil holder on the printer's 12" by 12" bed, so it was cut in half and reattached with screws. The vertical coils were made using 18AWG wire with a polyester coating with about 200 turns on each coil. It is rated for 16 amps for chassis wiring and 2.3 amps for power transmission. After the wire was wound, the outer layer was coated with stycast to glue the wires together. To minimize the stray magnetic field in the vertical direction, 700mA was provided using a GW Instek power supply.

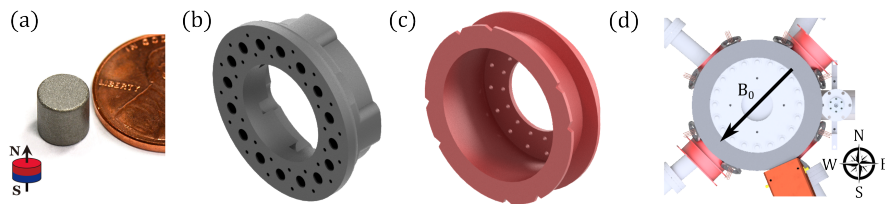


Figure 5.31: Our magnetic field comes from permanent magnets, with compensation done using coils. (a) Samarium cobalt magnets from CMS magnets were physically constrained within a 3-D printed holder (b). Over the magnets was a 3-D printed coil holder, that further ensures the magnets can't slip out of the holder.

In the final direction, another pair of compensation coils are used. A similar gauge wire was used with a polyimide coating; for these horizontal compensation coils, about 110 turns were used each and a current of 30mA is provided using a Keithley power supply. The unused pair of coils has a similar number of turns as these coils.

5.7 RF/Microwaves

A large focus of this experiment has been devoted to understanding the RF and microwave fields that we can apply to our ions. While the RF voltage for the ion confinement is generated using a Rohde and Schwarz function generator (later replaced by an Agilent), the rest of the RF and microwave tones are produced using ARTIQ (Advanced Real-Time Infrastructure for Quantum physics). Most of the RF components are from Minicircuits, although we have several custom-built components from Peter Yu in our department's electronic laboratory. Within our experiment, we lovingly refer to them as Peter Boxes.

5.7.1 ARTIQ Hardware

The ARTIQ system has been adopted by many groups in the trapped ion field, in large part due to its versatility, customization, and well-developed open source hardware and software database. Each ARTIQ crate consists of a rack-mountable enclosure and a Kasli device, which consists of a field programmable gate array

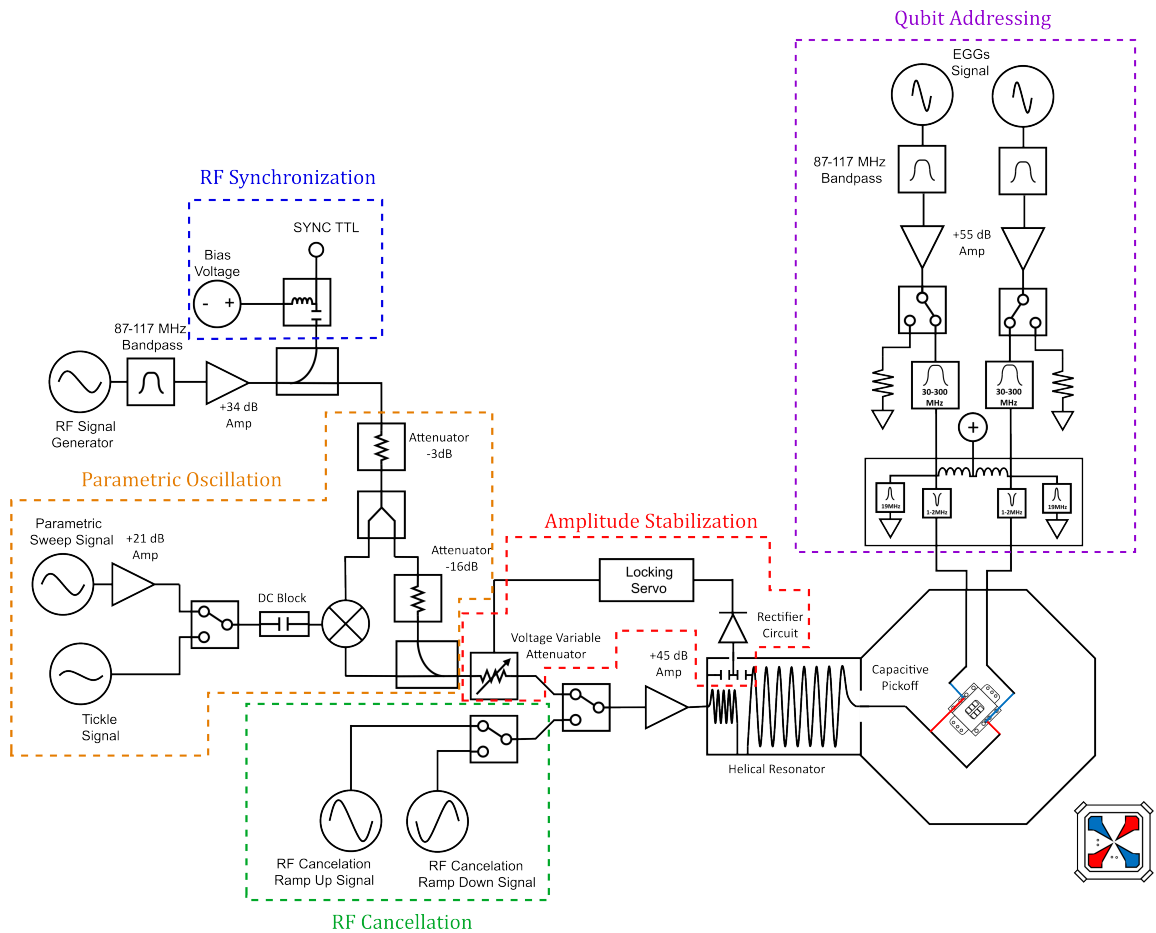


Figure 5.32: A somewhat outdated version of the RF board to demonstrate each RF system that was combined for this experiment. The qubit addressing system uses the ARTIQ AWG to produce signals that are filtered and amplified before being applied to the EGGs electrodes. Individual electrode control for RF/microwave signals requires both AWG channels to be in use, and a custom bias tee allows DC to be applied to each electrode. On the RF electrodes, we implemented an active trap stabilization system, a parametric board for shimming, RF synchronization, and an RF cancellation system to quickly reduce the trap field for loading. In the end, the locking and parametric control were moved to their own board, and the RF cancellation and synchronization were removed from lack of use.

(FPGA), RAM, and control capabilities for up to 12 daughter-boards. From there, different daughterboards are available for digital input-output, digital-to-analogue conversion, analogue-to-digital conversion, RF signal generation, and more. In our experiment we have three direct digital synthesizer (DDS) boards referred to as Urukul boards. Each Urukul has four DDS outputs, which we nominally use for controlling the laser AOM's as well as for applying parametric/tickle tones. For digital IO, the crate includes two 8-channel TTL boards. One of the boards is dedicated to input signals from the PMT, the line trigger, and the trap RF clock, with available inputs for other applications that arise. The other board is the TTL output board. One of the channels is used to initiate an integrator hold on our RF amplitude servo to halt feedback to keep the output constant, and the rest are sent to the amps we use for the AOM's and RF signals. Our spectroscopic search of the A -doublets of HCl^+ requires applying tones around 80MHz as well as the motional sidebands about this frequency. This operation is done using the ARTIQ phaser, a two output arbitrary waveform generator shown in Fig. 5.33. The tones for studying motional Raman are also generated using this device. We have two phasers in our experiment, one baseband variant and one which includes an upconverter to address higher frequencies. The final boards that our crate holds are not used on a regular basis. One is a low-noise clock distribution system. Next is the Fastino, ARTIQ's DAC board. Last is the Sampler, the ADC which is unused during typical operation but is needed for occasional readout of other devices for calibration or long-term measurement.

5.7.2 Peter Box

As briefly mentioned, there have been several cases during my PhD that we've required custom electronics, provided by Peter Yu in our department's electronic lab. In our experiment the complex devices he's designed and built include a laser current controller, a temperature controller, and a high voltage DC box (mentioned earlier). This section is more focused on various smaller devices he's built for our experiment and their application. The most important of these are the boxes he's built us for applying our different voltages onto the trap EGGS electrodes. Two different

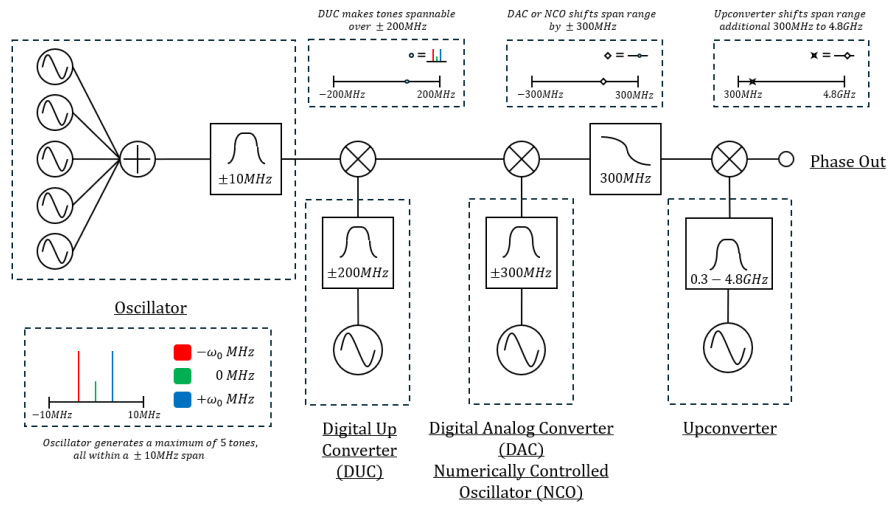


Figure 5.33: Each channel of the ARTIQ Phaser consists of five local oscillators, which must all be within $\pm 10\text{MHz}$. The oscillator tones then go through a digital up converter to make them scannable over $\pm 200\text{MHz}$, which is then shifted by a numerically controlled oscillator, which can shift the range by $\pm 300\text{MHz}$. Our models have a 300MHz low-pass filter that limits the range of the baseband AWG. The upconverter model, can further shift the span range up to 4.8GHz . Through the software, the frequency, phase, and amplitude of each oscillator can be set, allowing for versatility

versions have been developed for this task: each box operates as a filtered bias tee with two RF inputs, a single DC input, and two outputs. Special consideration is given to ensure that any RF trapping signal that is being picked up onto the EGGS electrodes is being properly filtered. If the electrodes are left floated, then the trapping voltages get picked up onto these electrodes with different voltages and phases, which results in the ion becoming unhappy. Unhappy ions have high heating rates and have trouble remaining trapped; for new experiments this can often be a leading challenge related to the inability to load. As such, these bias tees are designed to have notch filters for 19MHz (the trapping frequency) at each output. The rest of the components are for a more standard bias tee. A second version was built with the goal of including an 82MHz resonator for reducing insertion loss onto the electrodes for our HCl^+ spectroscopy. A couple transformer components were included, but the insertion loss was actually slightly more in this new box in the region of interest. The electronics are housed in boxes that I machined to connect directly onto the chamber's filter boards.

5.7.3 Revised Parametric and Locking Board

As mentioned earlier in section 5.3.4, the trap was originally locked using a capacitive divider scheme discussed in [K 16], whereby a 0.2pF and 20pF pair of capacitors picked off 1% of the output voltage of the resonator. While this particular method was eventually scrapped due to the noise picked up on the long coaxial cable used, locking electronics were merged with the parametric oscillation components to form a single board for all the operations that we apply to the RF electrodes. Typical operation involves consistently applying a voltage to trap the ions without introducing noise from the ARTIQ DDS (used for parametric oscillation) so the switch after the parametric amplifier is switched to the terminator. The Agilent signal generator outputs the 19MHz signal that goes through the Squareatron-5000, an ultra-low noise limiting amplifier. We'd like to thank Jeremy Metzner from Dave Allcock's research group for assembling the device for us. After the Squareatron, the signal goes through a voltage variable attenuator before being amplified. A coupler picks off a small part of the amplified signal prior to the resonator to lock with, going

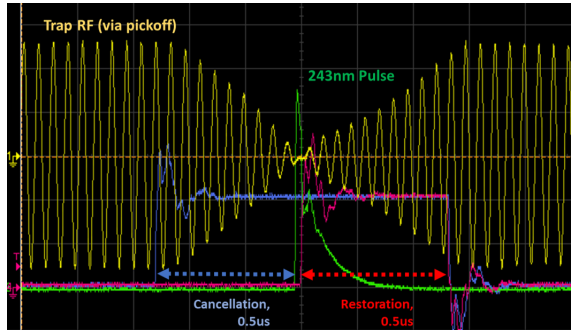


Figure 5.35: To demonstrate the viability of canceling the trapping RF during HCl^+ loading, the voltages are monitored on an oscilloscope. The yellow curve shows the trapping voltage signal get reduced by the cancellation tone (blue), before ramping back up (pink). The RF voltage is minimized when the 243nm laser pulse goes through the trap, shown here in green. The ramp down and ramp up pulses, have a larger amplitude in the beginning for fast response.

the RF trapping field can result in AC Stark shifts of the molecular states [Kel20], thereby moving the transitions needed by REMPI. The frequency measured in one ion trap may shift when trying to load that same molecule in a different ion trap, as was the case in this experiment, see section 5.2.2. A procedure for rapidly reducing the trapping voltage during the loading of HCl^+ was implemented from [Kel20], which can keep the Ca^+ stably confined during the reduction. While this procedure was integrated into our experiment, the frequency shift from the trapping field would be very small and be within ionization laser linewidth. Additionally, since the ionization laser is quite broad, the ro-vibrational state selection via REMPI is not viable. This detour did motivate another check of the resonator's performance via a ringdown time measurement, which has similar results to the measurement of the Q factor on a network analyzer.

CHAPTER 6

Electric-field Gradient Gates

In this chapter, I will cover the proposal that this experiment was conceived to implement, as well as the progress that was made towards achieving the goal. The original motivation behind this experiment was to create a system to implement laser-free molecular quantum logic [HC21]. Atomic ions have been a leading platform for quantum computing, with many systems driving transitions between different hyperfine states using high-power lasers. For atoms, transitions between hyperfine states can only be driven with magnetic dipoles or with Raman lasers. However, as we covered in section 4.7.0.1, molecules can have electric dipole transitions between hyperfine states if they are connected with different parity states. Furthermore, these hyperfine states fall within a convenient regime of the energy spectrum, the RF to microwave spectrum. That means the strong E1 transitions can be driven without the use of lasers, and since these states are on the order of MHz to GHz separated, they will have long spontaneous decay lifetimes (Fermi's golden rule). An additional motivation for molecules stems from their prolific energy structure. A more complex energy structure adds additional challenges, but offers greater potential for novel schemes, and if the transitions are drivable with off-the-shelf electronics, then the challenges become less daunting.

6.1 Molecular Quantum Logic Spectroscopy

As mentioned briefly in section 4.9, the motion of a trapped molecule is coupled directly to the motion of a co-trapped atom, within the same ion trap. By co-trapping with an atomic ion, the molecule can be cooled to its ground motional state without having to try driving transitions within the molecule. Since the ion is also within a cryogenic environment, it will thermalize to its environment, reducing to its ground

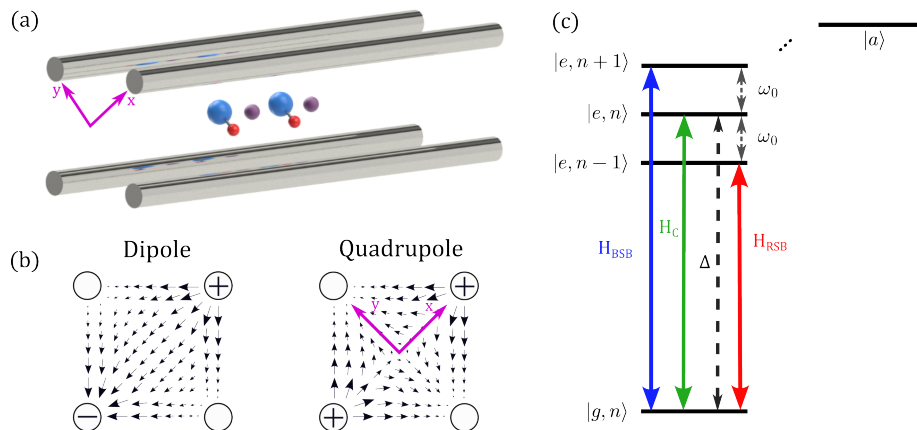


Figure 6.1: A schematic of the basic system for electric-field gradient gates (EGGs). Molecular ions are co-trapped with atomic ions for sympathetic cooling and state readout shown in (a). For illustrative purposes the blades are portrayed as rods. Electric fields are applied directly onto the trap electrodes (b), either in a dipole or a quadrupole configuration. The electric field in the dipole is constant, whereas in the quadrupole it is zero, but has a linear gradient. Two dipole addressable states $|g\rangle$ and $|e\rangle$ are separated in energy by Δ , with a third auxiliary state $|a\rangle$, shown in (c). Motional sidebands are produced at the ion trap's secular frequency ω_0 .

ro-vibrational state. At this point, the molecular state will only be characterized by its hyperfine state and parity state. If we can introduce an interaction that couples the motion of the molecule to its internal spin, then that change in motion can be read out by the atom. This method is frequently referred to as quantum logic spectroscopy [PW05] and has been implemented in other molecule/atom ion systems [Cho17, Fab16, D 25].

6.2 EGGs: The Theory

Electric-field gradient gates (EGGs) are produced by applying an oscillating electric field to our trap blades, with two important orientations being referred to as the dipole configuration and the quadrupole configuration, as shown in Fig. 6.1. The dipole configuration will produce an electric field at the i^{th} molecule in the trap that will take the form

$$\mathbf{E}_{E1}^{(i)} \cdot \hat{x}_j = -\frac{V_m \cos(\omega_m t)}{2r_0} \quad (6.1)$$

where the subscript j indicates a given motional mode. Operating in the quadrupole configuration will produce an electric field of the form

$$\mathbf{E}_{E2}^{(i)} \cdot \hat{x}_j = -\frac{2V_m \hat{x}_j \cos(\omega_m t)}{r_0^2}, \quad (6.2)$$

where we will write the position operator as some equilibrium offset and the displacement that stems from the normal modes,

$$\hat{x}_j^{(i)} = x_{eq}^{(i)} + \sum_j \sqrt{\frac{\hbar}{2m\omega_j}} b_j^{(i)} (\hat{a}_j + \hat{a}_j^\dagger). \quad (6.3)$$

Our experiment focuses on a trapped molecule with an electronic dipole moment $\mathbf{d}$ and an energy splitting of Δ . The Hamiltonian for a dipole in an electric field is $H_m = -\mathbf{d} \cdot \mathbf{E}_m(\mathbf{r}, t)$. The total Hamiltonian of our single ion system is written out as

$$\frac{H_{total}^{(i)}}{\hbar} = \sum_j \omega_j (\hat{a}^\dagger \hat{a} + \frac{1}{2}) + \frac{\Delta}{2} \sigma_z - \frac{\mathbf{d} \cdot \mathbf{E}_m(\mathbf{r}, t)}{\hbar}. \quad (6.4)$$

Moving our total Hamiltonian into the interaction picture with respect to the molecular Hamiltonian and the harmonic oscillator Hamiltonian requires the time evolution operator, $U(t) = E^{-i(\sum_j \omega_j (\hat{a}^\dagger \hat{a} + \frac{1}{2}) + \frac{\Delta}{2} \sigma_z)t}$, be applied to our total Hamiltonian. As a result the Hamiltonian reduces to the following

$$\frac{H_I^{(i)}}{\hbar} = E^{i(\sum_j \omega_j (\hat{a}^\dagger \hat{a} + \frac{1}{2}))t} E^{i(\frac{\Delta}{2} \sigma_z)t} \left(-\frac{\mathbf{d} \cdot \mathbf{E}_m(\mathbf{r}, t)}{\hbar} \right) E^{-i(\frac{\Delta}{2} \sigma_z)t} E^{-i(\sum_j \omega_j (\hat{a}^\dagger \hat{a} + \frac{1}{2}))t}, \quad (6.5)$$

which looks intimidating until we introduce our electric field terms. Recall that the dipole is an operator defined by

$$\mathbf{d} = \langle g | \mathbf{d} | e \rangle |g\rangle \langle e| + \langle e | \mathbf{d} | g \rangle |e\rangle \langle g| = \langle g | \mathbf{d} | e \rangle (\sigma_- + \sigma_+) = d\sigma_x \quad (6.6)$$

which indicates that for any electric field along the dipole direction, we will have the molecular component of the time evolution operator change our Hamiltonian. Through a few lines of algebra we can show that this transforms σ_x into $(E^{i\Delta t} \sigma_+ + E^{-i\Delta t} \sigma_-)$. In the case of the dipole configuration, the harmonic oscillator part of the time evolution operator commutes with the time-dependent Hamiltonian. The dipole Hamiltonian in the interaction picture is

$$\frac{H_{E1}^{(i)}}{\hbar} = \frac{\Omega}{2} ((E^{i(\Delta+\omega_m)t} + E^{i(\Delta-\omega_m)t}) \sigma_+^{(i)} + h.c) \quad (6.7)$$

where $\Omega = \frac{dV_m}{2r_o\hbar}$. For molecules with large splittings, we can ignore the first exponential as part of the rotating wave approximation. We will avoid doing so, to understand how our choice of molecule may affect our outcomes. Since this Hamiltonian is independent of the raising and lowering operators for the phonon number, we can tell that it drives carrier transitions only. The quadrupole configuration has a component, $\sum_j \sqrt{\frac{\hbar}{2m\omega_j}} b_j^{(i)} (\hat{a}_j + \hat{a}_j^\dagger)$, which will be transformed by the harmonic oscillator portion of the time evolution operator. The offset will not transform and will solely drive carrier transitions. The value $(\hat{a}_j + \hat{a}_j^\dagger)$ will transform to $\hat{a}_j E^{-i\omega_j t} + \hat{a}_j^\dagger E^{i\omega_j t}$. These will drive only sideband transitions. Our total Hamiltonian for the single ion quadrupole configuration is

$$\begin{aligned} \frac{H_{E2}^{(i)}}{\hbar} = \sum_j \frac{2\Omega}{r_o} \sqrt{\frac{\hbar}{2m\omega_j^{(i)}}} b(\hat{a} + \hat{a}^\dagger) (\sigma_+^{(i)} (E^{i(\Delta+\omega_m)t} + E^{i(\Delta-\omega_m)t}) + h.c) \\ + \frac{2\Omega x_{eq}^{(i)}}{r_o} ((E^{i(\Delta+\omega_m)t} + E^{i(\Delta-\omega_m)t}) \sigma_+^{(i)} + h.c) \end{aligned} \quad (6.8)$$

which includes both sideband and carrier transitions.

For practical purposes we want to consider the case where we apply simultaneous electric fields in the quadrupole configuration close to the first-order motional sidebands: $\omega_m = \omega_\pm = \Delta \pm (\omega_j + \gamma)$. Shimming the ion to the trap null will result in $x_{eq} = 0$, and defining the constant

$$\eta_j^{(i)} = \sqrt{\frac{\hbar}{2m\omega_j r_o^2}} b_j^{(i)} \quad (6.9)$$

allows Eqn.6.8 to be simplified as

$$\begin{aligned} \frac{H_{E2}}{\hbar} = 2\Omega \eta_j^{(i)} \sigma_x^{(i)} (e^{-i\gamma t} \hat{a}^\dagger + e^{i\gamma t} \hat{a}) \\ + 2\Omega \eta_j^{(i)} (\sigma_+^{(i)} \hat{a} (e^{i(2\Delta+\gamma)t} + e^{i(2\Delta-2\omega_0-\gamma)t} + e^{-i(2\omega_0+\gamma)t}) \\ + \sigma_+^{(i)} \hat{a}^\dagger (e^{i(2\Delta-\gamma)t} + e^{i(2\Delta+2\omega_0+\gamma)t} + e^{i(2\omega_0+\gamma)t}) + h.c). \end{aligned} \quad (6.10)$$

For convenience, the RWA will be applied at this step, removing all time dependent terms that oscillate at frequencies greater than $\gamma \ll \omega_0 \ll \Delta$, dramatically simplifying the Hamiltonian to

$$\frac{H_{E2}^{(i)}}{\hbar} = 2\Omega \eta_j^{(i)} (\sigma_+^{(i)} + \sigma_-^{(i)}) (e^{-i\gamma t} \hat{a}^\dagger + e^{i\gamma t} \hat{a}) = 2\Omega \eta_j^{(i)} \sigma_x^{(i)} (e^{-i\gamma t} \hat{a}^\dagger + e^{i\gamma t} \hat{a}). \quad (6.11)$$

A quantum non-demolition measurement will enable us to determine whether or not our ion is in the qubit subspace. We can accomplish this using our near-resonant

sideband tones in the quadrupole configuration. By applying these electric fields, we can increase the average phonon number, which will cause a measurable oscillation of our ion. Taking $\gamma \rightarrow 0$ and using Eqn. 6.11, we see that the time evolution operation will be given by a displacement

$$\begin{aligned}
 U_{E2}(t) &= e^{-2i\Omega\eta_j^{(i)}\sigma_x^{(i)}t(\hat{a}+\hat{a}^\dagger)} \\
 &= \hat{D}(2i\Omega\eta_j^{(i)}t)|-X^{(i)}\rangle\langle -X^{(i)}| + \hat{D}(-2i\Omega\eta_j^{(i)}t)|+X^{(i)}\rangle\langle +X^{(i)}|
 \end{aligned} \tag{6.12}$$

where the states $|\pm X^{(i)}\rangle = \frac{1}{\sqrt{2}}(|g^{(i)}\rangle \pm |e^{(i)}\rangle)$ are the eigenvectors of the $\sigma_x^{(i)}$ operator. Regardless of whether the molecule is in the ground or the excited state, as long as it is in that manifold, applying this operation will result in a displacement of the ion chain along the given mode j . The excitation generated by the state dependent force on the molecule will increase the energy in mode j by $\Delta E_j = \hbar\omega_j(2\Omega\eta_j^{(i)}t)^2$. Since the modes are shared by both the atomic and molecular ions, this increase in phonon number can easily be read out using the atomic ion.

6.3 EGGs: The Practice

There have been many challenges enacting this theory into practice. While we have estimates of what the energy levels of the molecule are based off of measured rotational constants of H^{35}Cl^+ , the exact energy levels for the different states have never been measured. To perform spectroscopy on the molecule, we use the same Hamiltonian as the two-qubit EGGs gate, a novel and previously untested method of driving a displacement. Additionally, there are 64 different states within the ground ro-vibrational state of the molecule, which are populated at random, so when we load a molecule, it can be within any of these states. For this reason, we would focus on the largest hyperfine $\mathbf{F}$ states, since they will have the greatest probability of being populated.

6.3.1 HCl^+ Sourcery

Loading Ca^+ is a simple task compared to loading our molecule. To load HCl^+ requires leaking low pressure gas ($\sim 10^{-6}\text{mBar}$) into our chamber and driving a 2+1 REMPI ionization transition within the molecule. Preliminary tests were carried out

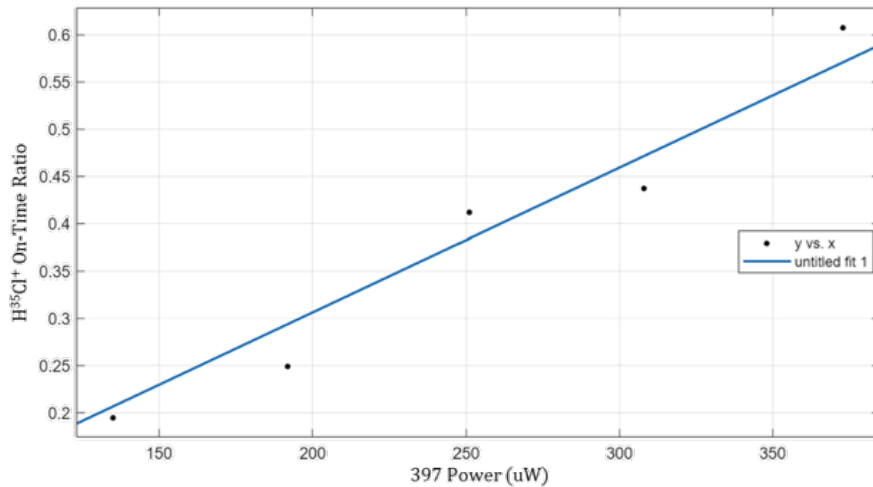


Figure 6.2: $\text{H}_2^{35}\text{Cl}^+$ Photodissociation Rate

prior to the design and construction of this experiment in the MOTION chamber by Hao. In that experiment he determined what pressures, pulse energies, and laser frequency were needed to load HCl^+ molecules within a cloud of Ca^+ . Mass confirmation was done using a time-of-flight mass spectrometer connected to the ion trap. Our loading gas was nitrogen with 40 ppm HCl . As mentioned in section 5.2.2, this gas would frequently result in other masses being loaded. Once we switched balance gases, we saw higher loading rates of HCl^+ . However, there are two isotopes of chlorine ^{35}Cl and ^{37}Cl . Estimates of the Λ -doublet transitions were made using constants for the former isotope. The latter did not have all the values measured/estimated, but would be a better choice given the mass ratio with calcium.

Reactions with H_2 to form H_2Cl^+ were observed in the MOTION chamber which also motivated our choice to be cryogenic. However, reactions are still relatively commonplace even in the best conditions. After a fresh cooldown, we can expect a couple hours without a reaction. Luckily, we discovered that with the $\text{H}_2^{35}\text{Cl}^+$ isotopologue, it would return to H^{35}Cl^+ shortly ($\sim\text{s}$) after letting high-power 397nm light through the trap. While we have not taken enough data to confirm, the dissociation data in Fig. 6.2 seems to indicate a linear relation and hence a 1+1 REMPD. However, the other isotopologue $\text{H}_2^{37}\text{Cl}^+$ would remain unchanged even with the 397nm light. An additional challenge we faced was that we seemed to load $\text{H}_2^{37}\text{Cl}^+$ more frequently than we'd expected based on chlorine isotope abundance (1:3 for ^{37}Cl compared to

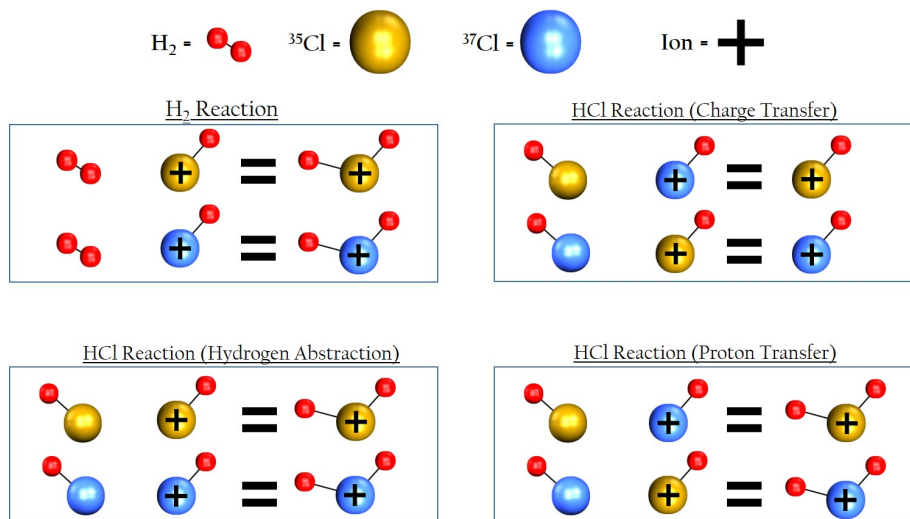


Figure 6.3: HCl⁺ Reaction Channels

³⁵Cl). Based on the results in [LKK21, Cha10], we believe that during the loading process H³⁵Cl⁺ or H³⁷Cl⁺ can also be reacting with another HCl molecule to form H₂Cl⁺ or to swap charge. HCl reactions, shown in Fig.6.3, can be characterized as hydrogen abstraction, whereby the hydrogen atom is removed from the colliding HCl molecule, as charge transfer between the two molecules, or as proton transfer, where the hydrogen nucleus on the ion is moved to the neutral molecule resulting in Cl and H₂Cl⁺. Combined with the photodissociation from 397nm light on H₂³⁵Cl⁺, this indicated that despite the lower isotopic abundance, H₂³⁷Cl⁺ was preferentially being loaded since H₂³⁵Cl⁺ could return to H³⁵Cl⁺ and eventually react to H³⁷Cl⁺ or H₂³⁷Cl⁺, whereas H₂³⁷Cl⁺ would be removed from the reaction pool.

To reduce loading H₂³⁷Cl⁺, we implemented the procedure shown in Fig. 6.4. During this procedure, the 397nm power is reduced to limit photodissociation of H₂³⁵Cl⁺ while the HCl leak is active. Once a dark ion is loaded, the 243nm light ($\sim 60\mu\text{J}$) is blocked and the leak valve is closed. The 397nm is still kept at low-power, as the neutral gas has time to freeze out. A couple minutes later, a high-power parametric voltage is applied to the trap electrodes to eject any undesired ions in orbit that had not crystallized. We assume that since these ions have not crystallized, they will be excited by frequencies corresponding to a trap empty of other massed particles. After a couple minutes of sweeping the ejection frequency, we assume only the crystallized ions remain and turn up the 397nm power to dissociate H₂³⁵Cl⁺ if it

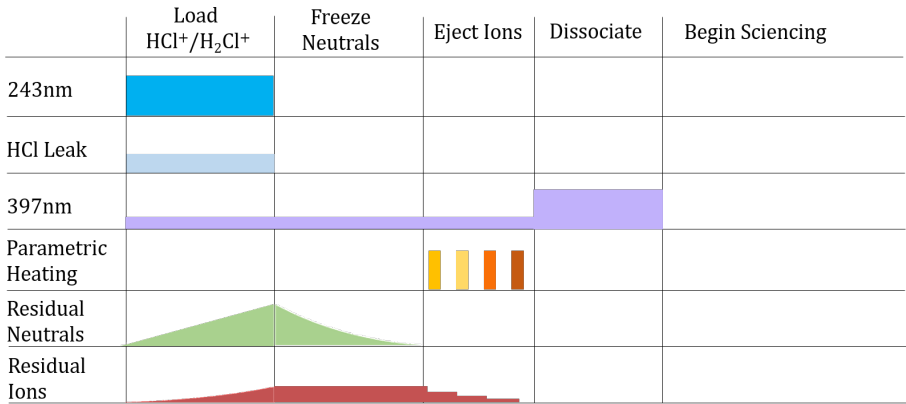


Figure 6.4: Procedure for loading HCl^+ starts with low power 397nm light to avoid photodissociating any $\text{H}_2^{35}\text{Cl}^+$ that is crystallizing. Once the ion has crystallized, the gas and 243nm light are turned off so that residual neutral gas can freeze out. An ejection step is implemented where we parametrically drive our electrodes to excite any high orbit ions that have not crystallized. Finally, we increase the 397nm power if we loaded $\text{H}_2^{35}\text{Cl}^+$ and need to dissociate it.

was loaded.

6.3.2 Coupling Voltage

Unlike with the trapping RF, which has a helical resonator to reduce insertion loss onto the electrodes for a high voltage [Hen12], the EGGs electrodes do not have any special resonator. Part of the reason is that the frequencies that we are introducing are much higher than the 19MHz that is used for trapping, and it becomes more difficult to design a resonator that works at these frequencies. The inductance from the wiring to the electrodes becomes a dominant term in the RLC circuit, setting a minimum limit to the inductance of the system. An additional reason, if that the EGGs scheme specifically involves applying at least two tones, separated by around 2-3MHz. A resonator like the one for trapping has a bandwidth of $\sim 100\text{kHz}$, which would only allow a single tone to reach the electrodes without being heavily attenuated.

The EGGs electrodes will have some insertion loss from an impedance mismatch, which needs to be calibrated to determine how much voltage is actually reaching the blades of the ion trap. Prior to this experiment, there wasn't a reliable way to

determine these voltages in situ. Using a technique outlined in the next chapter, we introduced a novel way to perform an in situ measurement of the voltage that is being applied to the electrodes using a measurement at the ion itself. The insertion loss has a frequency dependence that this method is able to measure, so we can determine how to best match the strength of the red and blue sideband tones. Additionally, this method can be phase sensitive to ensure that the two sidebands are in phase with one another.

The EGGs electrodes have the same length of wiring going from the feedthrough to the trap, but take slightly different paths through the cryogenic shielding. This and perhaps some other asymmetries between the EGGs electrodes, lead to a difference in phase between the two blades, referred to as Channel 0 and Channel 1. The ARTIQ phaser is able to provide individual control over the phase and amplitude of these two channels to compensate for the mismatch that occurs, but the calibration of these two channels is non-trivial. If we are trying to apply an electric field in the quadrupole configuration, and there is a phase or voltage imbalance, an electric field in the dipole configuration will be applied as well.

To test the field produced by each electrode, the coupling efficiency and phase delay of each electrode need to be individually measured. Applying an oscillating voltage onto a single electrode in a quadrupole trap can be decomposed into half quadrupole and half dipole, each in phase with each other. As discussed in the next chapter, applying three RF tones where two are at the red and blue motional sidebands of the third/carrier tone will result in a displacement, dependent on the voltages of all the tones, as well as the frequency and phase of the carrier tone. Thus, by a measurement of the phonon number produced in this operation, we can determine the voltage applied onto the electrode, as well as the phase of the carrier tone. Since the signal is controlled up until the vacuum feedthroughs, we can confirm the power and phase of all three tones as well as the power and phase of both channels, prior to attaching them to the electrodes. As such, the phase difference on the carrier tone should be representative of the phase difference between the two EGGs electrodes. This allows us to estimate the insertion loss and phase delay with an in situ measurement of the ion.

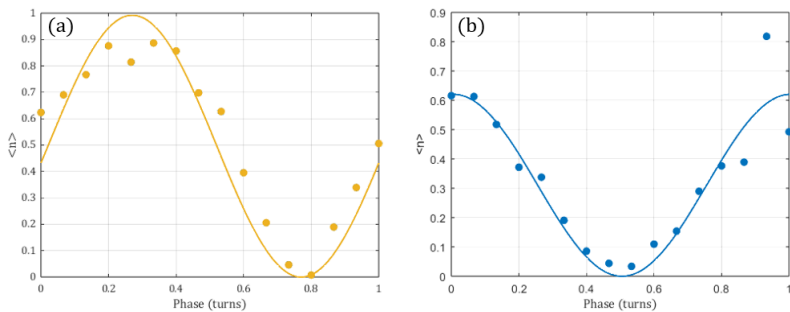


Figure 6.5: By applying tones to a single electrode, we should be generating equal parts dipole/quadrupole for the electric field. Applying three tones, a phase-sensitive measurement is performed, whereby we change the phase of just the red sideband. An initial test of the EGGs channel 0 (a), used 13dB attenuation compared to the measurement done on the channel 1 (b). Taking into account the peak phonon number generated by each channel, this resulted in ~ 12 dB difference in insertion loss between the two channels. Additionally, there is a phase difference between the two channels, $\sim \frac{\pi}{2}$ for the red sideband sweep that was performed. This corresponds to $\sim \frac{\pi}{4}$ difference between the carriers.

Furthermore, by using the shim electrodes to translate the ion near the trap center we are able to measure the imbalance between the two EGGs electrodes to probe the electric field, allowing us to map the field and determine our trap null in this direction at the desired frequency. As the ion is pushed out of the EGGs trap null, it should increase the projection onto the RF radial direction, resulting in an apparent increase in the coupling strength, set by normalizing to the minimum amplitude of either of the electrodes. Additionally, with these measurements we see that the RF micromotion null is at a different compensation voltage compared to the EGGs electrodes, suggesting that the parametric method for micromotion compensation may still lead to an offset for the EGGs dimension. Offsets from the null can produce a large AC Stark shift. By performing this measurement at several frequencies, we can determine that there is a large frequency dependence, requiring a careful calibration to confirm the correct shimming voltages during a spectroscopy sweep, shown in Fig. 6.6.

Each frequency may experience a different phase delay, as well as a phase shift from the different channels. To accurately determine the phases, we need to imple-

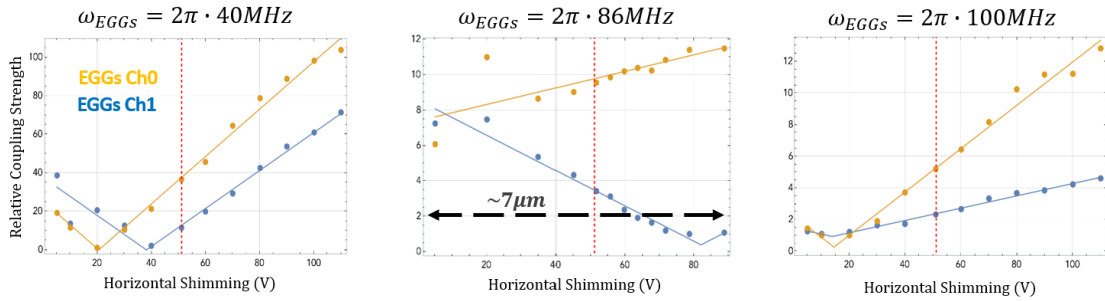


Figure 6.6: In order to map the trap null for the EGGs electrodes, the field strength needs to be estimated for each electrode via a two-tone motional Raman pulse. The red dotted line represents the location of the trapping RF micromotion null. The relative coupling strength is determined relative to the minimum voltage measured, using the conversion of phonon number to voltage outlined in the following chapter. Measurements were taken at three different frequencies, and by scanning nearly 100V on the horizontal shimming electrode, the ion is visibly displaced by $\sim 7\mu\text{m}$.

ment procedures that are phase-sensitive and will be able to probe a single phase at a time. Using the motional Raman method, we can produce a displacement when two tones are used, one in the dipole and one in the quadrupole configuration. In practice we need to calibrate to ensure that the tone is correctly in a given configuration and not in a linear combination of the two. Consider the case where we apply the RSB and the carrier tone on a single electrode. If we do a single displacement with these two tones, we will be unable to see any change in phonon number from a phase change on one of the tones since it will be a global phase. We need to introduce a separate displacement that is independent of the applied tones (and channel), which can be accomplished using a laser-based displacement. If we start with the laser-based displacement, the two-tone displacement will now be referenced to something else, and sweeping the RSB phase will result in an oscillation in the phonon number. However, the result is that we get the difference in phase between the two tones, not the individual values. Repeating these measurements with different combinations can provide enough relationships to calculate all the unknowns.

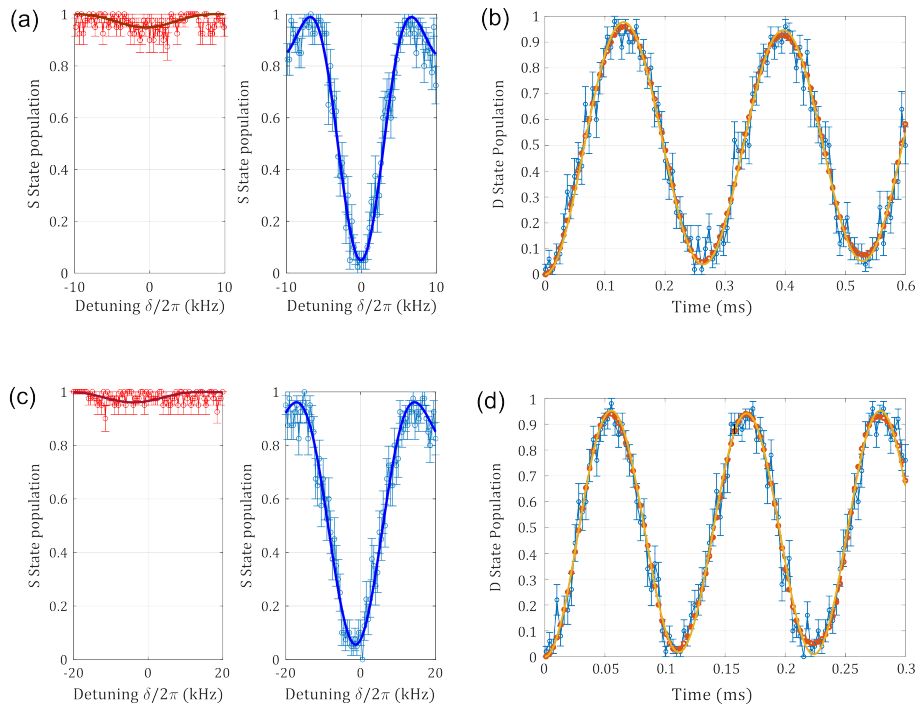


Figure 6.7: Attempts at searching for the transitions have been made using the radial RF modes, since the EGGs modes experience significantly higher heating and the axial modes are poorly coupled to the radial electric-field gradient. For a pair of Ca^+ and H^{35}Cl^+ , the two RF modes can be effectively cooled to the motional ground state by cooling every mode. (a)/(b) demonstrates an average phonon number of $\bar{n} = 0.058/0.017$ on the RF COM mode for 16ms of sideband cooling. (c)/(d) demonstrates an average phonon number of $\bar{n} = 0.046/0.017$ on the RF ROCKING mode for 8.888ms of sideband cooling. Each cycle of sideband cooling in (a)/(b) consists of {ER(13%), AR(10%), EC(15%), RR(13%), AC(13%), RC(36%)} where ($X\%$) denotes what percentage of that cycle the mode took up. (c)/(d) consists of {ER(13%), AR(10%), EC(15%), RC(19%), AC(20%), RR(23%)}. For ease, shorthand notation is used for each mode where the first letter denotes which direction (EGGs, RF, or Axial) and the second letter denotes if it is the COM mode or the ROCKING mode. (a)/(b) applies a $40\mu\text{s}$ spin polarization pulse every $640\mu\text{s}$, whereas (c)/(d) applies it every $191\mu\text{s}$.

6.3.3 Mode Choice

Various considerations have been made regarding which mode to drive for detecting transitions. Due to the poor projection of the electric-field gradient onto the axial modes, only the radial modes are considered for implementing EGGs. Additionally, the EGGs modes experience a significant heating rate from the high power amplifiers, and an even higher background heating rate from the motional Raman effect, due in part to the large geometric projection of the dipole tone. This leaves us with either the RF COM mode or the RF ROCKING mode. For the COM mode, the mode parameter for the Ca^+ ion is nominally 0.425 whereas for H^{35}Cl^+ it is 0.905, and for the ROCKING mode Ca^+ is nominally -0.905 whereas for H^{35}Cl^+ is 0.425. When b_{HCl} is large, the heating effect from EGGs will be greater; when b_{Ca} is large, the sideband cooling rate increases. Since these mode parameters have an inverse relationship to one another a choice needs to be made: try heating more or try scanning faster. While both options have been investigated, the typical choice is the latter for two additional reasons. The difference in sign between the atomic and molecular mode parameter for the ROCKING mode indicates that the heating effect from motional Raman will be weaker since there will be partial cancellation, which allows us to provide a larger dipole tone for greater linewidth broadening. And, since sideband cooling takes a significant portion of our pulse sequence, the speedup to the scans reduces the time that the molecular population could have redistributed into another state.

HCl^+ is highly reactive, with reactions occurring on the order of tens of minutes to several hours depending on vacuum conditions. It is not uncommon that a reaction can occur during or between datasets, resulting in $\text{H}_2^{35}\text{Cl}^+$. The change in secular frequencies from the reaction is on the order of kHz to low 10's kHz. When EGGs sweeps use red sideband RAP for readout, the robust nature of this method can let a reaction go undetected on the RF Rocking mode, which has $\sim 8\text{kHz}$ separation between the two molecules, whereas the RF COM mode has $\sim 40\text{kHz}$. We have characterized RAP to being robust for $\sim 20 - 50\text{kHz}$, and was motivated by the single point readout compared to the sideband ratio method which required both the RSB and BSB to be measured. This readout scheme can halve our required

probe duration, an important step for performing the spectroscopy.

6.3.4 Molecular Dipole Estimates

HCl⁺'s dipole moment is about 1.7 Debyes [Dal91], but an effective dipole exists for each transition, determined by how the dipole is oriented to the electric field. These transition dipole moments are the off-diagonal terms of the Hamiltonian

$$\frac{H_E}{\hbar} = -\boldsymbol{\mu}_e \cdot \mathbf{E} \quad (6.13)$$

using Eqn. A.1. Orientation of the magnetic field relative to the electric field (β) also must be considered. For π ($\Delta m_F = 0$) transitions, the electric field projection is along the dipole (internuclear axis), whereas $\sigma_{\pm}$ ($\Delta m_F = \pm 1$) transitions are orthogonal to the dipole. Since the dipole will be aligned to the magnetic field, this will look like

$$\langle \psi | \frac{H_E}{\hbar} | \psi' \rangle = -\mu_e E \cos(\beta) \langle \psi | D_{00}^{(1)} * T_0^1(\mathbf{L}) | \psi' \rangle \quad (6.14)$$

for π transitions and

$$\langle \psi | \frac{H_E}{\hbar} | \psi' \rangle = -\mu_e E \sin(\beta) \langle \psi | D_{0\pm 1}^{(1)} * T_{\pm 1}^1(\mathbf{L}) | \psi' \rangle \quad (6.15)$$

for $\sigma_{\pm}$ transitions. The effective dipole moments are displayed in Tab. 6.1.

6.3.5 Linewidth

As mentioned earlier, the transitions that we intend to drive are unknown, but many are estimated to exist within a 10MHz range. The linewidth of these transitions determines how easy it is to drive them. For the EGGs proposal, the linewidth will be given by

$$\Omega_{EGGS} = \frac{\Omega_0^2}{\sqrt{\Omega_0^2 + \delta^2}} \quad (6.16)$$

where

$$\Omega_0 = \eta_i \frac{2dV_m}{\hbar r_0} \quad (6.17)$$

which is the Rabi rate for the two-qubit gate. Note that Ω in Eqn. 6.11 is not the same as Ω_0 , since Ω_0 is used to make a Hamiltonian of the form

$$\frac{H}{\hbar} = \frac{\Omega_0 t}{2} \sigma_x \quad (6.18)$$

Transition	$I = I_{Cl} + I_H = 2$	$I = I_{Cl} - I_H = 1$
------------	------------------------	------------------------

π Transitions

$ \frac{7}{2}, \frac{7}{2}\rangle \Leftrightarrow \frac{7}{2}, \frac{7}{2}\rangle$	0.3	NA
$ \frac{7}{2}, \frac{5}{2}\rangle \Leftrightarrow \frac{7}{2}, \frac{5}{2}\rangle$	0.214286	NA
$ \frac{7}{2}, \frac{3}{2}\rangle \Leftrightarrow \frac{7}{2}, \frac{3}{2}\rangle$	0.128571	NA
$ \frac{7}{2}, \frac{1}{2}\rangle \Leftrightarrow \frac{7}{2}, \frac{1}{2}\rangle$	0.0428571	NA
$ \frac{5}{2}, \frac{5}{2}\rangle \Leftrightarrow \frac{5}{2}, \frac{5}{2}\rangle$	0.185714	0.3
$ \frac{5}{2}, \frac{3}{2}\rangle \Leftrightarrow \frac{5}{2}, \frac{3}{2}\rangle$	0.111429	0.18
$ \frac{5}{2}, \frac{1}{2}\rangle \Leftrightarrow \frac{5}{2}, \frac{1}{2}\rangle$	0.0371429	0.06
$ \frac{3}{2}, \frac{3}{2}\rangle \Leftrightarrow \frac{3}{2}, \frac{3}{2}\rangle$	0.06	0.22
$ \frac{3}{2}, \frac{1}{2}\rangle \Leftrightarrow \frac{3}{2}, \frac{1}{2}\rangle$	0.02	0.0733333
$ \frac{1}{2}, \frac{1}{2}\rangle \Leftrightarrow \frac{1}{2}, \frac{1}{2}\rangle$	0.1	0.166667

σ Transitions

$ \frac{7}{2}, \frac{7}{2}\rangle \Leftrightarrow \frac{7}{2}, \frac{5}{2}\rangle$	0.277746	NA
$ \frac{7}{2}, \frac{5}{2}\rangle \Leftrightarrow \frac{7}{2}, \frac{3}{2}\rangle$	0.363655	NA
$ \frac{7}{2}, \frac{3}{2}\rangle \Leftrightarrow \frac{7}{2}, \frac{1}{2}\rangle$	0.406579	NA
$ \frac{7}{2}, \frac{1}{2}\rangle \Leftrightarrow \frac{7}{2}, -\frac{1}{2}\rangle$	0.419913	NA
$ \frac{5}{2}, \frac{5}{2}\rangle \Leftrightarrow \frac{5}{2}, \frac{3}{2}\rangle$	0.20344	0.328634
$ \frac{5}{2}, \frac{3}{2}\rangle \Leftrightarrow \frac{5}{2}, \frac{1}{2}\rangle$	0.257333	0.415692
$ \frac{5}{2}, \frac{1}{2}\rangle \Leftrightarrow \frac{5}{2}, -\frac{1}{2}\rangle$	0.272943	0.440908
$ \frac{3}{2}, \frac{3}{2}\rangle \Leftrightarrow \frac{3}{2}, \frac{1}{2}\rangle$	0.0848528	0.311127
$ \frac{3}{2}, \frac{1}{2}\rangle \Leftrightarrow \frac{3}{2}, -\frac{1}{2}\rangle$	0.0979796	0.359258
$ \frac{1}{2}, \frac{1}{2}\rangle \Leftrightarrow \frac{1}{2}, -\frac{1}{2}\rangle$	0.244949	0.408248

Table 6.1: H^{35}Cl^+ effective dipoles calculated for a permanent dipole of 1.7Debye, and an angle of 60° between the magnetic field (dipole alignment) and the electric field (trap electrodes). The left column shows the transitions that are involved; for ease only positive states are shown but negative states will have the same values. The middle column is the condition that the hydrogen nuclear spin is aligned with the chlorine nuclear spin, and the right column is when it is anti-aligned.

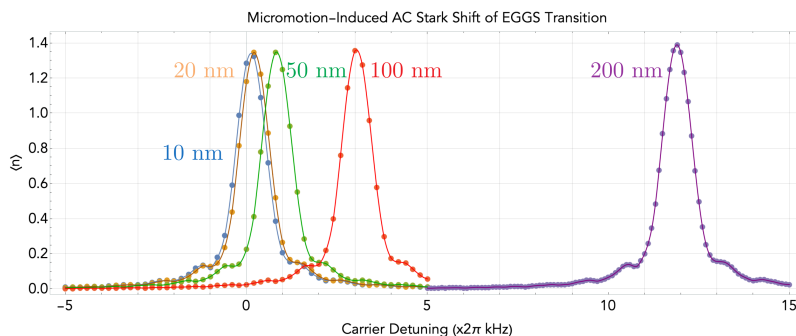


Figure 6.8: When the molecule is offset from the trap’s micromotion trap null, it will experience an AC Stark shift dependent on how far it is offset from the null. To minimize this effect, micromotion compensation needs to be done regularly when performing molecular spectroscopy.

which couples two states and follows the state probability given by Eqn. 3.42. Estimates using nominal quadrupole voltages and molecular dipole moments have $\Omega_0 \sim 300\text{Hz}$, which are extremely narrow transitions to attempt to drive. AC Stark shifts can come from the molecule displaced from the RF and EGGs nulls, as well as from an off-resonant dipole term coupling to other states. Regardless of the source, such a narrow transition can be shifted many times further than its own linewidth, which makes repeatable measurements particularly challenging.

6.3.5.1 Electric Dipole Leakage

The dipole field is proposed to drive the single qubit gates within our molecule, whereas a pure quadrupole is needed for state readout. Should there be an imbalance in the electrode voltages or phase, the desired quadrupole will have some dipole component. Consider the case in which each EGGs electrode has a single tone applied to it, one channel with $V \cos(\omega t)$ and the other with $V(1 + \varepsilon) \cos(\omega t - \varphi)$. This can be decomposed into two cases: there is $V \cos(\omega t)$ and $V \cos(\omega t - \varphi)$ in one case, and 0 and $V\varepsilon \cos(\omega t - \varphi)$ in the other. The latter was mentioned earlier as being made up equally of half dipole and half quadrupole. The former can be

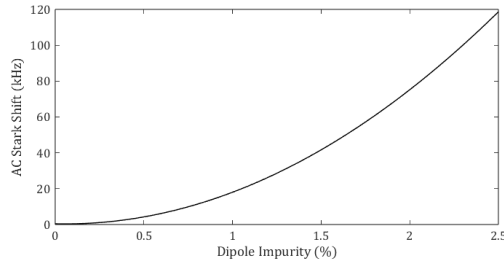


Figure 6.9: Simulated result from Qutip for the AC Stark shift that results from an off-resonant dipole coupling to an additional hyperfine state.

calculated by rewriting the voltages as

$$V \cos(\omega t) = V \left(\cos\left(\omega t - \frac{\varphi}{2}\right) \cos\left(\frac{\varphi}{2}\right) + \sin\left(\omega t - \frac{\varphi}{2}\right) \sin\left(\frac{\varphi}{2}\right) \right) \quad (6.19)$$

$$V \cos(\omega t - \varphi) = V \left(\cos\left(\omega t - \frac{\varphi}{2}\right) \cos\left(\frac{\varphi}{2}\right) - \sin\left(\omega t - \frac{\varphi}{2}\right) \sin\left(\frac{\varphi}{2}\right) \right). \quad (6.20)$$

Written in this way, it should be clear that one term makes up a quadrupole voltage and the other makes up the dipole, and a phase imbalance between the two applied tones results in a dipole tone being produced, off-resonant from the carrier by the secular frequency. The effect of this off-resonant dipole is an AC-Stark shift, which can dramatically shift the carrier frequency. So $\varphi \neq 0$ and $\varepsilon \neq 0$ will both create detrimental AC-Stark shifts. The origin of this shift comes from off-resonant couplings to other hyperfine states. The magnetic field produced in this experiment will produce a Zeeman splitting in H^{35}Cl^+ that is $\sim 1.8\text{MHz}$, which determines the difference in energy between a π , a σ_- , and a σ_+ transition. When trying to drive transitions using the RF COM mode ($\sim 1.4\text{MHz}$), a weak dipole term on the sidebands can produce a stronger off-resonant Rabi rate than the readout rate, since the dipole term does not include $\eta \sim 10^{-5}$. The effect is the AC Stark shift shown in Fig. 6.9.

6.3.6 Green EGGs

What color is between red and blue? Green EGGs is a scheme that we implemented to make both a Dr. Seuss reference and to try broadening the linewidth of the transitions. When a third tone is added to the EGGs quadrupole tones, is on resonance (hence green), and is applied within the dipole configuration then the linewidth will

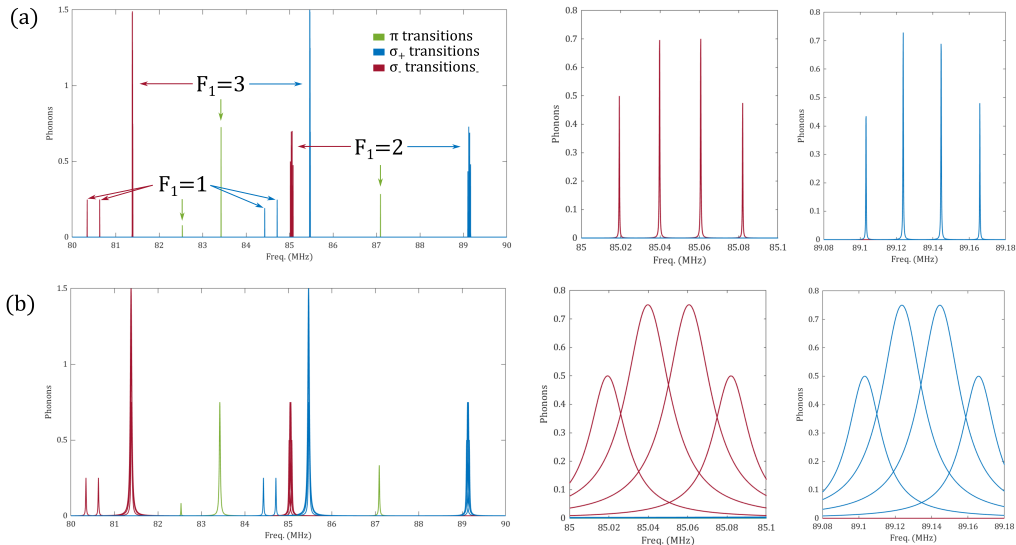


Figure 6.10: Standard EGGs (no carrier tone) produces the linewidths shown in (a) for each of the different transitions. The linewidths are given by equations 6.16 and 6.17, assuming 7V quadrupole tones are driving the RF ROCKING mode, $b_{HCl} = 0.42$ and $\omega_0 = 2\pi \cdot 1.142\text{MHz}$. To emphasize how narrow the transitions are ($\sim 2\pi \cdot 300\text{Hz}$), the $F_1 = 2$ $\sigma_{\pm}$ transitions are shown zoomed in. For ease the hydrogen nuclear spin has been excluded, since it has little effect on the predicted location of the peaks. In (b), we demonstrate what linewidths we would expect when we apply a 10mV dipole tone at the carrier to generate Green EGGs ($\sim 2\pi \cdot 25\text{kHz}$).

experience broadening dependent on the strength of the carrier tone. Since the EGGs Hamiltonian is effectively a Mølmer-Sørensen gate and the resonant tone is a single qubit gate, this method can be thought of as a continuous dynamic decoupling scheme. Dynamic decoupling schemes in traditional Mølmer-Sørensen based systems have been demonstrated in [MRS17, CH22] and similarly in magnetic field gradient systems [Sri20]. Switching into the interaction picture with respect to this additional tone results in a new Rabi rate given by

$$\Omega_{GE} = \frac{\Omega_0 \Omega_C^2}{\Omega_C^2 + \delta^2}, \quad (6.21)$$

where the value Ω_C is the dipole Rabi rate from the carrier tone. This Rabi rate indicates that the linewidth will be broadened based on the strength of the dynamic decoupling tone which, unlike the readout scheme, can be significantly faster since it is not reduced by the effective Lamb-Dicke parameter.

6.3.7 Background Heating

In order to determine whether a transition has been driven we measure the phonon number of the ion crystal at the mode of interest. Estimates of the peak phonon number are provided by the EGGs theory $\bar{n} = (2\Omega\eta t)^2$, but these excitations are typically small, so making sure the background phonon numbers are low ensures that the signal-to-noise ratio is large enough to confirm that heating is from driving a transition. Since we can sideband cool the ion chain well, the dominant background comes from either the motional squeezing effect or from the motional Raman background when doing the Green EGGs protocol.

6.3.7.1 Squeezing

Motional squeezing occurs when large enough voltages are applied parametrically on the trap electrodes. Traditional squeezing occurs when the parametric tones are applied at $\pm 2\omega_0$ [Bur20], however, through multiphoton processes, an effective squeezing unitary is produced by two tones separated by $2\omega_0$ that are applied in the quadrupole configuration (an $\hat{x}^2$ operation). This is unfortunately the exact way we apply the spectroscopy EGGs scheme. The quadrupole field in this case will be coupled to the charge of the ions, not the dipole of the molecules, such that the Hamiltonian will be written as

$$\frac{\hat{H}}{\hbar} = \omega_0 \hat{a}^\dagger \hat{a} + \Omega_q (\hat{a} + \hat{a}^\dagger)^2 \cos((\omega_d + \delta)t) \cos(\omega_0 t) \quad (6.22)$$

where $\Omega_q = \frac{eV_q x_0^2}{\hbar r_0^2}$ and ω_c is the carrier frequency. Solving the second order Magnus term (shown in the following chapter) will produce a squeezing time evolution operator with

$$g = -i\Omega_q^2 \left(\frac{\omega_0}{\omega_c^2 + \omega_0^2} \right) \quad (6.23)$$

where the squeeze strength $\xi = gt$. When the QHO is squeezed, even Fock states become populated, and the average phonon number increases by

$$\bar{n}_{sq} = \sinh^2(|gt|). \quad (6.24)$$

This background signal starts to become apparent ($\bar{n} \approx 0.1$) when the quadrupole voltages are $\sim 7V$ at 80MHz or $\sim 38V$ at 500MHz. Limiting the background from squeezing ends up putting a bound on how much EGGs voltage we can apply.

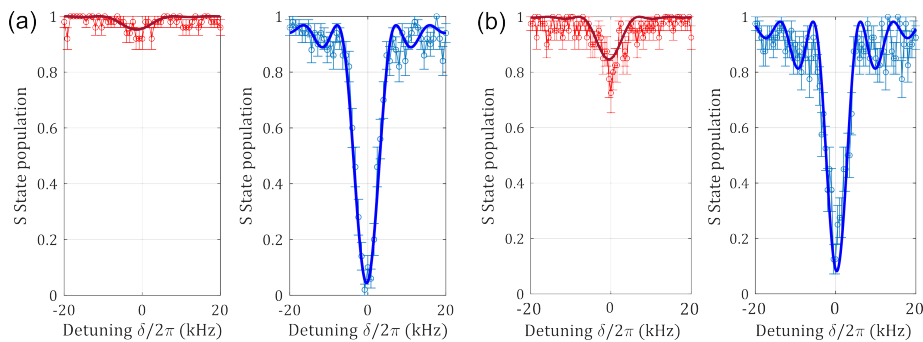


Figure 6.11: Residual background heating on the RF COM mode after (a) sideband cooling and (b) 1ms of EGGs with a weak dynamic decoupling term. Compared to the ROCKING mode, the COM mode is more susceptible to the motional Raman heating effect.

6.3.7.2 Motional Raman Background

Although the Green EGGs scheme allows us to increase the linewidth of the transitions, it can introduce a significant amount of background signal. The introduction of the third tone results in a displacement operator (covered in the next chapter). As a larger carrier tone is applied, the strength of the displacement will increase, which eventually starts to increase the background phonon number given by

$$\bar{n}_{QVSA} = \Omega_q^2 \Omega_d^2 t^2 \left(\sum_j b_j \frac{\omega_0}{\omega_c^2 - \omega_0^2} \right)^2 \quad (6.25)$$

where $\Omega_d = \kappa \frac{eV_d}{\hbar \omega_0} x_0$ and κ is a geometric projection onto a given direction. Since this displacement affects all ions, the mode parameter b_j must be summed for all ions prior to conversion to an average phonon number, such that the rocking mode will have a significantly reduced effect from the difference in sign. As such, despite the weaker predicted phonon number from the EGGs effect, by taking measurements using the ROCKING mode, a larger dynamic decoupling tone can be applied for broadening the transitions. Additionally, as with the second order squeezing effect, this displacement becomes dramatically weaker at higher frequencies, allowing for larger dipole voltages to be applied when probing for the $\sim 500\text{MHz}$ transitions compared to the $\sim 80\text{MHz}$ ones.

6.3.7.3 Phase Shift Keying

Suppression of the QVSA heating can be achieved using phase shift keying. Phase shift keying involves reversing the phase of the carrier during the experiment, so that any displacement from the QVSA effect gets canceled out once the dipole phase changes. The quadrupole tones, which are used to create the molecule's displacement operation, are unaffected by this phase reversal, so that the strength of the EGGS displacement remains the same. While this does a good job removing the background from QVSA, it only does so when on resonance. The lineshape of a phase shifted scan shows sharp increases in phonon number once moved off-resonance. As a result, when there is a drift in secular frequency, a large background signal appears. For a probe time of 1ms, this drift only needs to be 2kHz to cause a 0.8 increase in phonon number. This result is demonstrated in Fig. 6.12.

6.3.8 Redistribution

This experiment is designed around limiting any electric fields that could inadvertently move population out of the qubit manifold and into a different state. These redistribution pathways result in the population being in an ever-changing molecular state. Keeping the redistribution rates low is critical to getting reliable and consistent data, otherwise partway through an experiment the molecular state may leave the manifold and may not return.

6.3.8.1 Black Body Radiation

Covered a bit earlier in the prior chapter, considerations were made to try limiting the effects of black body radiation (BBR) that could reach our molecule. If the molecule were placed within a 4K box, the ground state would nearly never experience an excitation into a higher rotational state. However, since room temperature BBR is able to enter the 4K shielding to excite the ion, this lifetime is significantly reduced. If we consider the room temperature lifetime of the ground rotational state to be 6s, then the room temperature BBR would limit that lifetime to about 1220s given the transmission measured as well as the solid angle of the windows to the ion.

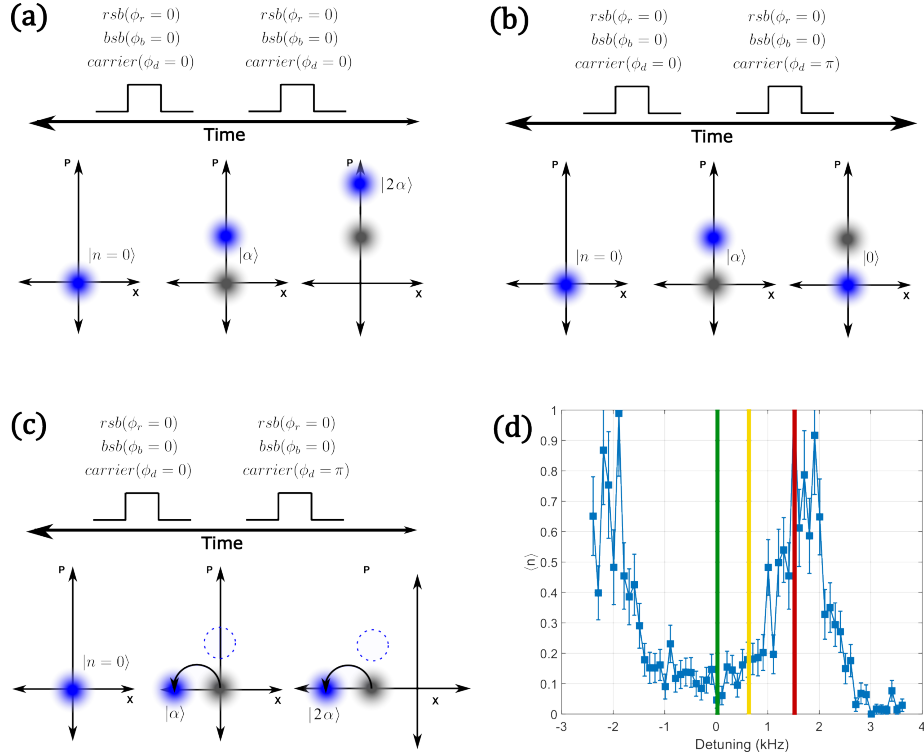


Figure 6.12: To remove residual heating caused by motional Raman, phase shift keying (PSK) was implemented. The pulse is separated into two sections. Without PSK, the sequence can be visualized as two identical displacements (a). If the carrier tone, has its phase flipped halfway through the sequence, the second displacement will cancel the first (b). However, this is only true for on-resonance displacements, and when a detuned displacement is considered, the PSK sequence can still result in heating in the form of sidebands (c). When the secular frequency drifts, it is possible that heating from these sidebands becomes troublesome (d). A prior measurement indicated the secular frequency was at the location marked by the yellow bar. The subsequent measurement now has the secular frequency at the green bar. Had the initial frequency drifted in the opposite direction by a similar amount, it would experience higher heating, shown by the red bar.

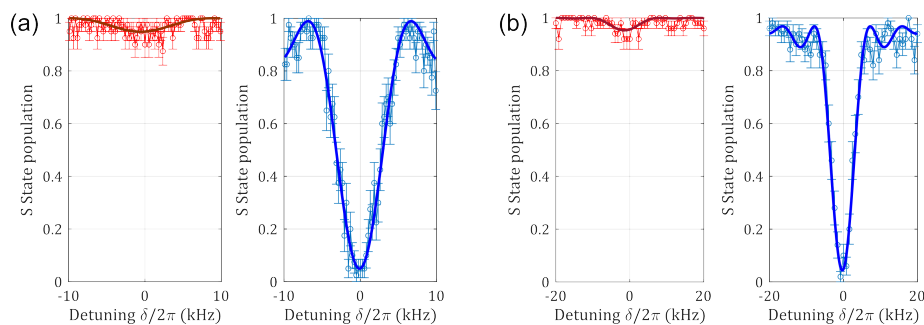


Figure 6.13: A reduction in the overall sideband cooling time is possible by optimizing the cooling of spectator modes. The RF COM mode in (a) is cooled with 16ms of sideband cooling on all modes and in (b) is cooled with only 8.888ms, cooling only five of the six modes.

Additionally, this is assuming the beryllium copper pieces have an emissivity of 0.3 and the copper pieces 0.1, the trap and its pieces are at 4K, and the BBR from 300K is not reflected back onto the ion. Assuming it is reflected, this lifetime reduces to 670s. Furthermore, if we assume the trap is not at 4K but instead 20K, as the temperature diode would suggest, then the lifetime would be reduced further to 558s.

6.3.8.2 Johnson-Nyquist Noise

One condition that was not initially considered, but later became a considerable variable later on was how the fundamental thermal effects may redistribute the molecular population. The Johnson-Nyquist noise from the room temperature RF components will result in a power spectral density of -174dBm per 1Hz. The rate of a spin flip is given by

$$\Gamma = \left| \frac{1}{\hbar} \int dt \langle \uparrow | dE(t) \sigma_x | \downarrow \rangle e^{i\omega t} \right|^2 \quad (6.26)$$

$$= \frac{1}{\hbar^2} \int dt \langle \uparrow | dE(t) \sigma_x | \downarrow \rangle e^{i\omega t} \int dt' \langle \downarrow | dE(t') \sigma_x | \uparrow \rangle e^{-i\omega t'} \quad (6.27)$$

$$= \frac{d^2}{\hbar^2} \int dt \int d\tau E(t) E(t + \tau) e^{i\omega \tau} \quad (6.28)$$

where $t' = t + \tau$, and

$$S_E(\omega) = \int dt \int d\tau E(t) E(t + \tau) e^{i\omega \tau} \quad (6.29)$$

is the electric field noise spectral density. For a given temperature and resistance the electric field noise spectral density for the radial modes is

$$S_E(\omega) = \frac{4k_bTR(\omega)}{r_0^2}. \quad (6.30)$$

Since we care about the voltage amplitude for redistribution, half of this spectral density will be considered, which results in a redistribution every ~ 2900 seconds. To get a better idea of what the rate will be for our experiment, we measured the power spectral noise of the SMA cable before it was attached to the chamber feedthrough. In the initial test, the EGGs amplifiers (+52dB) were not connected to a switch, so they would always be on, and we measured a power spectral noise of -110dBm in a 1Hz window. To convert the power spectral density to the electric field spectral density, we use

$$S_E(\omega) = \frac{2R}{r_0^2} S_P(\omega) \quad (6.31)$$

where R is assumed to be 50Ω and the electric field is a perfect dipole, which results in a redistribution event every ~ 1 ms. To mitigate this effect, the amplifiers go into high power switches (-53dB insertion loss) that disconnect them when not in use. When measuring the residual power noise, in this configuration, we reached the noise floor of the device, -150dBm at 1Hz, so the noise will be -160dBm at 1Hz, based off the previous measurement and the gain of the amplifiers. This would result in a redistribution event every ~ 100 seconds, which led us to limit our probe times to about two minutes per run. An additional measurement that was made following the amplifier switches, was a measurement of the EGGs mode heating rates shown in Fig. 6.14, confirming that the switches were reducing unnecessary heating on that mode.

6.4 Preliminary Results and Discussion

Many hours have been spent taking spectroscopic data on the transitions. Unfortunately, we've not taken definitive data that would allow us to confidently say that a transition has been driven. Criteria for confidence include: repeatability with and without the Green EGGs dipole term, broadening from the dipole term, consistency with expected phonon number and linewidth, and confirmation that a peak does not

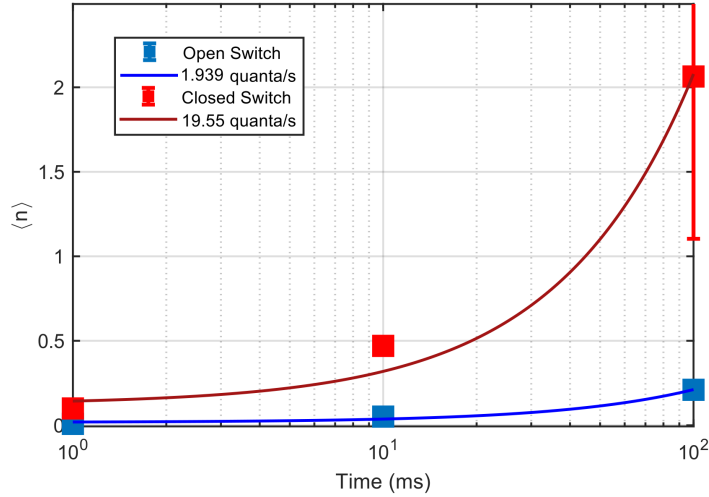


Figure 6.14: To determine how much of an effect the amplifiers were causing, we took measurements of the EGGs mode with them connected to a switch. The red data shows the heating rate when the switch was closed and the noise from the amplifier heated the ion. The blue data shows that when the switch is open, the heating rate reduces by an order of magnitude.

correspond to a PSK secular drift, a reaction, the crystal melting or an ion position swap, which requires randomized datasets. Our main focus has been around 83MHz, covering from 78MHz up to 90MHz, as well as transitions around 510MHz to 520MHz. For examples of possible peaks see Fig. 6.15 and 6.16. As mentioned these did not meet the confidence criteria. For example, while the peaks in Fig. 6.15 are reasonably close to the expected phonon number, the linewidth we'd expect from the applied dipole is significantly larger than the measured peaks. Additionally, as shown in Fig. 6.12, after this data was taken, a scan of the linewidth indicated the frequency the data was set to (yellow) was no longer at the minimum (green) and drift could easily push it into the heated (red) regime. Lastly, this data was taken using a linear sweep, in which case a drift into the heated region would result in several consecutive high points, whereas a randomized scan would only have consecutive high points in frequency if it were a real transition. In general, we have only seen pronounced peaks like these during linear frequency scans, and not from a randomized sweep. A more rigorous calibration of the voltages and phases needs to be done prior to the next round of EGGs spectroscopy in order to remove any

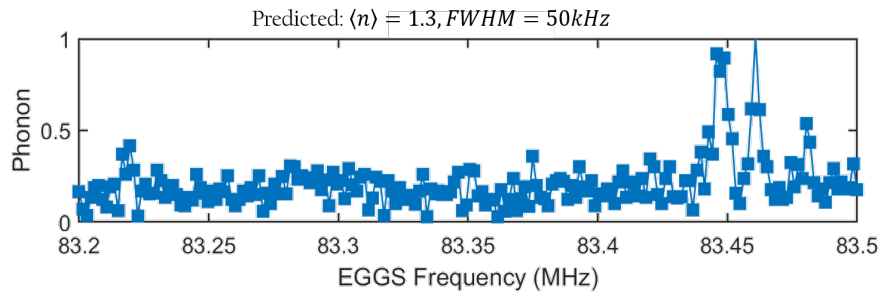


Figure 6.15: Sample dataset collected searching for a transition around 83.35MHz. Based on calibrations performed using second order squeezing and motional Raman tests, we estimated that the peak phonon number for a transition in this region would be $\bar{n} = 1.3$, and that, due to the dynamic decoupling term, the FWHM would be $\sim 50\text{kHz}$. The scan was performed using a linear sweep, from 83.2 to 83.5MHz with a 1.5kHz step, on the RF ROCKING mode, taking about 4 minutes to run. Subsequent runs showed higher heating rates and experienced more frequent crystal melting. I believe these peaks come from a drift in the secular frequency which hadn't been recalibrated in several hours. This region was the most studied, and we were unable to measure consistent and repeatable results.

possible unwanted dipole terms, via correct phases and balancing of the two EGS electrode voltages.

6.5 Raman Discussion

An alternative option for EGS spectroscopy is to implement molecular quantum logic spectroscopy schemes with laser-based Raman transitions for state detection. These schemes have been well developed in [Cho17, C 20, YC24, D 25, F 24] for state readout in molecular ion experiments. Theoretical estimates for implementing the scheme outlined in [Cho17] were done for H^{35}Cl^+ , with help by Prof. Michael Heaven at Emory for calculating the transition dipole moments of our molecule. In order to estimate the Raman rate

$$\Omega_{\text{Raman}} = \frac{\langle \psi_f | \mu_e E_2 | \psi_m \rangle \langle \psi_m | \mu_e E_1 | \psi_i \rangle}{4\hbar^2 \delta} (S_- + S_+) \quad (6.32)$$

we considered available lasers to estimate the maximum available optical power, the detuning from the optical electric dipole transition, and the minimum beam waist

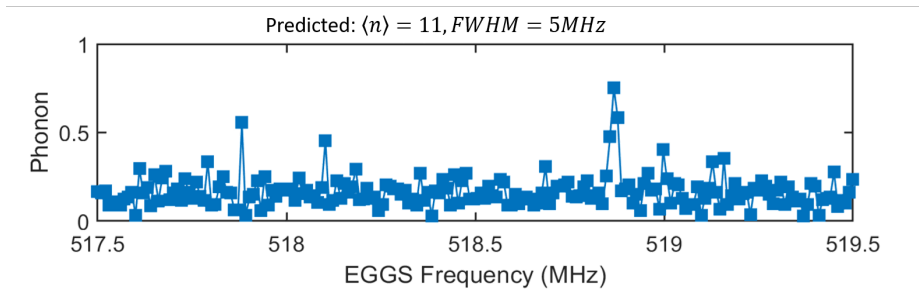


Figure 6.16: Sample dataset collected searching for a transition around 518.5MHz. Based on calibrations performed using second order squeezing and motional Raman tests, we estimated that the peak phonon number for a transition in this region would be $\bar{n} = 11$, and that, due to the dynamic decoupling term, the FWHM would be $\sim 5\text{MHz}$. Noticeably more power could be added to the trap electrodes before the background heating became problematic. The scan was performed using a linear sweep, from 517.5 to 519.5MHz with a 10kHz step, on the RF ROCKING mode, taking about 3.5 minutes to run. The secular frequency measurement in Fig. 6.12 was taken about a half hour after this dataset was collected, since subsequent runs showed higher heating rates as well, suggesting that these peaks were from the secular frequency drifting into a PSK sideband.

we could generate with a 150mm lens and a 1mm input Raman beam. The values

$$S_- = \sum_m \langle \psi_f | \hat{r} \cdot \hat{q}^{(2)} | \psi_m \rangle \langle \psi_m | \hat{r} \cdot \hat{q}^{(1)} | \psi_i \rangle \quad (6.33)$$

$$S_+ = \frac{\Delta - \omega_1}{\Delta + \omega_1} \sum_m \langle \psi_f | \hat{r} \cdot \hat{q}^{(1)} | \psi_m \rangle \langle \psi_m | \hat{r} \cdot \hat{q}^{(2)} | \psi_i \rangle \quad (6.34)$$

represent the angular momentum selection rules for the Raman light (ω_1) coupling to the excited state at Δ . The electric field generated by the laser is given by

$$E_j = \sqrt{\frac{2P_j}{\varepsilon_0 c A_j}} \quad (6.35)$$

where the subscript j denotes the two different Raman beams. Following [Choi17], to cancel the AC Stark shift, the laser configuration requires double the power on the σ_- polarized beam compared to the π polarized beam. For H^{35}Cl^+ , the transition dipole has been estimated to be 0.4 Debye and $S_- = \frac{1}{5}$. An estimate for a 532nm laser is that $P_1 = 1\text{W}$ and $P_2 = 2\text{W}$ with a $15\mu\text{m}$ waist would be required to create a $\sim 1\text{kHz}$ Raman rate.

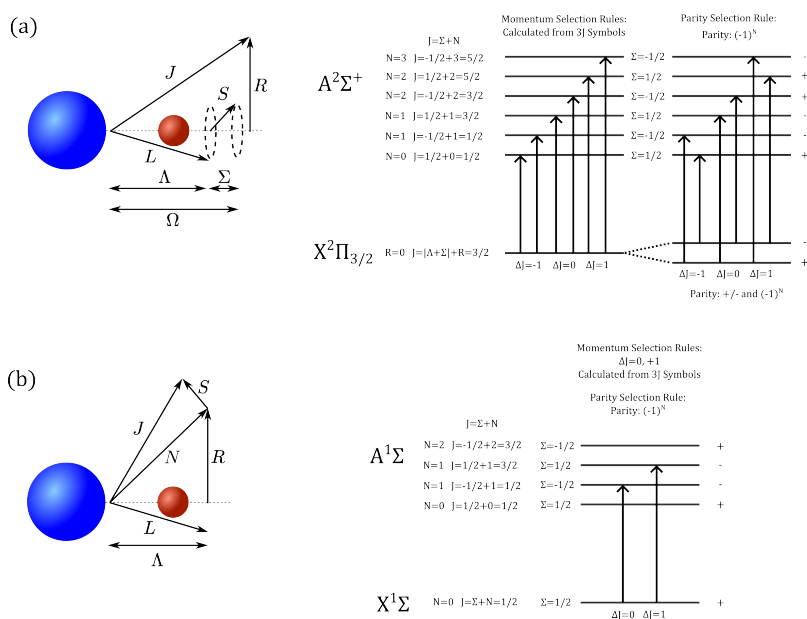


Figure 6.17: Hund's case (a) and (b) molecules couple to different states during a Raman process. For HCl^+ , the case (a) represents what states participate in this interaction. Since the Raman process is a two-photon process, the intermediate states must have a parity different from the original state. The parity of a Σ^+ state is determined solely by $(-1)^N$, whereas the parity of a Π state also includes the sign of the superposition ($\pm$) as shown in equations 4.33 and 4.34. Note that for the Σ^+ state, the quantum number $N = R$.

CHAPTER 7

Motional Raman

This section is dedicated to the theory and experimental results associated with the motional Raman technique discovered while introducing the Green EGGs scheme to the ions. The results from this study have been published in [WMH25] and this chapter will reflect this work. Since this work demonstrated the viability of an ion as a quantum vector signal analyzer, I will often refer to the scheme as such (QVSA).

7.1 Resonant Excitations

When a driving force is applied to a harmonic oscillator that is at the natural frequency of the oscillator, the force will produce a resonant excitation, quickly increasing the amplitude of oscillation. For the QHO defined by the ion trap, the force in question is generated by an electric field at the ion and when on resonance is referred to as a tickle. The potential of the oscillator is quantized by the amount of phonons within it, each with energy $\hbar\omega_0$. Photons introduced by the tickle will be at energy $\hbar(\omega_0 + \delta)$, where δ is a small detuning from resonance. The constant electric field at the center of the trap will introduce a Hamiltonian term

$$\hat{H} = -ex_0(\hat{a} + \hat{a}^\dagger)E \cos \omega t \approx -\frac{eE}{2}x_0(e^{-i\delta t}\hat{a} + e^{i\delta t}\hat{a}^\dagger) \quad (7.1)$$

where the magnitude of the electric field E , is assumed to be real for ease, producing a displacement, γ , given in magnitude by

$$|\gamma| = \frac{eE}{2\hbar}x_0t \operatorname{sinc}\left(\frac{\delta t}{2}\right) = \frac{\Omega_d}{2}t \quad (7.2)$$

having assumed the tickle began at $t = 0$. It is intuitive to define a dipole rate

$$\Omega_d = \frac{\kappa eV_d}{\hbar r_0}x_0 \quad (7.3)$$

where the electric field has been assumed to be a perfect dipole, i.e. $E = \frac{\kappa V_0}{r_0}$ with a projection of κ onto the mode of interest. This definition will be used later for any applied dipole field.

To think about this another way, consider the tickle as a signal that we are trying to detect, which can be pictured as a single photon at $\omega_s = \omega_0 + \delta$, which can be subtracted ($\hat{b}$) from or added ($\hat{b}^\dagger$) to the bath. In doing so, it can raise ($\hat{a}^\dagger$) or lower ($\hat{a}$) the number of phonons in the QHO since it is a linear force acting on the ion (Coulomb force). Thus a tickle is a single-photon/phonon process.

7.2 Motional Raman Excitations

Now consider the case in which the signal frequency ω_s is moved far off resonance $2\pi\delta t \gg 1$. In phase space it will make very small and very fast loops, but not produce any noticeable displacement. By introducing an additional probe photon into the system, such that it is the natural frequency away from the tickle $\omega_p = \omega_s \pm \omega_0$, a second-order heterodyne process can result in a beating of the two applied tones, one of which will appear at ω_0 . The second tone, however, needs to have a electric-field gradient, so that it produces a $\hat{a}^2$ and a $\hat{a}^{\dagger 2}$ term. The signal term introduces a single phonon step, whereas this probe term introduces a two-phonon step, with the heterodyne beating creating a single-phonon and triple-phonon step in the process, the latter of which is significantly weaker.

7.2.1 Theoretical Analysis

Since the signal that we intend to measure is applied onto our electrodes as a dipole tone, we denote its frequency as ω_d , whereas the probe tones at frequencies at $\omega_d + \delta \pm \omega_0$ are applied to the trap in a quadrupole configuration, as shown in Fig. 7.1. We assume $\omega_d \gg \omega_0 \gg \delta$. The motion of the ion can be described by the Hamiltonian

$$\frac{\hat{H}}{\hbar} = \omega_0 \hat{a}^\dagger \hat{a} + \Omega_q (\hat{a} + \hat{a}^\dagger)^2 \cos((\omega_d + \delta)t) \cos(\omega_0 t) + \Omega_d (\hat{a} + \hat{a}^\dagger) \cos(\omega_d t + \varphi_d) \quad (7.4)$$

where $\Omega_q = \frac{eV_q x_0^2}{\hbar r_0^2}$, $\Omega_d = \frac{\kappa e V_d x_0}{\hbar r_0}$, r_0 is the ion-electrode distance, $x_0 = \sqrt{\frac{\hbar}{2m\omega_0}}$ is the length scale of the QHO, and κ is a geometrical factor. In the interaction picture

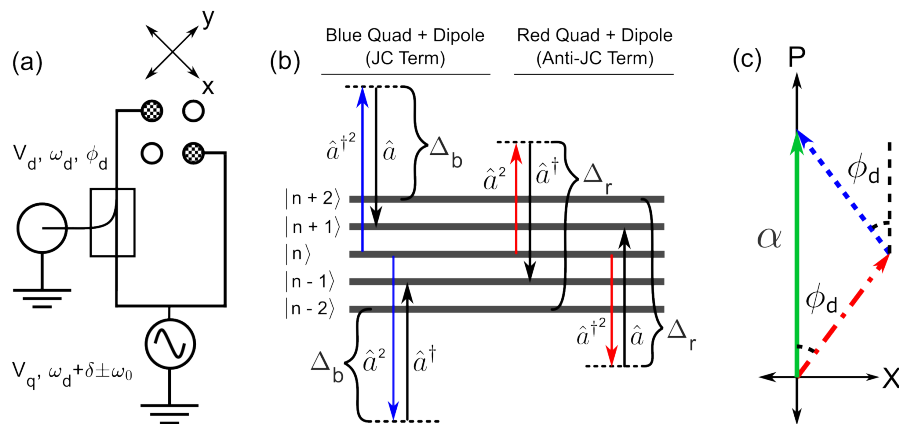


Figure 7.1: Implementation of the quantum vector signal analyzer (QVSA). (a) Simplified schematic of the QVSA drive coupling to the quadrupole ion trap. Two quadrupole tones ($\omega_d + \delta \pm \omega_0$) are coupled onto the electrodes to characterize an unknown dipole tone (ω_d). The dipole tone is easily supplied using either a BNC connector or a microwave horn. (b) Level diagrams for motional Raman transitions. The blue (red) arrow indicates the blue (red) quadrupole field, while the black arrow represents the dipole field. The combination of the blue quadrupole ($\omega_d + \delta + \omega$) and dipole fields results in displacement following the Jaynes–Cummings (JC) model, whereas the combination of the red quadrupole ($\omega_d + \delta - \omega$) and dipole fields results in displacement according to the anti-Jaynes–Cummings (Anti-JC) model. $\Delta_b = \omega_d - \omega_0$; $\Delta_r = \omega_d + \omega_0$; (c) Phase space diagram showing that the phase sensitivity of the QVSA displacement α arises from the interference between the displacements generated by the multiple Raman transitions; each transition is excited by one quadrupole and one dipole tone. We assume the initial phase of the quadrupole tones to be zero.

with respect to the harmonic oscillator, we have

$$\begin{aligned} \frac{\hat{H}_I}{\hbar} = & \Omega_q \left(\hat{a}^2 e^{-2i\omega_0 t} + 2 \left(\hat{a}^\dagger \hat{a} + \frac{1}{2} \right) + \hat{a}^{\dagger 2} e^{2i\omega_0 t} \right) \cos((\omega_d + \delta)t) \cos(\omega_0 t) \\ & + \Omega_d \left(\hat{a} e^{-i\omega_0 t} + \hat{a}^\dagger e^{i\omega_0 t} \right) \cos(\omega_d t + \varphi_d). \end{aligned} \quad (7.5)$$

To evaluate the propagator, we use the Magnus expansion, i.e. $\hat{U} = e^{\sum_i \hat{A}_i}$. The first-order Magnus term $\hat{A}_1$ consists of terms with amplitudes that scale as $\frac{\Omega_q}{\omega_d}$ and $\frac{\Omega_d}{\omega_d}$, which we assume to be small and therefore ignore.

The second order Magnus term is given by

$$\hat{A}_2 = \frac{1}{2} \left(-\frac{i}{\hbar} \right)^2 \int_0^t \int_0^{t'} \left[\hat{H}(t'), \hat{H}(t'') \right] dt'' dt'. \quad (7.6)$$

Again, we assume that terms proportional to $\frac{\Omega_q^2}{\omega_d^2}$ and $\frac{\Omega_d^2}{\omega_d^2}$ are small and lead to trivial light shifts, and thus can be ignored. This leaves only the terms that arise from the dipole-quadrupole interaction, which are responsible for the primary dynamics of interest. After applying the RWA to drop terms that oscillate at $2\omega_0$ and $2\omega_d$ and evaluating the time integrals, the propagator is evaluated as

$$\hat{U} = \exp \left[i\Omega_q \Omega_d \frac{\omega_0}{\omega_d^2 - \omega_0^2} \frac{\sin(\delta t - \varphi_d) + \sin(\varphi_d)}{\delta} (\hat{a} + \hat{a}^\dagger) \right] = \hat{D}(\alpha) \quad (7.7)$$

which is the form of a displacement operator with

$$\alpha = i\Omega_q \Omega_d \frac{\omega_0}{\omega_d^2 - \omega_0^2} \frac{\sin(\delta t - \varphi_d) + \sin(\varphi_d)}{\delta}. \quad (7.8)$$

In the $\delta t \rightarrow 0$ limit, Eq. (7.8) reduces to

$$\alpha = i\Omega_q \Omega_d \frac{\omega_0}{\omega_d^2 - \omega_0^2} t \cos(\varphi_d). \quad (7.9)$$

If we consider an imbalance of the phases of the quadrupole tones, defining the RSB (BSB) phase as φ_r (φ_b), then the Hamiltonian of the ion motion is

$$\begin{aligned} \frac{\hat{H}}{\hbar} = & \omega_0 \hat{a}^\dagger \hat{a} + \Omega_d (\hat{a} + \hat{a}^\dagger) \cos(\omega_d t + \varphi_d) \\ & + \frac{\Omega_q}{2} (\hat{a} + \hat{a}^\dagger)^2 (\cos((\omega_d + \delta + \omega_0)t + \varphi_b) + \cos((\omega_d + \delta - \omega_0)t + \varphi_r)). \end{aligned} \quad (7.10)$$

The resulting propagator is a displacement operator with

$$\alpha = i\Omega_q \Omega_d \frac{\omega}{\delta(\omega_d^2 - \omega^2)} e^{i\varphi_-} (\sin(\delta t - \varphi_d + \varphi_+) + \sin(\varphi_d - \varphi_+)) \quad (7.11)$$

where $\varphi_{\pm} = \frac{\varphi_b \pm \varphi_r}{2}$. Working in the near-resonance condition, $\delta t \ll 1$, the displacement becomes

$$\alpha = i\Omega_q\Omega_d \frac{\omega_0 t}{\omega_d^2 - \omega_0^2} \frac{e^{i(\varphi_d - \varphi_r)} + e^{-i(\varphi_d - \varphi_b)}}{2}. \quad (7.12)$$

The first (second) exponential in equation 7.12 is the phase term corresponding to the red (blue) sideband tone. Setting $\varphi_r = \varphi_b = 0$ leaves us with just a dependence on the signal's phase.

As the frequencies of the quadrupole tones are scanned across the dipole frequency, three peaks are observed in Fig. 7.2. Two peaks arise at $\delta \approx \pm 2\omega_0$ from a phase-independent interaction between the dipole tone and each quadrupole tone individually, while a central peak at $\delta \approx 0$ arises from the phase-dependent interaction between the dipole and both quadrupole tones, as shown in Fig. 7.7. While scalar spectrum analysis is therefore possible with only a single sideband tone, *vector* signal analysis requires the presence of all three tones to yield a phase dependence, and has the added benefit of a stronger interaction.

A similar Magnus expansion should be considered even when the dipole tone is not present. High-power quadrupole tones can produce intrinsic squeezing of the QHO, which will create background noise when trying to detect small displacements. Consider the effect of the two quadrupole tones in isolation, i.e. $V_d = 0$. For simplicity, choose $\varphi_r = \varphi_b = 0$. The interaction Hamiltonian of the ion motion is

$$\frac{\hat{H}_I}{\hbar} = \Omega_q \left(\hat{a}^2 e^{-2i\omega_0 t} + 2 \left(\hat{a}^\dagger \hat{a} + \frac{1}{2} \right) + \hat{a}^{\dagger 2} e^{2i\omega_0 t} \right) \cos((\omega_d + \delta)t) \cos(\omega_0 t). \quad (7.13)$$

For similar reasons to those above, we only need to consider the second-order Magnus term and apply the RWA to find the time evolution operator

$$\hat{U} \approx \exp \left[\frac{i\Omega_q^2 t}{2} \left(\frac{\omega_0}{\omega_d^2 - \omega_0^2} \right) (\hat{a}^2 + \hat{a}^{\dagger 2}) \right] = \hat{S}(\xi) \quad (7.14)$$

which is the form of a squeezing operator with $\xi \equiv -i\Omega_q^2 \left(\frac{\omega_0}{\omega_d^2 - \omega_0^2} \right) t$.

We now consider the interaction of all three tones in full. Combining equations 7.7 and 7.14 gives the second order Magnus term:

$$\hat{A}_2 = (\alpha \hat{a}^\dagger - \alpha^* \hat{a}) + \frac{1}{2} (\xi^* \hat{a}^2 - \xi \hat{a}^{\dagger 2}) \quad (7.15)$$

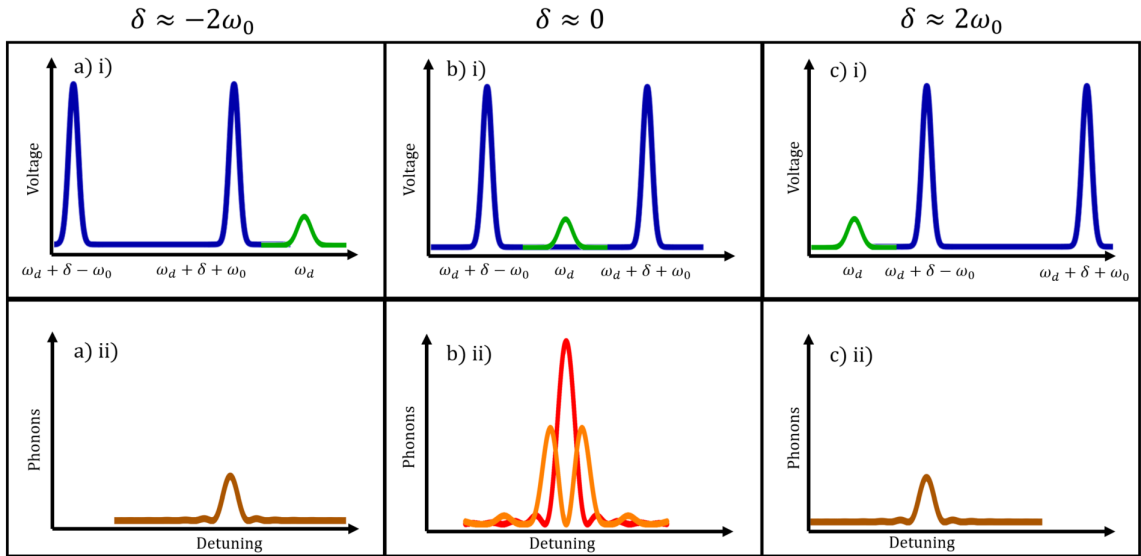


Figure 7.2: (a-c, i) Two quadrupole tones (blue) are swept in frequency across a weaker dipole tone (green). (a-c, ii) The resulting mean phonon number $\langle n \rangle$ produced from the corresponding interaction in (i). (a, i-ii) The interaction is primarily determined by that between the blue quadrupole tone at $\omega_d + \delta + \omega_0$ and the dipole tone at ω_d . This interaction is phase-independent. (b, i-ii) Both quadrupole tones at $\omega_d + \delta \pm \omega_0$ interact with the dipole tone at ω_d . The resulting interaction is phase-dependent, and can produce a resonance (red) when the phase relation satisfies $\varphi_d - \varphi_+ = 0$, where $\varphi_+ = \frac{\varphi_b + \varphi_r}{2}$, or an anti-resonance (orange) when $\varphi_d - \varphi_+ = \frac{\pi}{2}$. (c, i-ii) In contrast to (a), the interaction is primarily determined by that between the red quadrupole tone at $\omega_d + \delta - \omega_0$ and the dipole tone at ω_d , and is phase-independent.

where α is defined in Eqn. 7.9.

Using the Zassenhaus relation, the propagator of the interaction can be expressed as

$$\hat{U} = \hat{S}(\xi)\hat{D}\left(\sum_{j=1}^{\infty}\frac{i^j(-1)^{j+1}|\alpha||\xi|^{j-1}}{j!}\right) = \hat{S}(\xi)\hat{D}(\alpha') \quad (7.16)$$

$$\text{where } \alpha' \equiv \left|\frac{\alpha}{\xi}\right| (i \sinh |\xi| - \cosh |\xi| + 1). \quad (7.17)$$

The complete QVSA interaction thus consists of two parts: a displacement operator $\hat{D}(\alpha')$ and a residual squeeze operator $\hat{S}(\xi)$.

The resulting displacement α' is larger than that calculated in Eqn. 7.9 due to the intrinsic squeezing generated by the quadrupole tones, as described by Eqn. 7.14, an effect we refer to as *intrinsic amplification*. For small quadrupole voltages, i.e. as $\Omega_q/\omega_q \rightarrow 0$, the squeezing parameter becomes negligible, i.e. $\xi \rightarrow 0$, and thus $\alpha' \rightarrow \alpha$, reproducing equations 7.9 and 7.7. This is evident in Fig. 7.3 where $\langle n \rangle$ exhibits linearity in the low V_q regime, but becomes increasingly superlinear in the high V_q regime.

The residual squeezing results in an undesirable increase in the measurement background, which becomes particularly pronounced for longer measurement times (Fig. 7.3). However, the residual squeezing can be negated by applying an anti-squeezing operation $\hat{S}^\dagger(\xi)$ following the QVSA interaction (*intrinsic anti-squeezing*). In practice, this can be straightforwardly implemented by disabling the dipole tone and phase-shifting a single quadrupole tone by π .

7.2.2 Experimental Setup

Each QVSA experiment is comprised of four steps. A sample pulse sequence including quantum amplification is shown in Fig. 7.4. First, the ion is prepared in the motional ground state via sideband cooling. Second, the QVSA interaction is applied to induce a displacement α . Third, the motional state is projected onto the $\{S, D\}$ spin manifold via excitation on the first red/blue motional sideband for some given time t . This procedure can be expressed as a unitary operation $\hat{U}_{r/b}$. For sideband ratio measurements, we choose $t = t_\pi = \pi/\Omega_{01}$, where Ω_{01} is the Rabi frequency for the $|S, n=0\rangle$ to $|D, n=1\rangle$ transition. The spin state can in turn be read out via

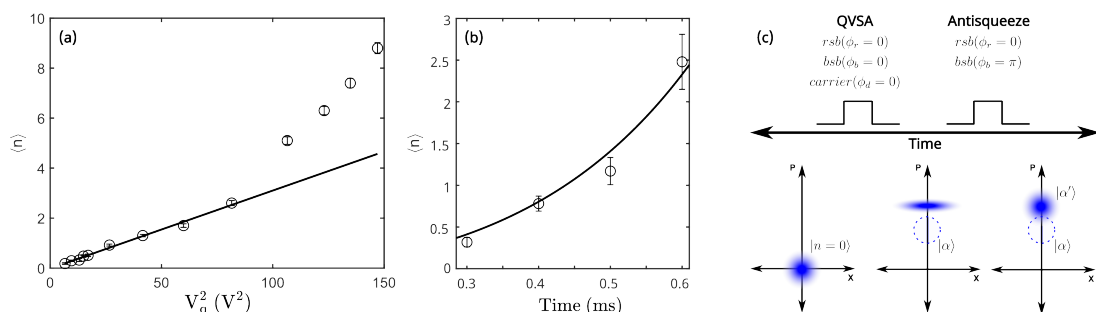


Figure 7.3: Onset of intrinsic amplification in the high V_q regime. (a) Heating from the QVSA interaction as a function of the quadrupole voltage V_q . The solid line is a linear fit to the data in the low V_q regime. The deviation of the data from the solid line in the high V_q regime arises from the intrinsic squeezing of the QVSA interaction, and can be leveraged to improve the measurement sensitivity. (b) Characterization of the intrinsic squeezing strength. The solid line is a fit using Eqn. 7.14 and yields a coupling strength $|z|/t \approx 2\pi \cdot 300$ Hz. Error bars represent one standard error. Pulse sequence for intrinsic amplification and associated Wigner functions. The dashed lines in the bottom panels depict a pure displacement to $|\alpha\rangle$ as would be expected. The QVSA pulse is comprised of a displacement operation followed by a squeeze operation, as illustrated by the colored ellipse. To convert from a squeezed state back to a coherent state $|\alpha'\rangle$ we antisqueeze by shifting the blue sideband phase by π .

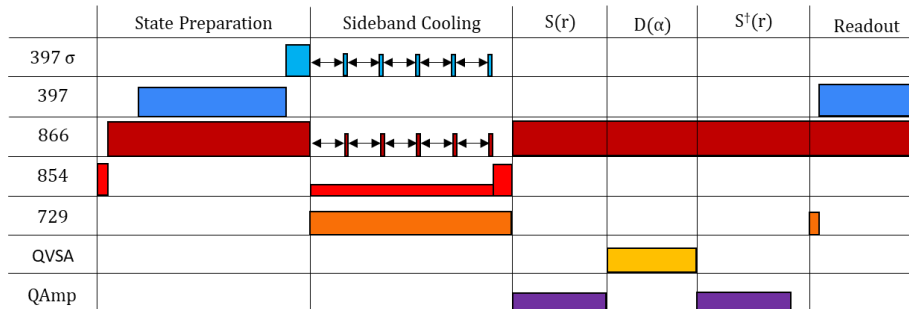


Figure 7.4: Pulse sequence for the QVSA with Quantum Amplification. The ion is Doppler cooled and optically pumped into the $|^2S_{1/2}, m_J = -\frac{1}{2}\rangle$ state. Continuous sideband cooling subsequently prepares the ion near the motional ground state $|n = 0\rangle$. Quantum Amplification is achieved by preparing the ion in a squeezed state before applying the QVSA pulse, which amplifies the displacement and minimizes uncertainty along the squeezing axis. The squeezing is then reversed via an anti-squeezing pulse, returning the ion to a coherent state.

state-dependent fluorescence, which is described by the projection operators $\{\hat{\Pi}_\mu\}$ where $\mu \in \{0 \text{ (dark)}, 1 \text{ (bright)}\}$. The final D-state probability can thus be expressed as $P_{r/b} = P_{r/b}(\mu|\alpha) = \text{Tr}\{\hat{\Pi}_\mu \hat{U}_{r/b}^\dagger \hat{\rho}(\alpha) \hat{U}_{r/b}\}$. Fourth, the ratio of the red and blue sideband D-state populations ($R = P_r/P_b$) is converted to a displacement and a mean phonon number at different detunings of the quadrupole tones (relative to the dipole tone), assuming a coherent state. The resulting profile is fitted with a sinc^2 function to extract the displacement. However, for Allan deviations, we instead measure only a single point at the secular frequency resonance where $\delta_q = \omega_0$.

Signals for the QVSA interaction are generated using the Sinara “Phaser” Arbitrary Waveform Generator (AWG). The AWG has two coherent output channels which are used to separately address a pair of quadrupolar trap electrodes. Dipole tones (i.e. $\hat{x}$ terms) and quadrupole tones (i.e. $\hat{x}^2$ terms) can be simply generated from the same AWG by setting an inter-channel phase delay of π and 0, respectively.

In practice, the QVSA signals are applied to excite motion *perpendicular* to the QVSA electrodes (i.e. along the y mode), which is made possible due to imperfections in trap geometry, though the effective dipole voltage is reduced by an experimentally determined geometric factor $\kappa = 1/42$. This is desirable since the x mode experiences a higher motional heating rate as a result of the electronics connected to the QVSA

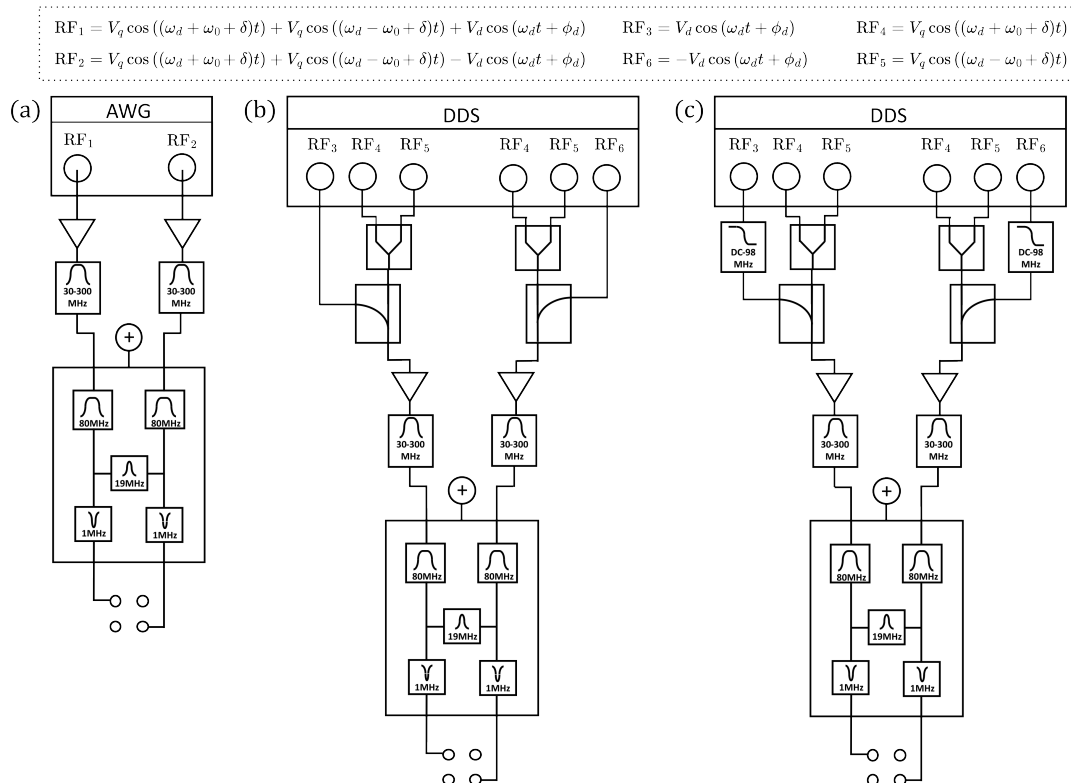


Figure 7.5: Example RF schematic of the QVSA signal generation apparatus. (a-c) Signals are amplified (Mini-Circuits ZHL-20W-13SW+) before filtering and DC biasing (to break the radial mode degeneracy). A narrow 19 MHz band-pass filter between the QVSA electrodes shorts any pickup from the 19 MHz trap electrodes, which improves trap stability. Different combinations of filters can be used for different frequency ranges. (a) Dipole and quadrupole tones are generated with two channels of an Arbitrary Waveform Generator (Sinara “Phaser”) by setting an inter-channel phase delay of π and 0, respectively. (b-c) QVSA signals are generated using two banks of synchronously clocked Direct Digital Synthesizers (DDSs) (Sinara AD9910 Urukul). This configuration allows the QVSA technique to function as a vector network analyzer (VNA) as the dipole tones can be separately passed through a Device Under Test (DUT).

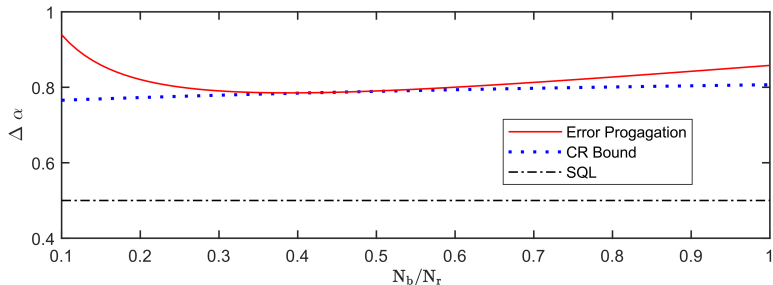


Figure 7.6: Uncertainty in the measured displacement $\Delta\alpha$ as a function of the relative number of red and blue sideband measurements used for the phase measurement in Fig. 7.10(b) in the main text. The Cramér-Rao bound (CRB) is saturated when $N_b/N_r \approx 0.4$. Experimental imperfections are included in the estimation.

electrodes.

Phase delays and latencies between the signals applied to the QVSA electrodes change the multipole moments of the field and can thus reduce the QVSA interaction strength and the motional coherence time. However, these can be simply calibrated by sweeping the inter-electrode phase delay at the RF source to maximize the resulting displacement.

Similarly, the QVSA signals can be aligned in phase space (Fig. 7.7) by sweeping the phase of a single quadrupole tone (e.g. φ_r , the phase of the red quadrupole tone) to maximize the resulting displacement. From equation 7.11, maximal displacement occurs when $\varphi_d = \varphi_+ = \frac{\varphi_r + \varphi_b}{2}$, where φ_r and φ_b are the red and blue quadrupole phases, respectively.

Calibration of the quadrupole amplitudes is achieved by applying a dipole tone together with a single quadrupole tone and equalizing the resulting displacement. The quadrupole voltages can in turn be determined absolutely by applying the two quadrupole tones at $\omega_d \pm \omega_0$ without the dipole tone to generate a motional squeezed state, from which the squeezing coupling strength (and thus the voltage) can be extracted.

7.2.3 State Readout

To date, the most sensitive measurement technique for displacement sensing is the overlap technique [WSH19], which employs Fock states to measure the expected state overlap $|\langle n | \hat{D}(\alpha) | n \rangle|^2$. However, due to the technical complexity of the overlap technique, we instead employ two more straightforward and well-established detection techniques: sideband ratio measurement and blue sideband (BSB) Rabi oscillations.

The sideband ratio measurement is preferred when the motional state is known to be coherent due to its high Fisher Information (Fig. 3.8). For other or unknown motional states, BSB Rabi oscillations are preferred due to their generality and ability to distinguish between types of motional states. The sideband ratio technique compares the excitation probabilities of the first red and blue sidebands. This technique is technically simple and offers immunity to systematic sources of error (e.g. imperfect state preparation, qubit frequency drifts, 729nm power fluctuations). However, the sideband ratio technique is limited to measurements of $\langle n \rangle > 1$, and is statistically inefficient when applied to squeezed states (Fig. 3.6 (c)).

Motional detection via BSB Rabi oscillations is accomplished by fitting the oscillations to their theoretical form to extract the relevant state parameter and the Fock state populations [MMK96a, LBM03, LKC15]. In contrast to the sideband ratio technique, this technique offers dynamic range and generality at the cost of lower Fisher Information and concomitant sensitivity: it can be used for measurements of $\langle n \rangle > 1$ and can be applied to an arbitrary motional state. We use this method specifically for quantum amplification data, which often results in $\langle n \rangle > 1$ and involves characterization of squeezed states. The BSB coherence time is $\approx 1400 \mu s$, which is limited by imperfect control, e.g. servo bumps [ANK15] and laser power fluctuations of the qubit addressing beam, and systematic drifts, e.g. secular frequency drifts and qubit frequency fluctuations.

In parameter estimation, the Cramér-Rao Bound (CRB) sets a lower bound on the achievable variance of an estimator $\Delta\alpha$ in terms of its Fisher Information F , i.e. $\Delta\alpha \geq \Delta\alpha_{CR} = \frac{1}{\sqrt{F}}$. Classically, the Fisher Information is calculated as

$$F(\alpha) = \sum_x \frac{1}{P(x|\alpha)} \left(\frac{\partial P(x|\alpha)}{\partial \alpha} \right)^2 \quad (7.18)$$

which is additive for repeated independent measurements. The maximal F over the estimator space is called the quantum Fisher information ($\mathfrak{F}$), which in turn leads to the quantum Cramér-Rao bound (QCRB). Here, we show that the CRB of our sideband ratio measurement can be saturated by adjusting the relative number of red and blue sideband measurements used to constitute R .

We define the inversion estimator [XCY19] $\hat{\alpha} = f^{-1}(R)$ to evaluate the uncertainty of the displacement α given that $R = P_r/P_b = f(\alpha)$. Assuming that the uncertainties of the red and blue sideband probabilities $\Delta P_{r/b}$ are dominated by quantum projection noise (which follows a binomial distribution and is thus $\Delta P_{r/b} = \sqrt{p_{r/b}(1 - p_{r/b})/N}$), propagation of uncertainty gives the uncertainty of the normalized inversion estimator (per experiment) as

$$\Delta\hat{\alpha} = \sqrt{\frac{1}{F_r N_r} \left(\frac{\partial\alpha}{\partial R}\right)^2 \left(\frac{\partial R}{\partial P_r}\right)^2 \left(\frac{\partial P_r}{\partial\alpha}\right)^2 + \frac{1}{F_b N_b} \left(\frac{\partial\alpha}{\partial R}\right)^2 \left(\frac{\partial R}{\partial P_b}\right)^2 \left(\frac{\partial P_b}{\partial\alpha}\right)^2} \sqrt{N_r + N_b} \quad (7.19)$$

where P_r (P_b) is the D-state population after a red (blue) sideband pulse, N_r (N_b) is the number of red (blue) sideband measurements, and F_r (F_b) is the Fisher Information per experiment for a red (blue) sideband measurement. The uncertainty of a binomial distribution with parameter p and repetitions N (also called quantum projection noise) is $\Delta P = \sqrt{p(1 - p)/N}$. To account for imperfections in the experimental procedure, we use an empirical formula $P_{r/b}^{real} = A P_{r/b}(\alpha) + B$, where $A \approx 0.94$ and accounts for decoherence processes, e.g. laser power fluctuations, while $B \approx 0.03$ and accounts for background noise, which for our experiment is dominated by the servo bump of the qubit addressing laser.

For comparison, the CRB per experiment calculated from Eq. (??) is:

$$\Delta\alpha_{CRB} = \sqrt{\frac{N_r + N_b}{N_r F_r + N_b F_b}}. \quad (7.20)$$

Fig. 7.6 shows both the CRB and $\Delta\hat{\alpha}$ alongside the SQL as a function of N_b/N_r for $\alpha \approx 0.68$. The estimator performance is maximized for $N_b/N_r \approx 0.4$, where it also saturates the CRB.

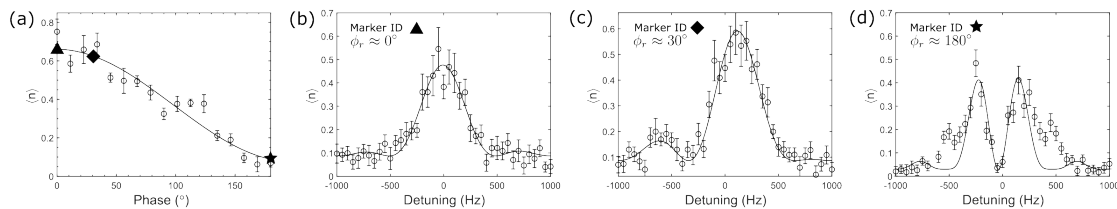


Figure 7.7: Phase dependence of the spectrum sweep procedure. The phase relation between the QVSA tones fundamentally changes the lineshape of the resonance. By sweeping just φ_r , the phase dependence is reduced to $\langle n \rangle \propto \cos^2(\frac{\varphi_r}{2})$. Panel (a) shows a phase sweep at the resonance (i.e. $\delta = 0$), with three characteristic phases marked by a triangle (b), a diamond (c), and a star (d). Voltage parameters vary between marked points. (d) illustrates the production of an antiresonance, which occurs when the red sideband is completely out of phase with the blue sideband (see Fig. 1(c) in the main text for a graphical representation). The amplitude discrepancy between (a) and (b-d) is due to a difference in the applied QVSA voltages. Error bars represent one standard error. The solid lines are fitted lines using equation 7.11.

7.2.4 Results

This technique was studied to understand its phase-sensitivity behavior, its wide-band applications, its compatibility with quantum enhancement methods, and its practical uses. Most frequency sensors that operate based off generating displacements typically produce a single displacement which is unable to provide information about the phase. Certain schemes, like Wigner reconstruction, are able to measure the phase by scanning phase space, but face a serious drawback in terms of the rate of data collection. Additionally, as was mentioned, when a signal tone moves too far from a sensor's resonance, the performance sharply drops. The heterodyne process of this method allows for a scannable range significantly larger than other quantum sensors. Quantum enhancement via squeezing is demonstrated to be compatible with this method, offering new avenues for improving sensitivity. Finally, we get to examine how this scheme has allowed us to reevaluate just what is needed to successfully implement EGGs in our system.

7.2.4.1 Phase Sensitivity

By applying both quadrupole tones, we introduce two effective displacements, one by the beating of the RSB and the carrier, and one by the BSB and the carrier. In phase space, we can imagine they undergo vector addition to create an overall displacement given by equation 7.12 which includes the phases of all three tones. For practical applications, we want to constrain $\varphi_r = \varphi_b = 0$, but we can see that if the phases of all the tones are unknown, this is challenging. When near resonance, the phase that can be measured by optimizing the displacement is given by $\varphi_d - \varphi_+$. Peak displacement occurs when

$$\varphi_d = \frac{\varphi_r + \varphi_b}{2} \quad (7.21)$$

which indicates that we may not measure the absolute phase of the dipole if the sidebands are imbalanced. Our measurement only provides information about one of the phases relative to the other two. Fig. 7.7 shows what happens when one of these phases is swept and the others are held constant. To properly calibrate the phase of each tone, we should introduce a constant displacement followed by displacement generated just by the carrier and a single sideband. Tickling on a given mode, can easily generate the constant displacement. Starting with the RSB, optimizing the displacement strength by sweeping the RSB phase will allow us to know it relative to the the phase of the tickle and the carrier. Repeating the measurement with the BSB allows us to determine the BSB phase relative to the tickle and carrier. Since the tickle and carrier phases are held constant during this calibration step, we are able to set $\varphi_r = \varphi_b$. Any QVSA test with both quadrupoles present will be able to measure the phase of an unknown tone.

7.2.4.2 Wideband Sensing

Demonstration of this method for wideband vector signal analysis of an unknown dipole tone $(V_d, \omega_d, \varphi_d)$ involved testing the performance of equation 7.8 at three different frequencies spread out orders of magnitude apart. As mentioned earlier, the QHO is first prepared in the vacuum state $|0\rangle$. Next, the quadrupole tones are applied with amplitude Ω_q and detuning δ for time t , creating a coherent state $|\alpha\rangle$.

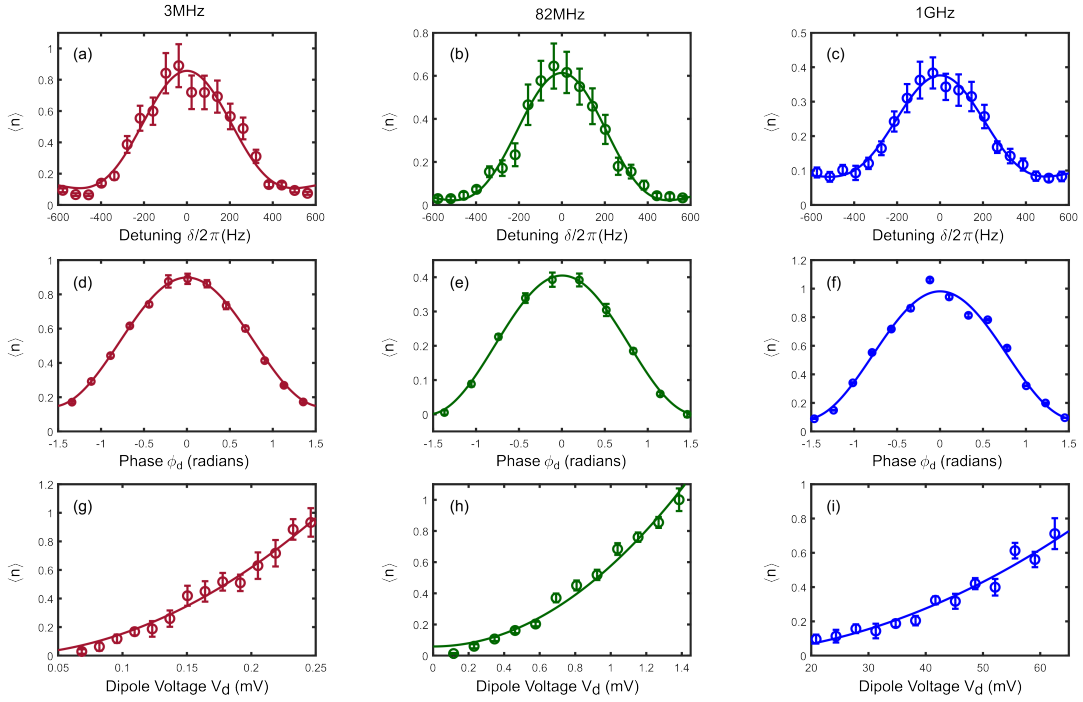


Figure 7.8: Wideband demonstration of frequency, phase, and amplitude sensing using the QVSA. (a)-(c) Dipole signals are detected by sweeping a pair of quadrupole tones across $\omega_d/2\pi = 3\text{MHz}$, 82MHz , and 1GHz , respectively. The lineshape $\langle n \rangle = |\alpha|^2$ is obtained by varying the detuning δ . (d)-(f) illustrate the dependence of $\langle n \rangle$ on ϕ_d for 3MHz , 82MHz , and 1GHz respectively. The phase dependence is measured with detuning $\delta = 0$. The quadratic dependence of $\langle n \rangle$ on V_d is measured in (g)-(i). The combination of frequency, phase, and amplitude sensing provides the means for vector signal analysis. Solid lines are fit using Eqn. 7.8 to extract voltages experienced by the ion. Error bars represent one standard error. Exact parameters used for each panel are listed in Tab. 7.1.

Subfigure	κV_d	V_q	Time	Pulse Shaping?
(a)	$(0.239 \pm 0.002) \text{ mV}$	$(8.5 \pm 0.1) \text{ mV}$	1ms	Yes
(b)	$(0.489 \pm 0.006) \text{ mV}$	$(2.88 \pm 0.03) \text{ V}$	1ms	No
(c)	$(42 \pm 3) \text{ mV}$	$(3.8 \pm 0.2) \text{ V}$	1ms	No
(d)	$(0.226 \pm 0.002) \text{ mV}$	$(9.17 \pm 0.01) \text{ mV}$	1ms	Yes
(e)	$(2.20 \pm 0.02) \text{ mV}$	$(0.515 \pm 0.005) \text{ V}$	1ms	No
(f)	$(0.31 \pm 0.02) \text{ V}$	$(0.84 \pm 0.05) \text{ V}$	1ms	No
(g)	See Figure	$(8.62 \pm 0.09) \text{ mV}$	1ms	Yes
(h)	See Figure	$(1.36 \pm 0.02) \text{ V}$	1ms	No
(i)	See Figure	$(3.9 \pm 0.3) \text{ V}$	1ms	No

Table 7.1: Table of parameters for Fig. 2

The mean phonon number $\langle n \rangle = |\alpha|^2$ of the QHO is measured from the relative intensity of the motional sidebands of the optical qubit transition. This process is repeated as the detuning δ is scanned to maximize the displacement and thus determine the unknown frequency ω_d (Fig. 7.8(a)-(c)). Once ω_d is determined, φ_d is found by repeating the above protocol at $\delta = 0$ while varying the phase of the applied quadrupole tones. Illustrated in Fig. 7.8(d)-(f), the displacement is maximized when $\varphi_d = 0$. With ω_d and φ_d determined, measurement of $\langle n \rangle$ is used to determine V_d . A similar measurement scanning the dipole voltage V_d is shown in Fig. 7.8(g)-(i).

Arguably the biggest selling point of this technique is its wideband sensitivity, which is noticeably larger than other previous quantum sensing techniques. Regardless, we demonstrate wideband operation, characterizing the electric field sensitivity at additional frequencies from 100 kHz to 1 GHz (Fig. 7.9), a frequency range $> 800\times$ larger than previous techniques. Performance closely matches that expected from equation 7.9 (solid red line). To remove the frequency dependence on the region of interest (ω_d) that quadratically reduces the strength of the displacement, equation

7.8 can be instead expressed in terms of the Mathieu q parameter, as shown by

$$\alpha = \beta \frac{q_q V_d}{r_0 \sqrt{m \omega_0}} \frac{\sin(\delta t - \varphi_d) + \sin(\varphi_d)}{\delta} \quad (7.22)$$

where β is a constant prefactor and $q_q = \frac{2eV_q}{mr_0^2\omega_q^2}$ for $\omega_q \approx \omega_d \gg \omega_0$ or $q_q = \frac{2eV_q}{mr_0^2(\omega_d^2 - \omega_0^2)}$ for $\omega_d \approx \omega_0$, which the subscript q indicates this is the Mathieu q parameter for the quadrupole tones. However, practical limitations constrain selection of q_q . Empirical results suggest that $q_q \leq 0.01$ is required in our experiment to prevent disturbance of trap dynamics, which limits the achievable V_q , and therefore sensitivity, at frequencies $\omega_d \lesssim 20$ MHz. For $\omega_d \gtrsim 20$ MHz, the voltage V_q required to maintain $q_q = 0.01$ exceeds component damage thresholds (28 dBm) of our experiment. Our practical inability to maintain $q_q = 0.01$ at higher frequencies causes a concomitant deterioration of sensitivity, reflected in the change in slope above 20 MHz in Fig. 4. Importantly, this limit is not fundamental. An additional power consideration comes from the intrinsic squeezing, which goes as Ω_q^2 whereas the QVSA effect goes as Ω_q , resulting in background noise that will eventually force us to increase the signal voltage (if possible), or to increase the probing time which is linear for both the intrinsic squeezing and the QVSA effect. Phase sensitivity is maintained until the $\omega_d \lesssim \omega_0$ regime, where only the blue quadrupole tone can be applied, preventing the interference that generates phase sensitivity. However, phase sensitivity could be recovered by interfering the QVSA displacement with a resonant displacement at ω_0 .

To contextualize the QVSA performance, Fig. 7.9 additionally compares electric field sensitivity across various quantum sensing techniques. Ref.3 [QHV22] reports phase-insensitive electric field measurements in NV centers from roughly 200 kHz to 1 MHz. Both Ref.2 [WSH19] and Ref.1 [GAL21] are phase-insensitive, using 150 entangled ions in a Penning trap and a single ion in a Paul trap, respectively, and are limited to sensing at the oscillator's secular frequency. Ref.6 [AGJ20] demonstrates the ability to perform highly sensitive measurements of the electric field strength, even for fields detuned by $\sim$ MHz from the oscillator's secular frequency, with phase sensitivity. While Ref. [MCC20] predicts that Rydberg atoms theoretically provide a tunable frequency range from kHz to THz, to our knowledge, the only experimental demonstrations within the MHz to GHz range are Ref.4 [LZL22] and Ref.5 [HPA22],

which are phase-insensitive with frequency ranges of 10 MHz and 4 MHz, respectively, both shown in Fig. 4. In comparison, the QVSA is the only method with phase sensitivity, demonstrating a frequency range across the low-frequency RF to UHF microwave bands, and sensitivities competitive with more specialized techniques.

Several straightforward modifications would significantly improve the QVSA performance. These include using a lighter mass ion and smaller ion-electrode distance (e.g. Be^+ with a surface trap [MKB19]), as well as higher damage threshold electronics - these form the red dotted line in Fig. 7.9. Longer motional coherence times would enable greater quantum amplification for greater improvements in sensitivity, denoted as the red dashed line in Fig. 7.9. Use of other non-classical motional states [HLJ13, JWN17, MMK96b, WSH19] and a low-noise pre-amplifier would yield a frequency sensitivity of $\approx 0.3 \text{ Hz}/\sqrt{\text{Hz}}$, a phase sensitivity of $\approx 3 \text{ mrad}/\sqrt{\text{Hz}}$, and an amplitude sensitivity of $\approx 0.2 \text{ pV}/\sqrt{\text{Hz}}$ ($2.8 \mu\text{V m}^{-1}/\sqrt{\text{Hz}}$).

Carrier frequency (MHz)	E-field sensitivity ($\frac{\text{mV}}{\text{m}\sqrt{\text{Hz}}}$)	Phase resolution (mrad)
0.1	8.3(1.1)	NA
0.6	2.1(3)	NA
3	2.2(4)	9.4(7)
30	8.1(1.7)	9.9(1.2)
100	66(14)	15.7(2.5)
200	$4.4(8) \cdot 10^2$	8.6(1.3)
350	$1.3(3) \cdot 10^3$	22.8(10.2)
600	$2.5(5) \cdot 10^3$	9.3(1.7)
800	$3.6(8) \cdot 10^3$	9.6(2.9)
1000	$1.0(2) \cdot 10^4$	11.7(2.2)

Table 7.2: The list of measurements shown in Fig 7.9. All values listed in the table are derived from an Allan deviation following the same procedure used in Fig. 7.9

7.2.4.3 Comparison to SQL

The standard quantum limit (SQL) of a measurement is the minimum achievable uncertainty when using classical states. For displacement sensing, the SQL is $\Delta\alpha_{\text{SQL}} =$

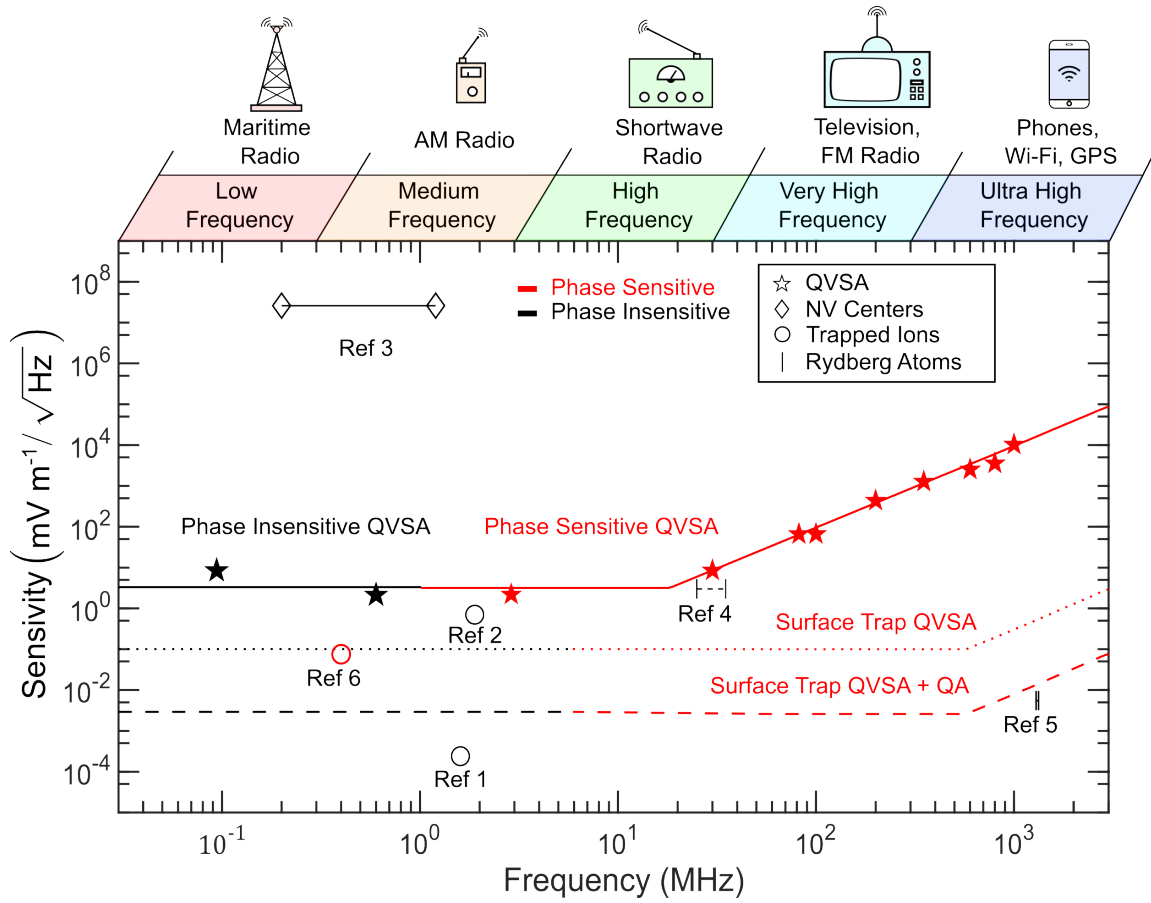


Figure 7.9: Comparison of electric field sensitivity between different quantum sensing techniques. Phase-insensitive methods are shown in black; phase-sensitive methods are shown in red; $\diamond$: NV centers; $\circ$: ions; $|$: Rydberg atoms; $\star$: our measurement. The length of lines corresponds to the frequency range of operation. The solid red line represents the estimated QVSA performance in our linear trap, which aligns closely with our measurements (exact measurement results are listed in Tab. 7.2). Quadrupole tones are applied with a constant Mathieu q -parameter of $q_q \approx 0.01$ to yield a nearly constant sensitivity from 100 kHz until ≈ 20 MHz. Above ≈ 20 MHz, practical limitations on power prevent the constant Mathieu q -parameter of $q_q = 0.01$ from being applied, causing the minimum achievable sensitivity to increase with frequency. The dotted red line indicates the estimated QVSA sensitivity if implemented on a surface trap with an ion-electrode distance of $40\mu\text{m}$, $^9\text{Be}^+$ as a trapped ion, a secular frequency of $2\pi \cdot 6$ MHz, and a maximum quadrupole power of 39.5dBm; The dashed red line represents anticipated near-term enhanced QVSA sensitivity in a surface trap with quantum amplification (QA). This entire comparison did not account for the scaling with the number of ions.

$\frac{1}{2}$ [WSH19, GAL21]. The SQL for the mean phonon number $\langle n \rangle$ is then given by

$$\Delta n_{\text{SQL}} = \frac{\Delta \alpha_{\text{SQL}}}{\frac{\partial \alpha}{\partial n}} = \sqrt{n}. \quad (7.23)$$

For frequency sensing with Rabi spectroscopy, the mean phonon number $\langle n \rangle$ expressed in terms of the dipole tone detuning δ is

$$\langle n \rangle = |\tilde{a}|^2 \frac{\sin^2(\delta t)}{(\delta t)^2} \quad (7.24)$$

where $|\tilde{a}|$ is the displacement amplitude and t is the QVSA interaction time. Using Eqn. 7.23, the SQL becomes $\Delta n = |\tilde{a}| \sin(\delta t) / (\delta t \sqrt{N})$, where N is the number of experiments. The frequency uncertainty of the dipole tone $\Delta \delta$ at the SQL is therefore:

$$\Delta \delta = \frac{\Delta n}{\frac{\partial n}{\partial \delta}} = \frac{(\delta t)^2}{\delta t^2 \cos(\delta t) - t \sin(\delta t)} \frac{1}{2|\tilde{a}| \sqrt{N}}. \quad (7.25)$$

Unlike in Ramsey spectroscopy, where the uncertainty is independent of detuning [IBB93], Rabi spectroscopy measurements require judicious selection of the read-out points. For the Allan deviation in Fig. 7.10(a), we use two points near the half-maxima, which balances the sensitivity with the signal-to-noise ratio (SNR). Under these conditions, $\sin(\delta t) / (\delta t) \approx 1/\sqrt{2}$. The SQL for frequency sensing is thus:

$$\Delta \delta \approx 2\pi \frac{0.209}{|\tilde{a}| t \sqrt{N}}. \quad (7.26)$$

Following Eqn. 7.24, the mean phonon number $\langle n \rangle$ expressed in terms of the dipole tone phase φ_d is:

$$\langle n \rangle = |\tilde{a}|^2 \cdot \sin^2(\vartheta + \varphi_d), \quad (7.27)$$

where ϑ is the variation of the initial phase. At the SQL, this becomes $\Delta n = |\tilde{a}| \cdot \sin(\vartheta + \varphi_d) / \sqrt{N}$, which allows us to calculate the phase uncertainty at the SQL:

$$\Delta \varphi_d = \frac{\Delta n}{\frac{\partial n}{\partial \varphi_d}} = \frac{1}{\cos(\vartheta + \varphi_d)} \frac{1}{2|\tilde{a}| \sqrt{N}}. \quad (7.28)$$

Similarly, for the Allan deviation in Fig. 7.10(b), two points are measured at the half-maxima (i.e. $\cos(\vartheta + \varphi_d) = 1/\sqrt{2}$). The SQL for phase sensing is thus:

$$\Delta \varphi_d = \frac{1}{|\tilde{a}| \sqrt{2N}}. \quad (7.29)$$

The mean phonon number $\langle n \rangle$ expressed in terms of the dipole tone amplitude V_d is:

$$\langle n \rangle = AV_d^2 t^2, \quad (7.30)$$

where $A = 1.1 \times 10^{13} \text{ V}^{-2}\text{s}^{-2}$ is a prefactor (measured using $V_q = 8.4\text{V}$ at 82MHz). The SQL is thus:

$$\Delta V_d = \frac{\Delta n}{\frac{\partial n}{\partial V_d}} = \frac{1}{2t\sqrt{AN}}, \quad (7.31)$$

where N is the number of experiments.

The relationship between voltage (V_d) and electric field (E_d) is given by $E_d = 0.715V_d/r_0$, as determined using COMSOL Multiphysics.

7.2.4.4 Quantum Amplification

Quantum enhancement can be generated using non-classical states to improve the sensitivity of a measurement. For this experiment, we implemented a specific type of enhancement called quantum amplification, which leverages squeezing and anti-squeezing operations to increase the strength of a displacement operator. By parametrically modulating our trap potential, we can easily introduce a squeezing operation with a short pulse [BSB19]. The strength of the squeezing operation is given by $\xi = re^{i\theta}$. Quantum amplification is generated by squeezing the vacuum state before the application of the QVSA interaction. Then, an anti-squeezing pulse of the same strength and π phase offset relative to the squeezing operator is applied to reverse the squeezing operation, returning the ion to a coherent state with an amplified displacement given by

$$\hat{S}^\dagger(\xi)\hat{D}(\alpha)\hat{S}(\xi)|0\rangle = \hat{D}(e^r\alpha)|0\rangle. \quad (7.32)$$

Verification of the anti-squeezing fidelity was performed by measuring the phonon number after applying anti-squeezing immediately after a squeezing operation. State readout with the BSB Rabi flopping allows us to ensure that the final state properly

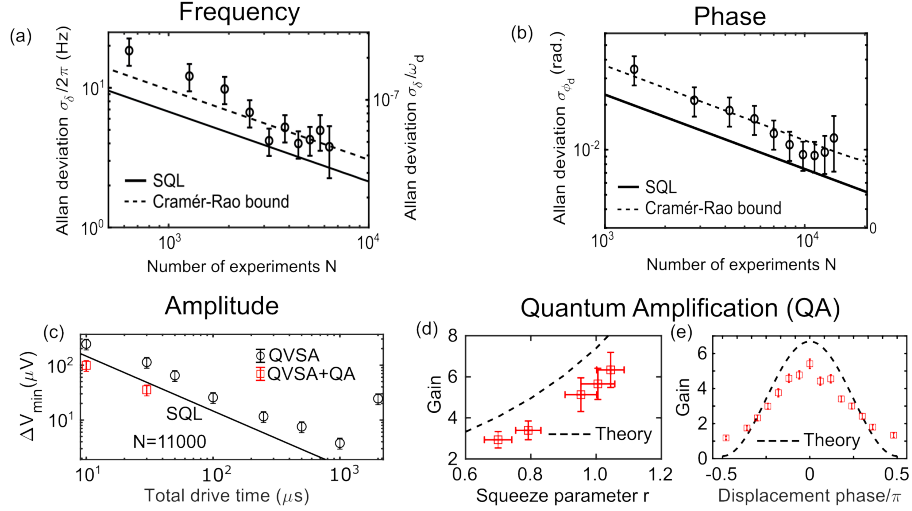


Figure 7.10: Frequency, phase and amplitude sensitivity of QVSA and integration of quantum amplification (QA). (a-b) Overlapping Allan deviation of frequency and phase of an 82 MHz dipole tone. (a) The measured frequency sensitivity is $39(9) \text{ Hz}/\sqrt{\text{Hz}}$ with a minimum resolution of $3.8(1.6) \text{ Hz}$ after 6300 experiments. (b) The measured phase sensitivity is $117(26) \text{ mrad}/\sqrt{\text{Hz}}$ with a minimum resolution of $9.1(2.0) \text{ mrad}$ after 9800 experiments. (a-b) The probe time is 2 ms and each experiment takes ~ 17 ms. Solid lines represent the corresponding standard quantum limit (SQL). Dashed lines represent the corresponding Cramér-Rao (CR) bounds, both of which are saturated by our measurements. (c-e) Integration of quantum amplification with the QVSA. (c) The minimum detectable voltage ($\Delta V_{\min}$) on the electrodes with and without quantum amplification. Each data point represents 11,000 experiments, each of which takes ~ 17 ms. The measured sensitivity to the voltage coupled onto the electrodes is $51(10) \mu V/\sqrt{\text{Hz}}$ with a minimum detectable voltage of $3.8(8) \mu V$. The electric field sensitivity is $66(13) \text{ mV m}^{-1}/\sqrt{\text{Hz}}$ with an electric field resolution of $4.9(1.0) \text{ mV/m}$. Notably, with QA the voltage sensitivity is $3.4(2.0) \text{ dB}$ and $2.8(2.0) \text{ dB}$ below the SQL for $10 \mu s$ and $30 \mu s$, respectively. (d-e) Quantum amplification through squeezing. By applying a squeeze operation $\hat{S}(r)$ before and an anti-squeeze operation $\hat{S}^\dagger(r)$ after $10 \mu s$ of QVSA operation, the mean phonon number $\langle n \rangle$ is amplified. The gain scales with the squeeze parameter r (d) and its phase relative to the QVSA displacement, with $r = 0.95$ (e). Dashed lines represent the maximum achievable gain from theory. Parametric squeezing is applied for $30 \mu s$ at roughly 75 mV . (a-e) Error bars represent one standard error.

fits to a vacuum state and residual squeezing is minimized. Using the maximum coupling strength $r/t = 2\pi \cdot 5.5(2)$ kHz applied for $30\mu\text{s}$, we recover $\langle n \rangle = 0.03(2)$, which is indistinguishable from the initial state. Fig. 7.10 shows quantum amplification of $\langle n \rangle$ as a function of r , with a maximum gain in $\langle n \rangle$ of 6.3(9). Maximal amplification requires orientation of the QVSA displacement along the squeezing axis in phase space (Fig. 7.10(e)). This is achieved by adjusting the phases of the quadrupole tones by equal and opposite amounts. Discrepancies between the achieved and predicted gains likely arise from phase instability between the squeezing and displacement operations. We characterize the voltage sensitivity in the manner previously described and find a metrological gain $10\log_{10}(\Delta V_{\text{QA}}/\Delta V_{\text{SQL}})^2 = 3.4$ (2.0) dB and 2.8(2.0) dB below the SQL for interaction times of $10\mu\text{s}$ and $30\mu\text{s}$, respectively (Fig. 3(c)). Our achievable amplification is currently limited by the increased motional decoherence of a squeezed state.

7.2.4.5 Comparison to VNA

The relevance of the QVSA technique is demonstrated through two examples. We demonstrate the functionality of the QVSA technique for vector network analysis by characterizing the transfer function of a commercial low-pass filter (Mini-Circuits SLP-100+). For this measurement, V_d and φ_d are measured as a function of frequency with and without the filter inserted. The RF configuration used for these measurements is shown in Fig. 7.5(b-c). For normalization, the transfer function of the dipole tone is measured without the Device Under Test (DUT) (i.e. the low-pass filters) in place (Fig. 7.5(b)). At each frequency, the RSB phase φ_r is scanned, and the resulting displacement is fitted to Eqn. 7.8 in the main text to extract the voltage (V_n) and phase (φ_n) of the measured signal, similarly as shown in Fig. 7.8. Next, DUTs are connected in series with the relevant dipole tone DDS channels (RF3 and RF6) respectively as shown in Fig. 7.5(c), which alters the amplitude and phase of the dipole field experienced by the ion. A similar measurement is repeated to extract the new voltage (V_f) and phase (φ_f) sensed by the ion. Finally, the phase delay from low-pass filters can be expressed as $(\varphi_f - \varphi_n)$ and attenuation can be written as $20\log_{10}(V_f/V_n)$. The data is shown in Fig. 7.11 alongside the measurement from a

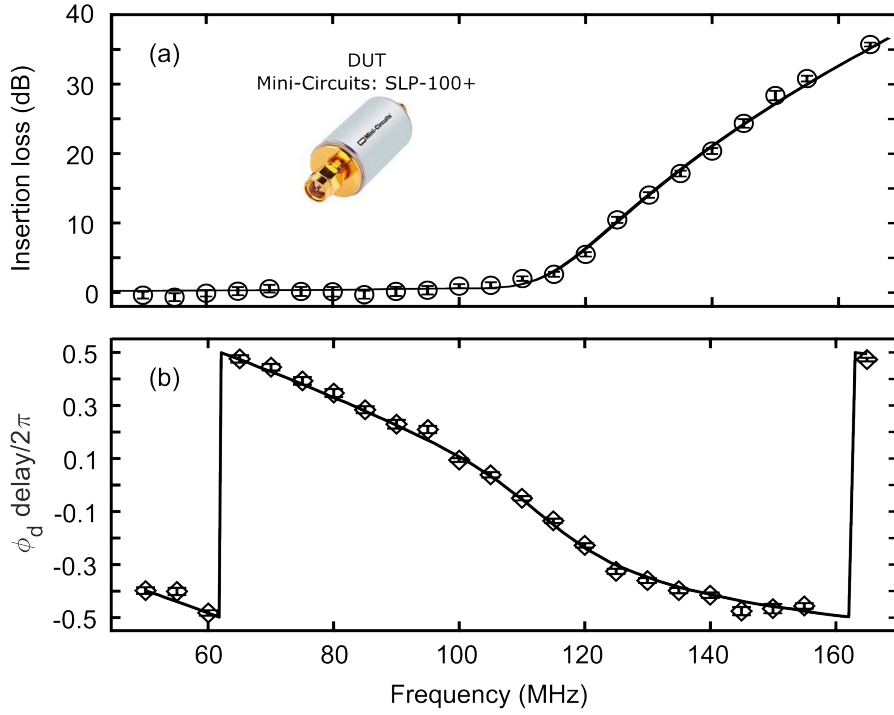


Figure 7.11: Transfer function of a commercial low-pass filter. (a-b) $\circ$ and $\diamond$ are measured through the QVSA. Solid lines are measured via a commercial vector network analyzer. Each point is composed of $N \approx 13500$ experiments. Error bars represent one standard error. No re-scaling or offset is applied to the data.

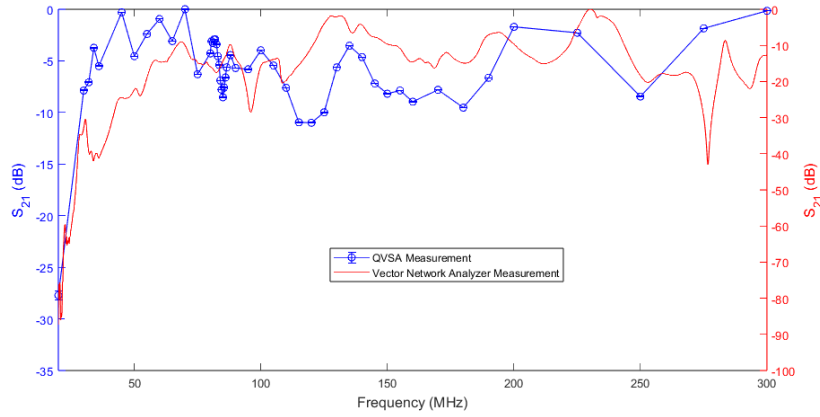


Figure 7.12: Insertion loss of the ion trap system measured using the QVSA and a commercial vector network analyzer (VNA). Blue (red) data shows the insertion loss measured by the QVSA (VNA). Error bars for the QVSA measurements represent one standard error and are too small to be seen.

standard network analyzer (Agilent 8714ES), which shows excellent agreement.

The same procedure was also used to calibrate the wideband transfer function of a qubit control line, shown in Fig. 7.12. The measured coupling efficiency is compared against that from a traditional measurement using a capacitor divider connected to trapping RF electrodes and a vector network analyzer over the frequency range accessible with our DDS (20 MHz - 300 MHz). As shown in Fig. 7.12, the traditional measurement fails to accurately report the filter function. Changes to the measurement conditions required to interface the network analyzer, e.g. impedance changes at measurement ports, likely account for this discrepancy, serving as an archetypal example of the difficulty of accurately calibrating qubit signals.

CHAPTER 8

Motional Raman: Subharmonics

An investigation into the useful field-sensing properties of subharmonic motional Raman terms was performed and published in [HH26]. The motional Raman process can be extended to higher order terms as more photons get involved within the process. Fig. 8.1 demonstrates how an even number of tones can produce a displacement, forming either a Jaynes-Cummings interaction as shown in (a) and (b), or an Anti-Jaynes-Cummings interaction in (c) and (d). These terms are characterized by how the signal and probe tones are applied to the trap electrodes, and by the relative frequencies of these tones. Each term will be able to generate a displacement, so only a single condition needs to be investigated to understand the dynamics. For example, Fig. 7.1 (b) indicates that for quadrupole sidebands and a carrier dipole, the blue sideband and the carrier produce a Jaynes-Cummings term and the red sideband and the carrier produce an Anti-Jaynes-Cummings term, and yet we have shown each will produce comparable displacements. We will go on to show that the motional Raman method investigated in the earlier chapter is the case of $K=2$, where K is the number of photons present in the subharmonic process. For generality, we will proceed by investigating a single one of these cases, and consider a signal tone at ω_s with voltage V_s and phase φ_s applied in the quadrupole configuration (i.e. a linear field gradient), and the probe tone applied at ω_p with both a dipole component (i.e. a uniform field) given by V_d and a quadrupole component given by V_q . The probe tone will be applied at $\omega_p = \omega_s + \frac{2\omega_0}{K} + \delta$, where ω_0 is the resonance frequency of the QHO mode and δ is a small detuning from the resonance. This would be case (c) in Fig. 8.1. The Hamiltonian of the ion motion under these fields is:

$$\begin{aligned} \frac{\hat{H}}{\hbar} = & \omega_0 \hat{a}^\dagger \hat{a} + \Omega_s (\hat{a} + \hat{a}^\dagger)^2 \cos(\omega_s t + \varphi_s) \\ & + \Omega_d (\hat{a} + \hat{a}^\dagger) \cos(\omega_p t) + \Omega_q (\hat{a} + \hat{a}^\dagger)^2 \cos(\omega_p t), \end{aligned} \quad (8.1)$$

where $\Omega_q = \frac{eV_q\eta^2}{\hbar}$, $\Omega_d = \frac{eV_d\eta}{\hbar}$, $\Omega_s = \frac{eV_s\eta^2}{\hbar}$, and η is the effective Lamb-Dicke parameter. The dipole tone Ω_d is included as it carries a lower order of η than quadrupole tones and modifies the motional Raman interaction to produce a displacement, rather than a squeeze operator. This is beneficial as coherent states are more easily manipulated and, given our readout procedure, provide significantly higher Fisher information than squeezed states. We can try solving this Hamiltonian using the Magnus expansion as we did in the K=2 motional Raman described in the previous chapter. However, instead we'll introduce the Floquet theory method to do so.

8.1 Floquet Theory of Subharmonics

In the interaction picture with respect to the bare QHO, the Hamiltonian given by Eqn. 9.1 becomes a sum of eight Hermitian components, each comprised of a factor Ω_i , ladder operators, and a time-dependent phase $e^{\pm i\omega_i t}$, where $\{\Omega_1 = \Omega_q, \omega_1 = \omega_p + 2\omega_0\}$, $\{\Omega_2 = \Omega_q, \omega_2 = \omega_p - 2\omega_0\}$, $\{\Omega_3 = \Omega_q, \omega_3 = \omega_p\}$, $\{\Omega_4 = \Omega_d, \omega_4 = \omega_p + \omega_0\}$, $\{\Omega_5 = \Omega_d, \omega_5 = \omega_p - \omega_0\}$, $\{\Omega_6 = \Omega_s, \omega_6 = \omega_s + 2\omega_0\}$, $\{\Omega_7 = \Omega_s, \omega_7 = \omega_s - 2\omega_0\}$, and $\{\Omega_8 = \Omega_s, \omega_8 = \omega_s\}$ [?].

The resultant dynamics can be analyzed via multimode Floquet theory [ESM05, WLS22], where the process can be understood as driving virtual motional transitions in a multimode Floquet space. The resulting effective Hamiltonian can be represented as an eight-mode Floquet Hamiltonian [ESM05, WLS22]:

$$\hat{H} = \sum_{K=1}^{\infty} \sum_{i=1}^8 \left(\hat{H}_1^{(i)} + \hat{H}_2^{(i)} + \cdots + \hat{H}_K^{(i)} \cdots \right) \times e^{-i(\sum_{k=1}^8 c_k \omega_k)t} e^{-i(c_6 + c_7 + c_8)\varphi_s} \quad (8.2)$$

While lower-order terms are readily obtained using methods from Refs. [ESM05, WLS22], deriving general analytical expressions for the K th-order term $\hat{H}_K^{(i)}$ is non-trivial. Nonetheless, the essential behaviors are evident from Eqn. 8.2. A resonant displacement results when $\sum_k c_k \omega_k = 0$. Since $c_k \in \mathbb{Z}$, this results in the following restrictions: for the probe tones, $\sum_{k=1}^5 c_k = \pm K/2$, and for the signal tone, $\sum_{k=6}^8 c_k = \mp K/2$.

This result has several consequences. First, δ becomes amplified by a factor

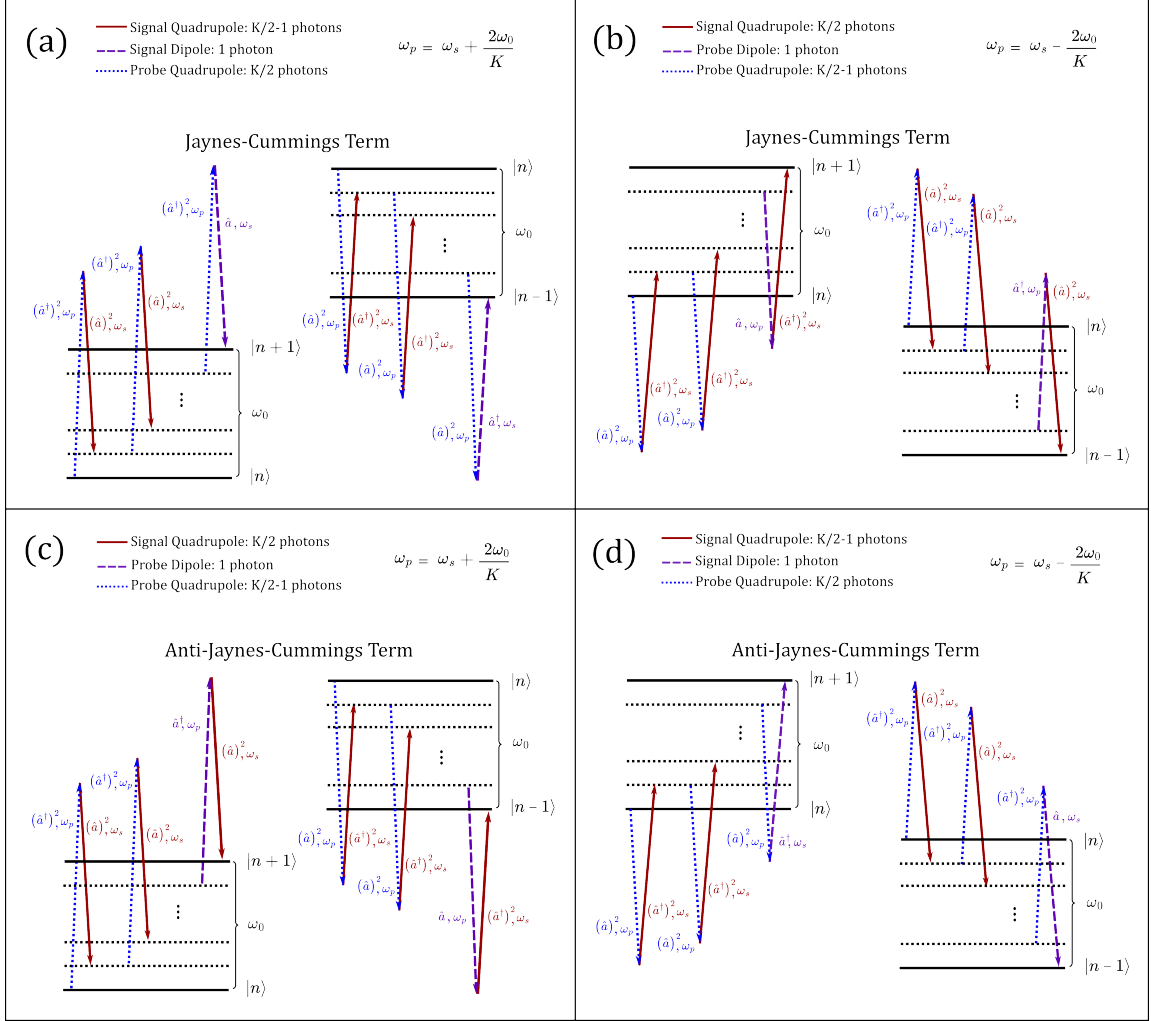


Figure 8.1: Displacement operations are generated by Jaynes-Cummings and Anti-Jaynes-Cummings terms in an effective Hamiltonian. For motional Raman interactions composed of K photons, where K is an even number, there are several possible configurations that can generate a displacement. (a) and (b) show the generation of a Jaynes-Cummings interaction and (c) and (d) show the creation of an Anti-Jaynes-Cummings interaction. In (a) the signal tone is applied on the electrode with both a dipole and a quadrupole component, whereas the probe tone is purely quadrupolar at $\omega_p = \omega_s + \frac{2\omega_0}{K}$. In (b) the signal tone is applied with only a quadrupole component, and the probe tone as both a dipole and quadrupole at $\omega_p = \omega_s + \frac{2\omega_0}{K}$. In (c) the signal tone is applied with only a quadrupole component, and the probe tone as both a dipole and quadrupole at $\omega_p = \omega_s - \frac{2\omega_0}{K}$. In (d) the signal tone is applied as both a dipole and a quadrupole, and the probe tone is a quadrupole at $\omega_p = \omega_s - \frac{2\omega_0}{K}$.

$\sum_{k=1}^5 c_k = K/2$, narrowing the linewidth relative to a linear measurement by $2/K$. Second, the K th-order Hamiltonian $H_K^{(i)}$ involves $K-1$ commutators [ESM05, WLS22]. Since all Fourier components $H_K^{(i)}$ are comprised of only first-order ($\hat{a}$, $\hat{a}^\dagger$) or second-order ($\hat{a}^2$, $\hat{a}^{\dagger 2}$, $\hat{a}^\dagger \hat{a}$) operators, the K th-order Hamiltonian is nonvanishing only when the number of first-order operators is exactly one, which occurs when the dipole tone contributes only one term. These determine the number of tones needed at each frequency, such that the displacement α is

$$\alpha = \beta(\omega_s, K, \omega_0) \Omega_d \Omega_q^{(K-2)/2} \Omega_s^{K/2} e^{-i\varphi_s K/2} \text{sinc}(K\delta t/4), \quad (8.3)$$

where β is a function of the signal frequency ω_s , the QHO frequency ω_0 , and subharmonic order K , independent of δ . The multimode Floquet process can be illustrated as a multiphoton transition, shown in Fig. 8.6(a). The dash-dotted line represents the subharmonic excitation for a quadrupole-only configuration (ii), while the dashed lines indicate excitation for a quadrupole/dipole configuration (iii).

8.2 Simulations

Simulations were conducted in QuTiP by time-evolving the initial state under the influence of the subharmonic excitation Hamiltonian in Eqn. 9.1 via the time-dependent Schrödinger equation. Constituent data points in Figs. 8.2(a-b) were obtained by simulating the system at different detunings δ to extract a lineshape, which was fitted using a sinc-squared lineshape to extract the maximum mean phonon number, linewidth, and center frequency. This procedure was repeated at different values of the target variable to extract the dependence on the target variable. The subharmonic excitation Hamiltonian was pulse-shaped in simulation using a tapered-cosine window with $100\mu\text{s}$ rise and fall times and a total length of 1.2ms . The basis size was varied for each data point to reduce simulation overheads and was confirmed to be sufficiently large to contain all simulation dynamics. The Fock state populations and Wigner functions were extracted for each simulation to confirm that a coherent state was produced.

The mean phonon number scales as V_d^2 for all orders of subharmonic excitation, where V_d is the dipole voltage, indicating an overall linear dependence of the displace-

ment on V_d , in agreement with the theoretical analysis and the empirical formulae in Eqn. 8.3. Simulations that instead vary the quadrupole voltages V_s, V_q show that the resultant phonon number scales as $(V_s V_q)^{2(K-1)}$, indicating that the displacement scales as $K - 1$, again in agreement with the theoretical analysis and the empirical formula 8.3.

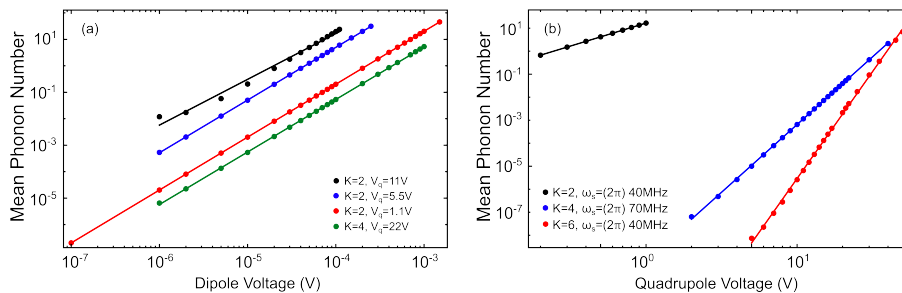


Figure 8.2: (a, b) Dependence of the subharmonic excitation on the applied dipole voltage V_d and the applied quadrupole voltages V_s, V_q , respectively. Results are simulated using a $\omega = 1$ MHz secular frequency. A linear model $y = mx + b$ is fitted to the log-log transform of the results to extract the power-law dependence at each subharmonic order. (a) Simulated results at all subharmonic orders exhibit an approximately quadratic dependence of mean phonon number on dipole voltage V_d , equivalent to a linear dependence of displacement on V_d . $\omega_s/2\pi = 40$ MHz. (b) Simulated results exhibit a power-law scaling of the mean phonon number with V_q , using $V_d = 1$ mV, that agrees well with the predicted scaling of $2(K - 1)$.

Simulations in Fig. 8.3 examine the dependence of the mean phonon number $\langle n \rangle$ on the signal frequency ω_s . These results demonstrate that $\langle n \rangle \propto 1/\omega_s^{2K}$, indicating that the displacement scales as $\alpha \propto 1/\omega_s^K$.

8.3 Standard Quantum Limit

Here we derive a general expression for the frequency uncertainty $(\Delta\delta)$ for the K -th order subharmonic. Since only the δ -dependent term is relevant for deriving the uncertainty $(\Delta\delta)$, Eqn. 8.3 in the main text can be simplified as:

$$\alpha = |\tilde{a}| \frac{\sin(K\delta\tau/4)}{(K\delta\tau/4)}, \quad (8.4)$$

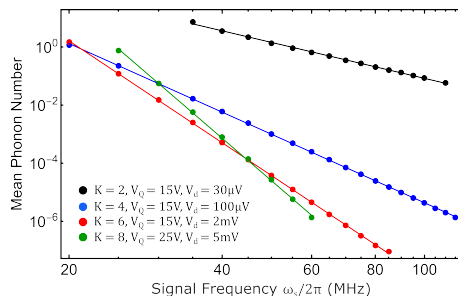


Figure 8.3: Dependence of the subharmonic excitation on the signal tone frequency ω_s . Results are simulated using $\omega = 1$ MHz secular frequency. A linear model $y = mx + b$ is fitted to the log-log transform of the results to extract the power-law dependence at each subharmonic order. Simulated results exhibit a power-law scaling of the mean phonon number with ω_s in good agreement with the predicted scaling of $\langle n \rangle \propto \omega_s^{2K}$, or equivalently, $\alpha \propto 1/\omega_s^K$.

where $\tilde{a}$ represents the product of all prefactors multiplying the *Sinc* function in Eqn. 8.3 and corresponds to the maximum displacement in phase space, τ is the interrogation time, and N is the number of experiments. $\tilde{a}$ is typically fixed at approximately 0.6 across different experiments and values of K .

The frequency uncertainty $\Delta\delta$ can be obtained through standard error propagation:

$$\Delta\delta = \frac{\Delta\alpha}{\frac{\partial\alpha}{\partial\delta}} = \frac{4\Delta\alpha}{K\tau} \frac{(K\delta\tau/4)^2}{(K\delta\tau/4 \cos(K\delta\tau/4) - \sin(K\delta\tau/4)) |\tilde{a}|\sqrt{N}}, \quad (8.5)$$

where $\Delta\alpha$ denotes the uncertainty in α for a single measurement. Its value depends on the Fisher information associated with the measurement technique and is ultimately bounded by 0.5 for a classical state. The actual frequency uncertainty is determined from an Allan deviation analysis, denoted σ_{ω_s} . In this approach, two operating points close to the half-maximum of the signal are used, which optimally balance sensitivity and signal-to-noise ratio. Under these conditions, $\text{sinc}(\frac{K}{4}\delta\tau) = 1/\sqrt{2}$. Eqn. 8.5 thus becomes:

$$\Delta\delta \approx 2\pi \frac{4\Delta\alpha}{K} \frac{0.418}{|\tilde{a}|\tau\sqrt{N}}, \quad (8.6)$$

Increasing K thus leads to a smaller uncertainty of δ .

To benchmark our method, we calculate the best sensitivity achievable with the corresponding linear generator using classical states, or equivalently, the SQL associ-

ated with that linear generator. This SQL corresponds to the special case of $K = 2$, which can be obtained directly from Eqn. 8.6. The expression is shown below:

$$\Delta\delta_{SQL}^{K=2} \approx 2\pi \frac{0.418}{|\tilde{a}|\tau\sqrt{N}}. \quad (8.7)$$

where $|\tilde{a}|$ is the displacement, τ is the interrogation time, and N is the number of experiments.

A common method for determining the mean phonon number $\langle n \rangle$ of different quantum states involves measuring the excited state population (P_r) after a pulse applied for $t = \pi/\Omega_{01}$ on the red motional sideband of the $|^2S_{\frac{1}{2}}, m_J = -\frac{1}{2}\rangle \Rightarrow |^2D_{\frac{5}{2}}, m_J = -\frac{5}{2}\rangle$ qubit transition, where Ω_{01} is the Rabi frequency on the blue motional sideband on the same transition between Fock states $n = 0$ and $n = 1$.

In Fig. 8.4 (a), we plot the mapping of phonon number onto the excited state population for both a coherent state and a squeezed state using our protocol. As can be seen, due to the form of a squeezed state in the Fock basis, the slope of the mapping is much less steep for a squeezed state than for a coherent state. Intuitively, this means that squeezed states will provide worse performance during the readout step. More quantitatively, in the framework of quantum metrology, better sensitivity corresponds to a larger Fisher information. For the same mean phonon number, the Fisher information for readout of a squeezed state (F_s) is lower than for that of a coherent state (F_c) for our readout scheme. Specifically, the Fisher information of our readout is $F(n) = \sum \frac{1}{P_r} \left(\frac{\partial P_r}{\partial n} \right)^2 = \frac{1}{P_r(1-P_r)} \left(\frac{\partial P_r}{\partial n} \right)^2$. From the relation between excited state population (P_r) and phonon number shown in Fig. 8.4(a), the ratio of the Fisher information between a coherent and a squeezed state is shown in Fig. 8.4(b); the improved sensitivity of the coherent state is apparent as F_c/F_s exceeds unity for all $\langle n \rangle$, demonstrating that coherent states yield higher Fisher information and therefore improved detection sensitivity, making them advantageous for precision measurement.

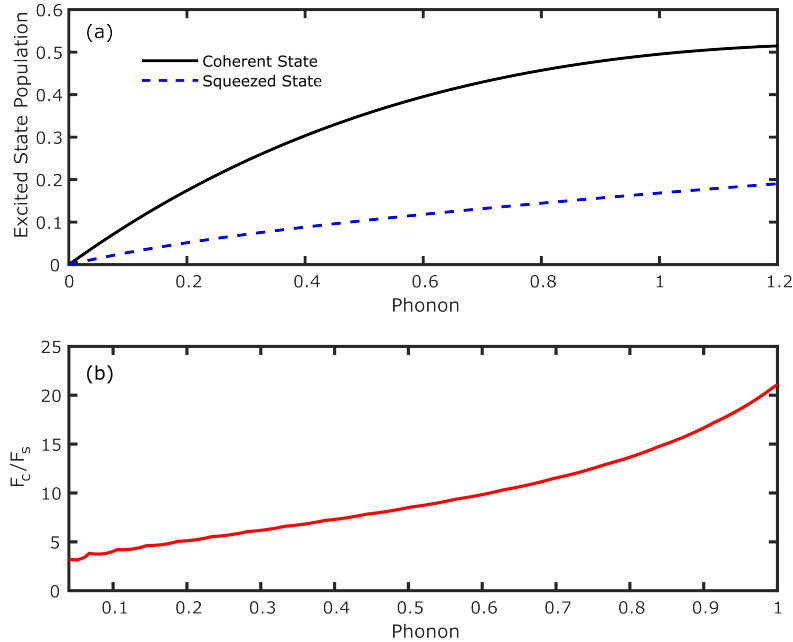


Figure 8.4: Characterizing the sensitivity of phonon readout for coherent and squeezed states. (a) The relation between population probability in the excited state $|^2D_{\frac{5}{2}}\rangle$ and phonon number for different motional states. The population is measured after applying a red sideband Rabi oscillation for interrogation time $\tau = \pi/\Omega_{01}$, where Ω_{01} is the Rabi frequency for the $|^2S_{\frac{1}{2}}, m_J = -\frac{1}{2}, n = 0\rangle$ to $|^2D_{\frac{5}{2}}, m_J = -\frac{5}{2}, n = 1\rangle$ transition. (b) The Fisher information for a coherent state (F_c) is greater than the Fisher information of a squeezed state (F_s) for any mean phonon number, making coherent states a more desirable quantum state for precision measurements.

8.4 Results

8.4.1 Experimental Procedure

To verify this result and demonstrate the technique, we realize a QHO using a single trapped $^{40}\text{Ca}^+$ ion with a radial (axial) secular frequency $\omega_r \approx 2\pi \cdot 1$ MHz ($\omega_z = 2\pi \cdot 0.7$ MHz). Doppler cooling of the ion followed by resolved-sideband cooling on the optical qubit transition prepares both the radial (axial) motional mode near the ground state with $\langle n \rangle \approx 0.01(1)$. The dephasing time τ_c of the radial (axial) mode without applying external fields is ≈ 2 ms (≈ 20 ms).

Subharmonic excitation can be driven on any QHO mode by applying to the

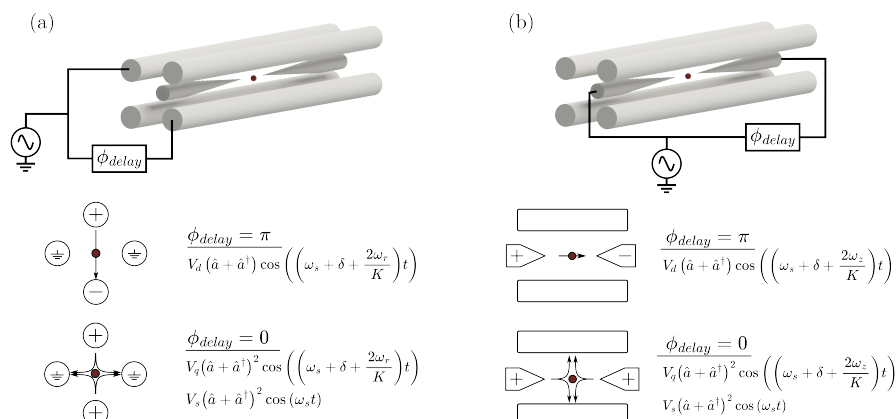


Figure 8.5: Experimental setup for generating radial and axial subharmonic results.

trap electrodes both the signal tone at frequency ω_s in a quadrupole configuration and the probe tone at a frequency $\omega_p = \omega_s + 2\omega_{r/z}/K + \delta$ with both dipole and quadrupole components. In what follows, we assume that all AC stark shifts of the QHO frequency from the quadrupole tones are absorbed into the resonance condition. Pulses are applied with a tapered-cosine envelope to mitigate off-resonant excitation [ZHM19].

8.4.2 Ramsey Spectroscopy

First, we confirm that the dynamics of subharmonic excitation agree with our model in Eqn. 8.3. We employ a motional Ramsey sequence [WSH19], depicted in Fig. 8.6(b), to demonstrate that subharmonic excitation is a K -th order process. Following cooling near the motional ground state $|0\rangle$, a resonant subharmonic excitation pulse is applied for interrogation time τ to generate a displacement $\hat{D}(\alpha)$. After free evolution for T_{delay} , a second resonant subharmonic excitation is applied with relative phase φ_s to generate a secondary displacement $\hat{D}(\alpha')$. For a K th-order process, this leads to a phase shift $K\varphi_s/2$ of the displacement, confirmed in Fig. 8.6(d) with varying order. Further, Rabi oscillations on the blue motional sideband confirm that subharmonic excitation induces a displacement (insets in Fig. 8.7 (a, b)).

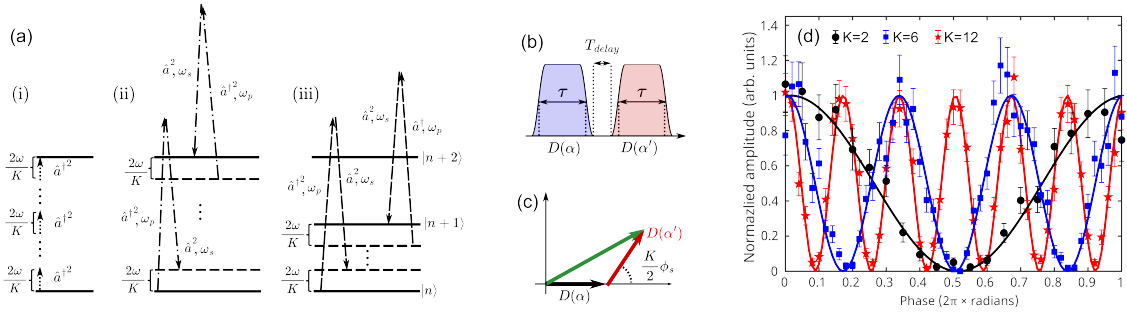


Figure 8.6: Energy-level diagrams for motional subharmonic excitation and verification via motional Ramsey spectroscopy. (a) Level diagrams for a K th-order parametric excitation (i), quadrupole subharmonic Raman excitation (ii) and quadrupole/dipole subharmonic Raman excitation (iii). (b-d) Experimental verification of accelerated dynamics via a motional Ramsey sequence with a higher-order subharmonic excitation. (b) Simplified pulse sequence of the motional Ramsey scheme used to examine the phase response. A subharmonic excitation pulse is applied for interrogation time τ to generate a displacement $\hat{D}(\alpha)$. Following free evolution for time T_{delay} , a second subharmonic excitation pulse of equal area is applied with phase φ_s relative to the initial pulse to generate a second displacement $\hat{D}(\alpha')$, mapping the phase onto the ion's motional state. (c) Motional Ramsey protocol in phase space. The phase φ_s rotates the second displacement $\hat{D}(\alpha')$ relative to the first. The phase dependence at order K thus corresponds to the periodicity of the Ramsey fringes. (d) Ramsey fringes generated by scanning the phase φ_s of the second subharmonic excitation pulse at different orders K . Here, $\tau = 1$ ms and the delay time is $T_{\text{delay}} = 50 \mu\text{s}$. Solid lines are fits of the data to a sine. Excitation amplitudes for all subharmonic orders K are normalized for comparison. Error bars represent one standard error.

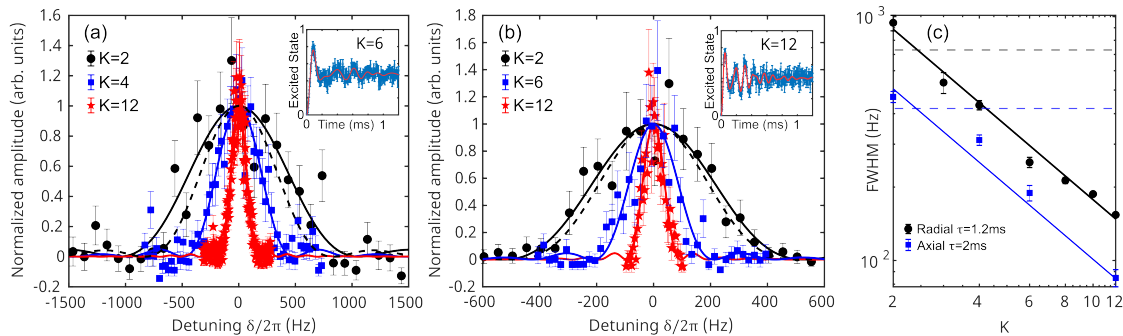


Figure 8.7: Linewidth narrowing beyond the linear Fourier transform limit (FTL) using subharmonic excitation. (a, b) Detection of an “unknown” signal with subharmonic excitation at $\omega_s/2\pi = 70$ MHz on the radial mode (a) and $\omega_s/2\pi = 80$ MHz on the axial mode (b). The “unknown” frequency is determined by scanning the probe detuning to maximize the signal amplitude. For visibility, fit amplitudes are normalized to unity, with corresponding data rescaled by the same factor. Solid lines are fits of the data to a sinc-squared lineshape. Insets in (a, b): Blue sideband Rabi oscillations characterize the motional state after subharmonic excitation. Solid red lines are fits assuming a motional coherent state. (c) Dependence of linewidth on subharmonic order K . The linewidth is defined as the Full-Width at Half Maximum (FWHM). The solid line is a linear fit to the data with no offset. (a, black data in c) An effective interrogation time of $\tau = 1.2$ ms used at different values of K . (b, blue data in c) $\tau = 2.0$ ms at different values of K . τ is the pulse width of a square pulse of equivalent amplitude and area. (a, b, c) Dashed lines show the FTL of the applied tapered-cosine pulse envelope. Error bars represent one standard error.

8.4.3 Linewidth Narrowing Beyond Linear FTL

We examine the line-narrowing of our protocol, relative to the linear case, by conducting spectroscopy at different subharmonic order K . Specifically, we measure the frequency of a signal tone by sweeping the probe detuning δ across the subharmonic resonance (Fig. 8.7 (a,b)), then measure the resultant mean phonon number $\langle n \rangle$ using the sideband ratio technique [WMH25]. Linewidths were extracted by fitting a sinc-squared profile $|\beta(\omega_p)|^2$ to the data (Fig. 8.7 (c)). At $K \geq 4$, the linewidth of the subharmonic surpasses the linear case (dashed lines in Fig. 8.7 (c)), with improvement by approximately 6 times over the linear case at nonlinear $K = 12$. To demonstrate arbitrary frequency operation, we excite the $K = 6$ subharmonic at $\omega_s/2\pi = 70, 80$ and 200 MHz.

From Fig. 8.7 (c), the linewidths narrow as $2/K$. The $2/K$ improvement is the maximum achievable and requires a square pulse envelope; non-square pulse envelopes, e.g. Blackman or Gaussian envelopes, result in reduced narrowing due to the nonlinear dependence on amplitude. We use a tapered-cosine pulse (Fig. 8.6 (b)) to retain the narrowing effect while suppressing off-resonant excitation.

We next examine the *useful* metrological gain relative to the best sensitivity achievable with the corresponding linear generator using classical states, or equivalently, the SQL of $K=2$. The subharmonic lineshape is first measured, as in Fig. 8.7 (a,b). Subsequent frequency measurements are made at the two half-maxima of the lineshape, then fitted to determine the center frequency. These are used to compute the overlapping Allan deviation (σ_{ω_s}), shown in Figs. 8.8 (a,b) for an 80 MHz signal at various K . For comparison, the solid line represents the corresponding SQL for $K=2$ ($\Delta\delta_{SQL}^{K=2}$) computed from the pulse area. A minimum frequency resolution of 0.85(18) Hz was achieved at $N \approx 16000$ with $K = 12$ on the axial mode. The maximum metrological gain [PSO18] of $20\log_{10}(\Delta\delta_{SQL}^{K=2}/\sigma_{\omega_s}^{min})=12.3(9)$ dB over the SQL of $K=2$ was realized at $N \approx 1600$ with interrogation time $\tau = 2$ ms. Likewise, a gain of 4.9(8) dB was obtained on the radial mode with $K = 24$ and $\tau = 2$ ms, with a minimum resolution of 2.3(2) Hz at $N \approx 7200$. The reduction in measurement performance on the radial mode results from increased off-resonant excitation and greater secular frequency instability. Higher power requirements at higher orders

result in a practical limit of $K = 24$ ($K = 12$) on the radial (axial) mode; likewise, an increase in sensing frequency results in increased power requirements. This limit is not fundamental. These effects can be mitigated by optimizing the RF coupling efficiency to reduce the required input powers.

The achieved 12.3(9) dB gain is comparable to that obtained with a linear measurement on a nonclassical $|n \approx 20\rangle$ Fock state [DLC24]. However, there is a crucial difference. Non-classical states enhance sensitivity in parameter estimation by increasing separation between superposed states of a system, which results in greater phase accumulation for improved Fisher information [DC17]. However, non-classical states decohere faster - for example, the coherence time of a Fock state $|n\rangle$ worsens as $1/n^p$, where $p \geq 1$ [TMK00, MKT00]. This reduces *usable* measurement times to largely eliminate any metrological gain over the SQL in the context of ultimate precision. Unlike commonly used active approaches that can mitigate the reduced coherence time associated with nonclassical states—such as decoherence-free subspaces developed for spin systems [BCM19] or bosonic quantum error-correction codes such as binomial codes [MSB16], our protocol requires only classical input states and derives metrological gain from a *nonlinear* interaction. As a result, it passively avoids the excess decoherence typically introduced by nonclassical probe states, thereby improving the *ultimate* precision of frequency sensing for bosonic systems such as motional modes.

8.4.4 Wideband performance

To confirm the wide frequency operation of our protocol, we characterized the $K=6$ subharmonic resonance at three different signal frequencies $\omega_s/2\pi = 70, 80$ and 200 MHz (Fig. 8.9). Traces are normalized to maximum excitation and exhibit highly overlapping lineshapes, making our protocol uniquely suited to the challenges of wideband quantum sensing. Sensing with the $K = 6$ subharmonic at $\omega_s = 200$ MHz required an RF power of roughly 33 dBm. Hardware upgrades, such as an improved coupling of the signal onto the trap electrodes, should enable sensing up to ≈ 400 MHz.

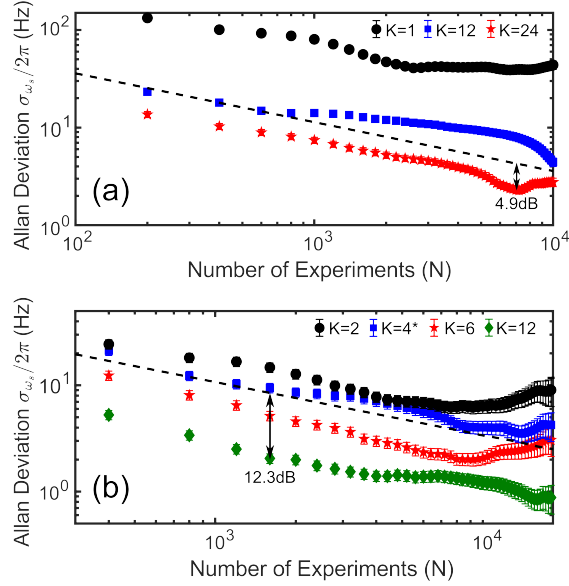


Figure 8.8: Frequency resolution of the subharmonic protocol. (a-b) Overlapping frequency Allan deviations for an 80 MHz tone at different subharmonic orders K on the radial (a) and axial (b) modes. Each experiment takes ~ 17 ms. Solid lines represent the corresponding SQL for the given interrogation time, which correspond with the SQL of the $K = 2$ subharmonic. (a) Subharmonic excitation is probed on the radial mode with interrogation time $\tau = 2$ ms. A minimum frequency resolution $\sigma_{\omega_s}^{min}/2\pi = 2.3(2)$ Hz is achieved with $K = 24$ order at $N \approx 7200$. A maximum metrological gain of 4.9(8) dB is similarly achieved with $K = 24$ at $N \approx 7000$. (b) Subharmonic excitation is probed on the axial mode with $\tau = 2$ ms. A minimum frequency resolution $\sigma_{\omega_s}^{min}/2\pi = 0.85(18)$ Hz is achieved with $K = 12$ at $N \approx 16000$. A maximum metrological gain of 12.3(9) dB is achieved with the same $K = 12$ at $N \approx 1600$. Error bars represent one standard error.

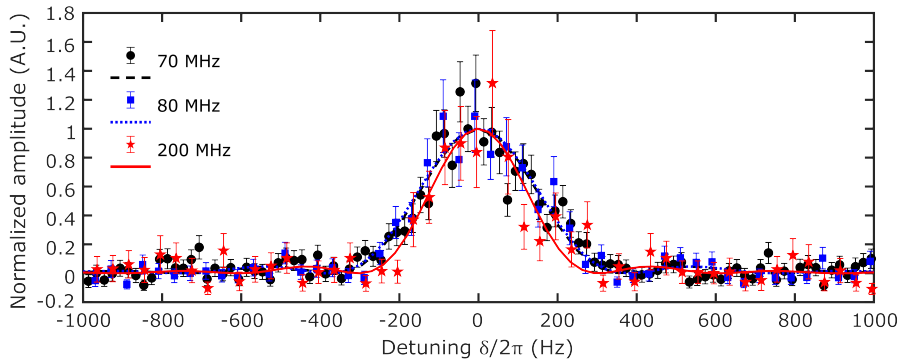


Figure 8.9: $K=6$ subharmonic excitation at different signal frequencies $(\omega_s/2\pi)=(70, 80 \text{ and } 200 \text{ MHz})$. Lines are fits to the data using a sinc-squared profile. All data are normalized by the peak amplitudes of their fit. The interrogation time is fixed at $\tau = 1.2 \text{ ms}$. Error bars represent one standard error.

8.4.5 Minimum Frequency Uncertainty

Finally, a minimum frequency uncertainty $\sigma_{\omega_s}^{min}/2\pi = 0.56(32) \text{ Hz}$ was achieved at 80 MHz with $\tau = 5 \text{ ms}$ on the axial mode. To our knowledge, this surpasses the precision of previous frequency measurements in the radio-frequency band using QHO-based system and sets a new state-of-the-art for these measurements. This measurement could easily be improved by approximately 4 times by improving damage thresholds of system electronics, with further gains achievable by extending motional coherence times.

To achieve a state-of-art frequency resolution for an 80 MHz signal tone, we excited the $K = 12$ subharmonic resonance on the axial mode with an interrogation time up to $\tau = 5 \text{ ms}$, as illustrated in the inset of Fig. 8.10. The Allan deviation was determined by measuring two points at the FWHM of the motional spectrum. A minimum frequency resolution of $\sigma_{\omega_s}/2\pi = 0.56(32) \text{ Hz}$ was achieved after $N = 9600$ experiments. The motional coherence time for the axial mode is 20 ms. The primary limitation on the achievable minimum frequency resolution is the damage threshold of trap electronics (5 Watts). Measurements at other values of K at $\tau = 5 \text{ ms}$ were not pursued to avoid risking damage to the electronic hardware. Upgrades to system electronics would allow excitation of subharmonic orders beyond $K = 12$, improving the minimum resolution by a factor of at least $4\times$.

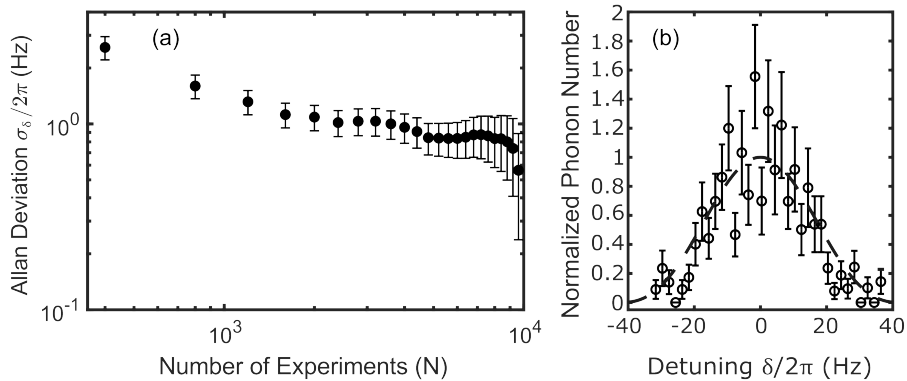


Figure 8.10: High resolution scan of an 80 MHz probe tone using the $K = 12$ subharmonic resonance on the axial mode. (a) An overlapping Allan deviation yields a minimum frequency uncertainty $\sigma_{\omega_s}/2\pi = 0.56(32)$ Hz at $N = 9600$. The interrogation time is fixed at $\tau = 5$ ms. Each individual experiment takes 22 ms and is comprised of cooling pulses, the subharmonic excitation sequence, and state detection. (b) Motional excitation spectrum generated by scanning the frequency of the probe tone. Error bars represent one standard error.

8.5 Further Discussion

8.5.1 Comparison to Quantum Enhanced States

Though nonclassical states are used to improve sensitivities [BSB19, WSH19, MKB19, GBS17, HLJ13], they incur a penalty of excess decoherence due to their sub-Planck structure [Zur01, TMK00]. This is well-noted with GHZ states [DRC17], which can be used to enhance a signal by a factor of M , where M is the number of probes in the GHZ state. However, in the presence of pure dephasing, GHZ states decohere M times faster, which negates their M -fold gain to yield *no* net metrological gain. Similarly, when sensing on bosonic modes (such as those of a trapped ion), Fock superposition states $|\psi_n\rangle = \frac{|0\rangle + |n\rangle}{\sqrt{2}}$ offer Heisenberg-limited n -fold gain, but decohere in excess by a factor n^2 for a net *reduction* in metrological potential [MKB19, TMK00, MKT00].

In this context, our nonlinear protocol can be used to achieve *ultimate* metrological gain as it yields amplification while still creating a coherent state - as coherent states are classical and lack the sub-Planck structure of nonclassical states, they incur

no excess decoherence, which, when combined with our nonlinear protocol, allows for net, *penalty-free* metrological gain.

To concretely compare the metrological potential of our nonlinear protocol in the presence of decoherence, we simulate the following motional Ramsey protocol for frequency sensing: the state is first displaced, then subjected to free evolution for a delay time T_D , then antidisplaced [WSH19, MKB19, MKW19]. The state is coupled to a phase reservoir over the entirety of the protocol to induce dephasing. This coupling is selected to give $T_2 = 2$ ms. Amplification strategies using nonclassical states typically apply this motional Ramsey to the state, then apply time-reversal dynamics (i.e. undo the nonclassical state) to remove any added noise [BSB19, BSK21]. For example, quantum-enhanced frequency sensing with Fock states uses a protocol of state creation ($|0\rangle \rightarrow |n\rangle$), displacement, free evolution, antidisplacement, and state reversal ($|n\rangle \rightarrow |0\rangle$) [WSH19]. For simplicity, we assume the nonclassical state creation and reversal are instantaneous. By contrast, our nonlinear subharmonic protocol involves no nonclassical state dynamics; instead, the displacement (and antidisplacement) is simply generated using a higher-order subharmonic resonance - here, we use the $K = 10$ subharmonic. As a reference, we show the bare $K = 2$ linear protocol, which represents the standard quantum limit. For fair comparison between nonclassical protocols and our nonlinear protocol, the displacement of the motional Ramsey is kept constant at $\alpha = 0.1$, while the amplification of the nonclassical states (characterized by the mean phonon number $\bar{n}$ of the amplified state) is kept constant across the nonclassical states, i.e. $\bar{n} = 5$.

To determine the ultimate achievable metrological gain of each protocol, we simulate a frequency linescan at each delay time T_D . The maximum frequency Fisher information at each delay time is then calculated from the linescan using $F(\delta) = \frac{(\partial_\delta P)^2}{\text{var}(P)}$ [WSH19], from which the sensitivity σ_δ can be extracted (via $\sigma_\delta = \sqrt{\frac{1}{F(\delta)/T}}$).

As shown in Fig. 8.11, although Fock states achieve the best sensitivity when using a nonclassical strategy, this improvement is surpassed by our nonlinear protocol using the $K = 5$ subharmonic. Again, this is due to the fact that our nonlinear protocol results in the creation of a classical coherent state, which suffers no excess decoherence. In this sense, such a nonlinear protocol can be said to be *penalty-free*.

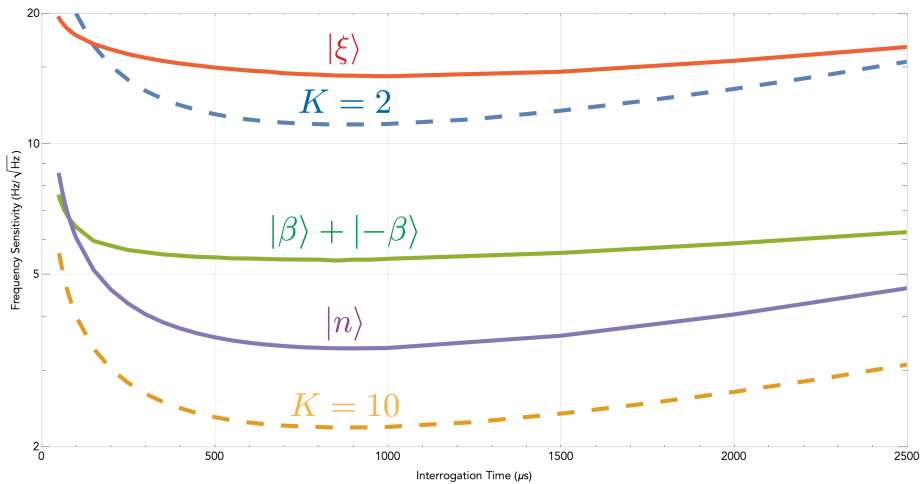


Figure 8.11: Simulated frequency sensitivity using a motional Ramsey protocol (displace - delay - antidisplace) with different amplification strategies in the presence of dephasing ($T_2 = 2$ ms). The amplification-free case is represented by the $K = 2$ linear protocol, which uses $|\alpha = 0.1\rangle$. Quantum amplification with nonclassical states is implemented by applying this $K = 2$, $|\alpha = 0.1\rangle$ linear protocol to the nonclassical state, then time-reversing the nonclassical dynamics. We simulate quantum amplification of the motional Ramsey using a Fock state ($|n\rangle$, $n = 5$), a squeezed state ($|\xi\rangle$, $\xi = 1.544$), and a cat state ($|\beta\rangle + |-\beta\rangle$, $\beta = 2.236$). The parameters of the nonclassical states have been chosen to keep the mean phonon number $\bar{n} = 5$. Amplification with our nonlinear protocol is implemented by instead generating the displacement of the motional Ramsey using a higher-order subharmonic, shown here as the $K = 10$ subharmonic. For fair comparison, the displacement is kept the same as that with the nonclassical protocols, i.e. $\alpha = 0.1$.

8.5.2 Odd Subharmonics

So far, the discussion has been focused on the even subharmonics. An investigation of the level diagram given in Fig. 8.1 shown makes it apparent that if an odd number of tones are present, then they are unable to properly produce the Jaynes-Cummings terms with $\omega_p = \omega_s \pm \frac{2\omega_0}{K}$. However, by using a different probe tone frequency it is possible to generate the Jaynes-Cummings terms (and AJC terms) using an odd number of phonons. Fig. 8.12 demonstrates how one of these terms can be constructed. The signal is applied in the quadrupole configuration, and the probe is applied with both a dipole and a quadrupole component. When the probe frequency

is $\omega_p = \frac{K-1}{K+1}\omega_s + \frac{2\omega_0}{K+1}$, the sum of the photon frequencies is the splitting in the energy between two adjacent levels in the QHO. By changing whether the dipole is on the carrier or the probe tone, or whether the desired Hamiltonian follows the JC or AJC model, will result in acceptable frequencies given by

$$\omega_p = \frac{K \mp 1}{K \pm 1}\omega_s \pm \frac{2\omega_0}{K \pm 1}. \quad (8.8)$$

A quick simulation confirmed this behavior but was not investigated further, since we did not foresee additional benefits to implementing these odd harmonics. Their nonlinear behavior will produce similar results to the even harmonics, while probing at a frequency much further away from the signal. In principle, since they can be further away from the signal, this may offer some practical application if there is large insertion loss near the signal frequency, however, as K increases, the probe frequency will approach the signal, so this application only works for small K .

8.5.3 Superharmonics

Superharmonic excitations were the first indication of higher-order harmonics appearing within our experiment. We would see mysterious heating as we tried to drive the EGGs transitions in our molecule, which persisted after the molecule was removed from the trap. They appeared at what seemed to be strange intervals, but a pattern was clear from the numerology: we were seeing steps related to fractions of our trapping RF and our secular frequencies. Applying attenuators at those frequencies failed to remove the heating, and this method allowed us to track down a tone leaking from the ARTIQ Phaser, which was outside the attenuation band and was beating with the trapping RF, as shown in Fig. 8.13. The signal that was leaking from the phaser was $\omega_x = 69.4445\text{MHz}$, and we detected peaks at $\omega_x \pm \frac{2}{3}\Omega_{rf} \pm \frac{1}{3}\omega_0$ and at $\omega_x \pm \frac{1}{3}\omega_0$. Superharmonic tones are also present at $N \Omega_{rf} \pm \omega_0$, where N is an integer. The highest superharmonic term we measured was $N = 11$.

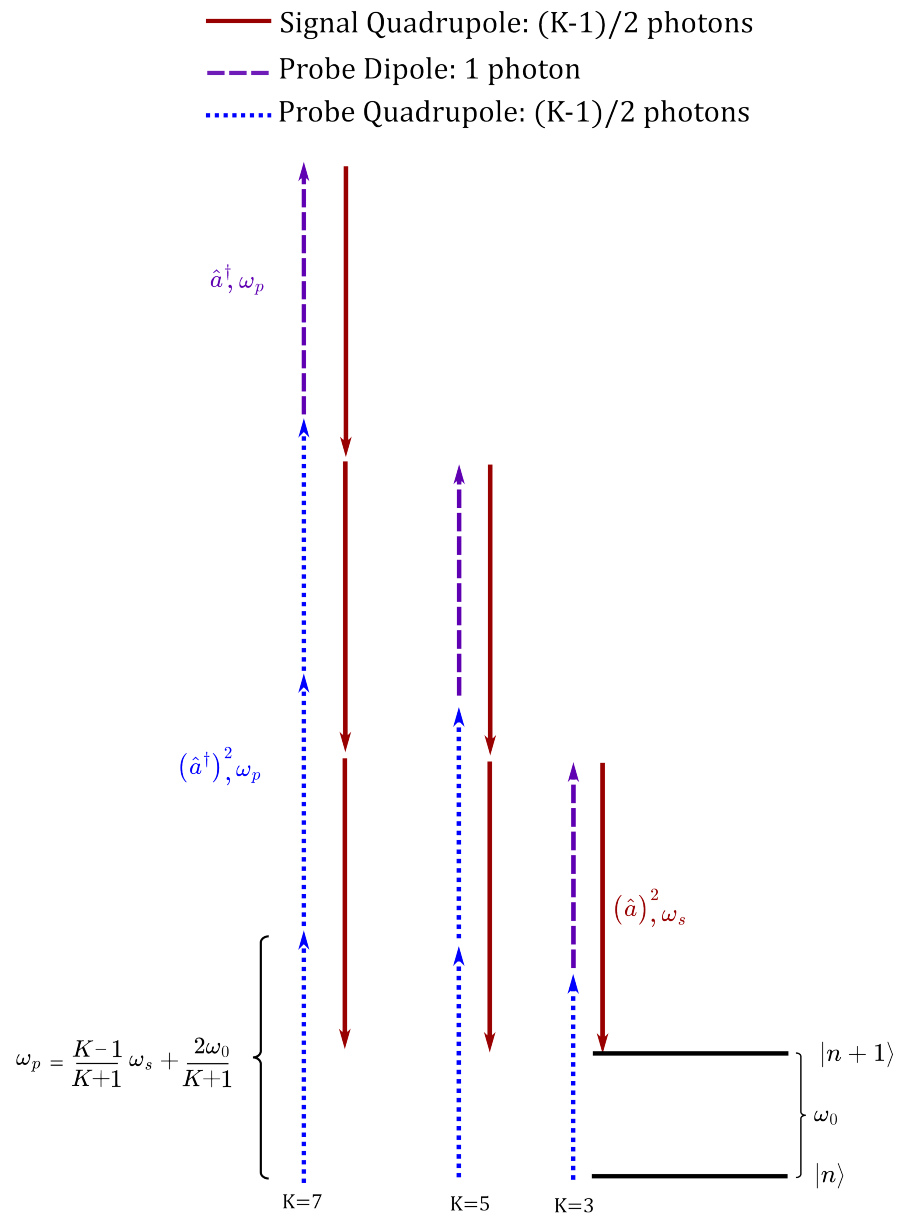


Figure 8.12: Odd K-harmonics for K=3,5, and 7. In this picture the signal is a pure quadrupole, whereas the probe tone has both dipole and quadrupole components, producing a Jaynes-Cummings term.

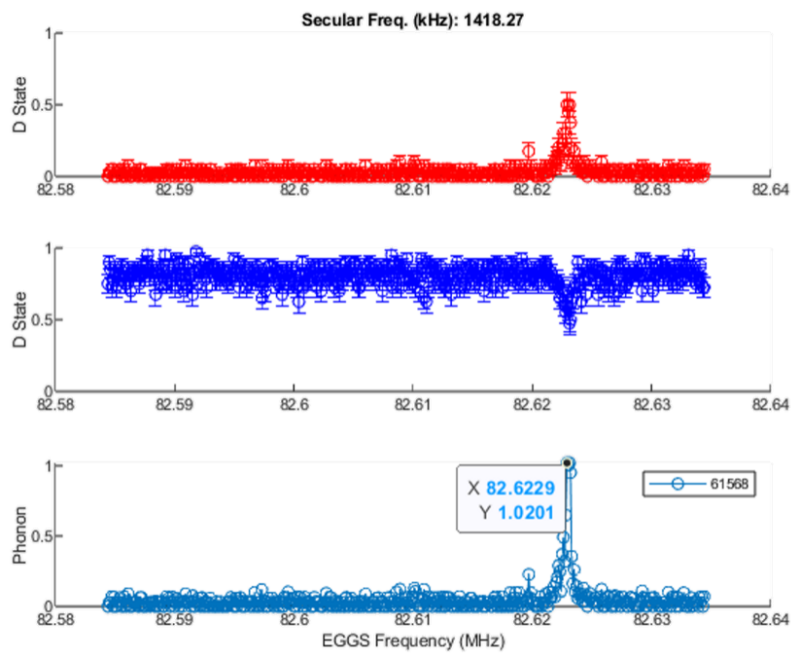


Figure 8.13: What led us to investigate the subharmonic effects was heating due to leakage from the ARTIQ phaser. Shown here is a peak that corresponds to $\omega_x + \frac{2}{3}\Omega_{rf} + \frac{1}{3}\omega_0$, where $\omega_x = 69.4445\text{MHz}$ is the leakage from the phaser.

CHAPTER 9

Super-resolution

The work shown in this chapter is in the process of being published and can be found in [Hao25].

9.1 Rayleigh's Curse

Fundamental metrological challenges arise when the statistical distance between two quantum states goes toward zero [Woo81], which largely originate when the quantum states from two signals, e.g. closely spaced frequencies or spatial features, undergo evolution and overlap substantially within the resolution limit (e.g. linewidth or diffraction limit), such as depicted in Fig. 9.1(a). As a result, their distinguishability is reduced and the underlying parameters cannot be reliably measured. For decades, this phenomenon, known as Rayleigh's curse [TNL16], was thought to set an ultimate limit in both frequency spectroscopy and imaging, as it was believed that the conventional resolution limit dictated the minimum overlap between quantum states.

Rayleigh's curse for signal detection can be introduced when two unknown oscillating, uniform electric fields are present at frequencies ω_1 and ω_2 . The resolution of these tones scales as the inverse of the probing time T , by a probe tone at ω_p in an electric quadrupole configuration. Heterodyne mixing from motional Raman generates a displacement to sense the unknown tones. The Hamiltonian of the ion motion under these fields is

$$\begin{aligned} \frac{\hat{H}}{\hbar} = & \omega_o \hat{a}^\dagger \hat{a} \\ & + \Omega_d (\hat{a} + \hat{a}^\dagger) (\cos(\omega_1 t + \varphi_1) + \cos(\omega_2 t + \varphi_2)) \\ & + \Omega_p (\hat{a} + \hat{a}^\dagger)^2 \cos(\omega_p t), \end{aligned} \quad (9.1)$$

where ω_o is the natural frequency of the QHO, $\varphi_{1/2}$ represents the phase of the

relative signal, $\Omega_d = \frac{eV_d\eta}{\hbar}$, $\Omega_p = \frac{eV_p\eta^2}{\hbar}$, V_d is the voltage of the unknown tones, V_p is the probe voltage, and η is the effective Lamb-Dicke parameter [WMH25]. For simplicity, the amplitudes of the two unknown tones are assumed to be equal.

Tuning the probe tone such that $\omega_p = \omega_\mu + \omega_0 + \delta$, where $\omega_\mu = \frac{\omega_1 + \omega_2}{2}$ for some fixed $\Delta\varphi = \varphi_2 - \varphi_1$, creates a coherent state $|\alpha\rangle$ following the conventional QVSA protocol based on the motional Raman effect. The resulting displacement, ignoring any intermodulation between the two dipole frequencies, can be expressed as:

$$\alpha = \sum_{i=1}^2 i\Omega_{\text{eff}} e^{-i\varphi_i} \frac{e^{i\delta_i t_0} - e^{i\delta_i(T+t_0)}}{\delta} = \sum_{i=1}^2 i\Omega_{\text{eff}} e^{i\delta_i t_0} e^{-i\varphi_i} \frac{1 - e^{i\delta_i T}}{\delta} \quad (9.2)$$

where $\Delta\omega = (\omega_2 - \omega_1)$ is the frequency difference between the unknown tones, $\Omega_{\text{eff}} = \frac{\Omega_d \Omega_p \omega_0}{(\omega_\mu \pm \Delta\omega/2)^2 - \omega_0^2}$ is the effective Rabi frequency, T is the interrogation time and t_0 denotes the start time of the sequence. The detuning in our displacement operator is defined as $\delta = \omega_q - \omega_d - \omega_0$, such that the detunings for each signal tone can be expressed as $\delta_1 = \omega_q - \omega_1 - \omega_0 = \delta - \Delta\omega/2$ and as $\delta_2 = \delta + \frac{\Delta\omega}{2} - \omega_0$. As a result, the displacement will be given by

$$\alpha(\Delta\omega, \Delta\varphi, T, t_0) = i\Omega_{\text{eff}} \left(e^{i(\delta - \Delta\omega/2)t_0} \frac{1 - e^{i(\delta - \Delta\omega/2)T}}{\delta - \Delta\omega/2} + e^{-i\Delta\varphi} e^{i(\delta + \Delta\omega/2)t_0} \frac{1 - e^{i(\delta + \Delta\omega/2)T}}{\delta + \Delta\omega/2} \right). \quad (9.3)$$

In a typical experiment we can treat $t_0 = 0$ since we apply our coherent operations immediately after preparing the QHO in its motional ground state. We have explicitly allowed for the two electric fields to be incoherent, i.e. $\varphi_1 \neq \varphi_2$, and we assume that these phases jump shot-to-shot but are roughly stable during the time T . Ignoring a global phase, 9.3 can then be reduced to

$$\alpha(\Delta\omega, \Delta\varphi, T, 0) = i\Omega_{\text{eff}} \left(\frac{1 - e^{i(\delta - \Delta\omega/2)T}}{\delta - \Delta\omega/2} + e^{-i\Delta\varphi} \frac{1 - e^{i(\delta + \Delta\omega/2)T}}{\delta + \Delta\omega/2} \right), \quad (9.4)$$

with $\Delta\varphi = \varphi_2 - \varphi_1$.

We assume that the two unknown tones are incoherent, i.e. $\Delta\varphi$ is random but slowly varying over the course of a measurement trial. (If the two tones are coherent, i.e. $\Delta\varphi$ is fixed, then super-resolution can also be achieved by using the method outlined here.) Therefore, the state of the oscillator is described by the density

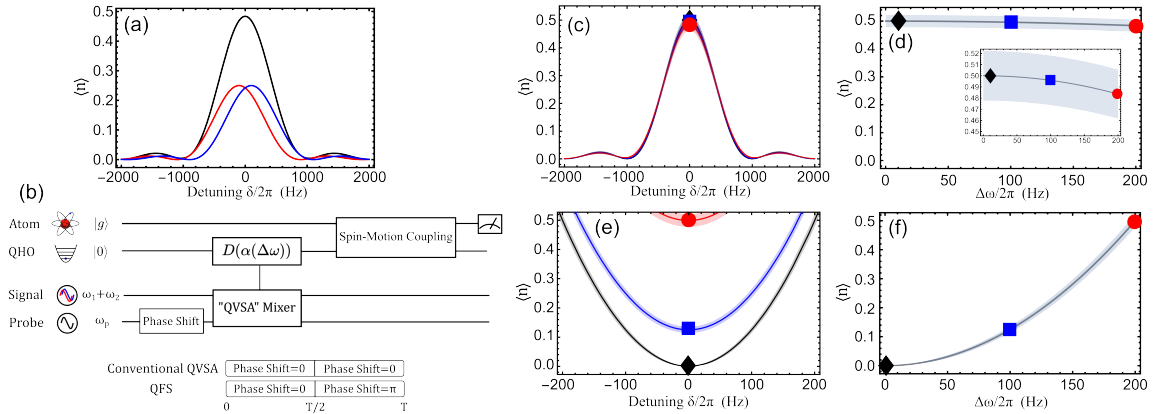


Figure 9.1: Spectroscopy of two incoherent tones using conventional quantum vector signal analysis (QVSA) and Quantum Fluctuation Suppression (QFS) techniques. (a) Conventional spectroscopy with two incoherent signal tones. When the separation between the two tones falls below the traditional linewidth limit, their corresponding peaks (red and blue traces) merge (black trace) and become unresolvable. (b) Quantum circuits implementing conventional QVSA and QFS protocols for detecting the frequency difference $\Delta\omega = \omega_2 - \omega_1$. The QFS protocol is implemented by applying a π phase shift to the probe tone at time $T/2$. The QVSA mixer is realized by a motional Raman transition and heterodynes the quadrupolar probe and unknown signal tones, resulting in a displacement of the QHO [WMH25]. Expected (c) conventional and (e) QFS spectroscopy generated by scanning the probe detuning (δ) at several different $\Delta\omega$. (c, d, e, f) Expected mean phonon number $\langle n \rangle$ under the conventional protocol (d) and the QFS protocol (f) as a function of $\Delta\omega$ at zero detuning $\delta = 0$. Shaded regions represent the uncertainty $\sigma_{\langle n \rangle}$ due to phonon shot noise for $N = 1000$ trials. (d) Using the conventional protocol, the dominant source of shot noise in the measured $\langle n \rangle$ is independent of $\Delta\omega$, obscuring any dependence of $\langle n \rangle$ on $\Delta\omega$ and rendering $\Delta\omega$ unresolvable. (f) Using the QFS protocol, sensitivity to $\Delta\omega$ is recovered by nullifying the $\Delta\omega$ -independent shot noise at $\delta = 0$. Here, the interrogation time is $T = 1$ ms.

matrix

$$\hat{\rho} = \frac{1}{2\pi} \int |\alpha(\Delta\omega, \Delta\varphi, T, 0)\rangle \langle\alpha(\Delta\omega, \Delta\varphi, T, 0)| d(\Delta\varphi) \quad (9.5)$$

$$= \frac{1}{2} (|\alpha(\Delta\omega, 0, T, 0)\rangle \langle\alpha(\Delta\omega, 0, T, 0)| + |\alpha(\Delta\omega, \pi, T, 0)\rangle \langle\alpha(\Delta\omega, \pi, T, 0)|), \quad (9.6)$$

and the phonon number is $\langle n \rangle = \frac{|\alpha(\Delta\omega, 0, T, 0)|^2 + |\alpha(\Delta\omega, \pi, T, 0)|^2}{2}$. By scanning the probe detuning δ , the spectrum of the unknown tones can be characterized at different $\Delta\omega$ (Fig. 9.1(c)).

However, in the super-resolution limit where $\Delta\omega T \ll 1$, it is straightforward to show that $|\alpha(\Delta\omega, 0, T, 0)|^2 \approx 4\Omega_{\text{eff}}^2 T^2 - \frac{1}{3}\Omega_{\text{eff}}^2 T^4 \Delta\omega^2$ and $|\alpha(\Delta\omega, \pi, T, 0)|^2 \approx \frac{1}{4}\Omega_{\text{eff}}^2 T^4 \Delta\omega^2$. The shot noise of a phonon measurement is given by σ_n , which, in the conventional protocol, results in the uncertainty of $\Delta\omega$ given by

$$\sigma_{\Delta\omega} = \frac{\sigma_n}{\partial n / \partial \Delta\omega} = \frac{12\sqrt{2}}{\Omega_{\text{eff}} T^3 \Delta\omega}, \quad (9.7)$$

which diverges as the two signal tones converge together to ω_μ ($\Delta\omega \rightarrow 0$), leading to Rayleigh's curse. Fundamentally, this divergence results from the shot noise introduced by the $\Delta\omega$ -independent displacement, $2\Omega_{\text{eff}}T$, which dominates and obscures any dependence on $\Delta\omega$, shown in Fig. 9.1(d). To achieve frequency super-resolution, this $\Delta\omega$ -independent displacement must therefore be removed.

9.1.1 Quantum Fluctuation Suppression

We've introduced a protocol for lifting Rayleigh's curse for a pair of electronic signals. To effect super-resolution, we apply a quantum fluctuation suppression (QFS) sequence by shifting the phase of the displacement operator by π at the midpoint of each interrogation time ($\frac{T}{2}$) [?]. The total displacement is

$$\begin{aligned} \alpha_{\text{SR}}(\Delta\omega, \Delta\varphi, T, 0) &= \alpha(\Delta\omega, \Delta\varphi, T/2, 0) \\ &\quad + e^{i\pi} \alpha(\Delta\omega, \Delta\varphi, T/2, T/2) \end{aligned} \quad (9.8)$$

which can be expanded to

$$\begin{aligned}
\alpha_{SR} &= \imath \Omega_{\text{eff}} \left(\frac{1 - e^{\imath(\delta - \frac{\Delta\omega}{2})\frac{T}{2}}}{\delta - \frac{\Delta\omega}{2}} + e^{-\imath\Delta\varphi} \frac{1 - e^{\imath(\delta + \frac{\Delta\omega}{2})\frac{T}{2}}}{\delta + \frac{\Delta\omega}{2}} \right) \\
&+ \imath e^{\imath\pi} \Omega_{\text{eff}} \left(e^{\imath(\delta - \frac{\Delta\omega}{2})\frac{T}{2}} \frac{1 - e^{\imath(\delta - \frac{\Delta\omega}{2})\frac{T}{2}}}{\delta - \delta\omega} + e^{\imath(\delta + \frac{\Delta\omega}{2})\frac{T}{2}} e^{-\imath\Delta\varphi} \frac{1 - e^{\imath(\delta + \frac{\Delta\omega}{2})\frac{T}{2}}}{\delta + \frac{\Delta\omega}{2}} \right) \\
&= \imath \Omega_{\text{eff}} \left(\frac{\left(1 - e^{\imath(\delta - \frac{\Delta\omega}{2})\frac{T}{2}}\right)^2}{\delta - \frac{\Delta\omega}{2}} + e^{-\imath\Delta\varphi} \frac{\left(1 - e^{\imath(\delta + \frac{\Delta\omega}{2})\frac{T}{2}}\right)^2}{\delta + \frac{\Delta\omega}{2}} \right) \quad (9.9)
\end{aligned}$$

For $\delta = 0$ – i.e. $\omega_q = \omega_\mu + \omega$, the super-resolution displacement can be reduced to

$$\alpha_{SR} = \imath \Omega_{\text{eff}} \left(e^{-\imath\Delta\varphi} \frac{\left(1 - e^{\imath\Delta\omega\frac{T}{4}}\right)^2}{\frac{\Delta\omega}{2}} - \frac{\left(1 - e^{-\imath\Delta\omega\frac{T}{4}}\right)^2}{\frac{\Delta\omega}{2}} \right). \quad (9.10)$$

The resultant mean phonon number $\langle n \rangle_{SR}$ for $\delta = 0$ is

$$\langle n \rangle_{SR} = |\alpha_{SR}|^2 = 256 \frac{\Omega_{\text{eff}}^2}{\Delta\omega^2} \sin^4 \left(\frac{\Delta\omega T}{8} \right) \sin^2 \left(\frac{\frac{\Delta\omega T}{2} - \Delta\varphi}{2} \right) \quad (9.11)$$

leading to Fig. 9.1(e). Operating in the super-resolution limit $\Delta\omega \ll \frac{1}{T}$, the mean phonon number for coherent tones with $\Delta\varphi = 0$ is

$$\langle n \rangle_{\Delta\varphi=0} \approx \frac{\Omega_{\text{eff}}^2}{256} \Delta\omega^4 T^6, \quad (9.12)$$

while $\Delta\varphi = \pi$ produces

$$\langle n \rangle_{\Delta\varphi=\pi} \approx \frac{\Omega_{\text{eff}}^2}{16} \Delta\omega^2 T^4. \quad (9.13)$$

For a pair of incoherent tones, the phase $\Delta\varphi$ will be random, and the mean phonon number will be calculated as

$$\langle n \rangle_{\Delta\varphi} = \langle |\alpha|^2 \rangle_{\Delta\varphi} = \frac{1}{2\pi} \int_0^{2\pi} 256 \frac{\Omega_{\text{eff}}^2}{\Delta\omega^2} \sin^4 \left(\frac{\Delta\omega T}{8} \right) \sin^2 \left(\frac{\Delta\omega T/2 - \Delta\varphi}{2} \right) d\Delta\varphi \quad (9.14)$$

$$= 128 \frac{\Omega_{\text{eff}}^2}{\Delta\omega^2} \sin^4 \left(\frac{\Delta\omega T}{8} \right). \quad (9.15)$$

As before, introducing the super-resolution limit $\Delta\omega T \ll 1$ allows Eq. 9.15 to become

$$\langle n \rangle_{SR} = \langle |\alpha_{SR}(\Delta\omega)|^2 \rangle_{\Delta\varphi} \approx \frac{\Omega_{\text{eff}}^2}{32} \Delta\omega^2 T^4. \quad (9.16)$$

As a result, $\Delta\omega$ becomes 'encoded' into the mean phonon number $\langle n \rangle_{\text{SR}}$, which can be efficiently measured in a QHO system, as shown in Fig. 9.1(f). The uncertainty in $\Delta\omega$ can be obtained from standard error propagation and Eq. 9.16, resulting in

$$\sigma_{\Delta\omega}^{SR} = \frac{\sigma_{n_{SR}}}{\partial n_{SR} / \partial \Delta\omega} = \frac{2\sqrt{2}}{\Omega_{\text{eff}} T^2}. \quad (9.17)$$

Notably, this result is independent of $\Delta\omega$, resulting in an uncertainty $\sigma_{\Delta\omega}$ that does not diverge as $\Delta\omega \rightarrow 0$, thereby enabling super-resolution measurement.

9.1.2 Quantum Fisher Information

An alternative way to consider the uncertainty of $\Delta\omega$ is to implement the Quantum Fisher Information (QFI). Fisher Information serves as a powerful tool for various sensing protocols and relies on an appropriate choice of estimator. For this protocol, the inversion estimator is employed to determine the mean phonon number $\langle n \rangle$ [WMH25]. Specifically, for a coherent state $|\alpha\rangle$, $\langle n \rangle$ can be inferred from the ratio of the motional sideband excitation probabilities P_r and P_b . Here, $P_{r/b}$ is the single-shot probability of a dark-state measurement following a red (blue) sideband Rabi pulse for duration $t_\pi = \pi/\Omega_{01}$, where Ω_{01} is the Rabi frequency of the $|^2S_{1/2}, n=0\rangle \leftrightarrow |^2D_{5/2}, n=1\rangle$ transition. This follows the same discussion provided in the motional Raman section.

To simulate incoherence between the signal tones, their relative phase $\Delta\varphi$ is randomly drawn from a uniform distribution over $[0, 2\pi]$ for each measurement. Despite this phase randomization, precise estimation of $\langle n \rangle$ remains possible. The estimator can be adapted by instead first averaging the detection probabilities over all repetitions before applying the inversion estimator. This phase-averaged estimator for $\langle n \rangle$ is thus:

$$\tilde{n} = g\left(\frac{\langle P_r \rangle_{\Delta\varphi}}{\langle P_b \rangle_{\Delta\varphi}}\right) = g\left(\frac{k/N}{j/M}\right), \quad (9.18)$$

where k (j) is the number of dark-state measurements observed in N (M) trials after application of a Rabi pulse on the red (blue) motional sideband. The conversion function $g(x)$ from the averaged sideband ratio to $\langle n \rangle$ is shown in Fig. 9.2.

The density matrix for a pure state parameterized by ϑ is $\rho_\vartheta = |\psi_\vartheta\rangle \langle \psi_\vartheta|$ and has

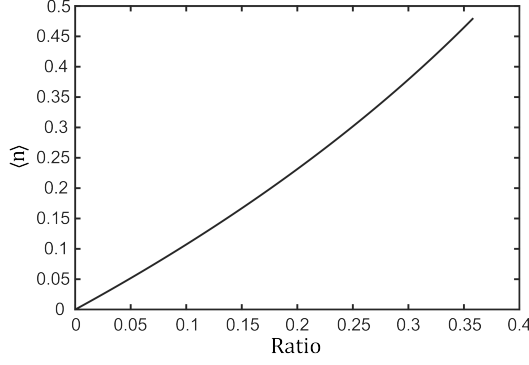


Figure 9.2: The conversion function $g(x)$ used to convert from a sideband ratio measurement to a mean phonon number $\langle n \rangle$ when the relative phase $\Delta\varphi$ varies randomly between measurements.

a Quantum Fisher Information (QFI)

$$\mathcal{F}_Q(|\psi_\vartheta\rangle) = 4 \left(\langle \dot{\psi}_\vartheta | \dot{\psi}_\vartheta \rangle - |\langle \dot{\psi}_\vartheta | \psi_\vartheta \rangle|^2 \right) \quad (9.19)$$

where $|\dot{\psi}_\vartheta\rangle = \frac{d|\psi_\vartheta\rangle}{d\vartheta}$. For frequency sensing, the parameter that our protocol senses is the frequency separation $\Delta\omega$ using a coherent state $|\alpha\rangle = e^{i\alpha(\hat{a} + \hat{a}^\dagger)}|0\rangle$. Using $|\dot{\alpha}\rangle = i\frac{\partial\alpha}{\partial(\Delta\omega)}(\hat{a} + \hat{a}^\dagger)|\alpha\rangle$, the QFI becomes

$$\mathcal{F}_Q(|\alpha_{\Delta\omega}\rangle) = 4 \left(\langle \dot{\alpha}_{\Delta\omega} | \dot{\alpha}_{\Delta\omega} \rangle - |\langle \dot{\alpha}_{\Delta\omega} | \alpha_{\Delta\omega} \rangle|^2 \right) = 4 \left| \frac{\partial\alpha}{\partial(\Delta\omega)} \right|^2. \quad (9.20)$$

From Eq. 9.15, the displacement after QFS takes the form

$$\langle \alpha_{\text{SR}} \rangle = \frac{16\Omega_{\text{eff}}}{\Delta\omega} \sin^2 \left(\frac{\Delta\omega T}{8} \right) \sin \left(\frac{\Delta\omega T/2 - \Delta\varphi}{2} \right). \quad (9.21)$$

In the super-resolved limit $\Delta\omega T \ll 1$, Eq. 9.20 thus simplifies to $\langle \alpha_{\text{SR}} \rangle = \frac{\Omega_{\text{eff}} T^2 \Delta\omega}{4} \sin \left(\frac{\frac{\Delta\omega T}{2} - \Delta\varphi}{2} \right)$.

The density matrix of the system under the QFS protocol is then obtained by integrating over the relative phase, written as

$$\hat{\rho} = \frac{1}{2\pi} \int |\alpha_{\text{SR}}(\Delta\omega, \Delta\varphi, T, 0)\rangle \langle \alpha_{\text{SR}}(\Delta\omega, \Delta\varphi, T, 0)| d(\Delta\varphi). \quad (9.22)$$

The QFI is similarly obtained using the same integration (convexity of the QFI [?]) to produce

$$\mathcal{F}_Q(|\alpha_{\Delta\omega}\rangle) \leq \frac{\Omega_{\text{eff}}^2 T^4}{8} (1 + (\Delta\omega T)^2) \approx \frac{\Omega_{\text{eff}}^2 T^4}{8}. \quad (9.23)$$

According to the quantum Cramér–Rao bound, $\sigma_{\Delta\omega}^Q = \frac{1}{\sqrt{\mathcal{F}_Q(\Delta\omega)}} \geq \frac{2\sqrt{2}}{\Omega_{\text{eff}} T^2}$. The QFS limit in Fig. 9.8(c) is based on this expression.

It is straightforward to see that this expression yields the same uncertainty as Eqn. 9.17, indicating that direct phonon-number measurements can saturate the quantum Cramér–Rao bound. Conceptually, QFS operates in phase space in a manner analogous to zero-delay Ramsey spectroscopy on the Bloch sphere. $\mathcal{F}_Q(\Delta\omega)$ effectively takes a form similar to that of Ramsey spectroscopy [WSH19], suggesting a potential for ultra-performant frequency sensing with our approach.

9.2 Experimental Procedure

Implementation of super-resolution involved the same experimental setup as was described in the previous chapter on subharmonic motional Raman. Since the premise of this experiment involves an additional tone and a phase change, modifications are made to the code that controls the AWG rather than the hardware. To verify and demonstrate our technique, we utilize a QHO realized on the axial motional mode of a single trapped $^{40}\text{Ca}^+$ ion with secular frequency $\omega_0 \approx 2\pi \cdot 700$ kHz. The axial mode is chosen for its ≥ 20 ms motional coherence time. The ground motional state of the axial mode is initially prepared via Doppler cooling of the ion followed by resolved-sideband cooling on the optical $^2S_{1/2} \leftrightarrow ^2D_{5/2}$ qubit transition to yield a mean phonon number $\langle n \rangle \approx 0.01(1)$.

For all experiments, the two “unknown” signal tones are applied as uniform electric fields by coupling voltages to the trap electrodes in a dipole configuration with frequencies $\omega_i = \omega_\mu \mp \Delta\omega/2$ for $i = 1, 2$ and relative phase $\Delta\varphi$. Control operations are applied exclusively to the probe tone, which is similarly applied, though in a quadrupole configuration. Prior to application of the super-resolution protocol, ω_μ is first obtained using conventional QVSA spectroscopy. While this scan is run by setting the two tones in-phase with one another, the effect of incoherent tones can be replicated by taking many different phases and averaging the results without accounting for the phase.

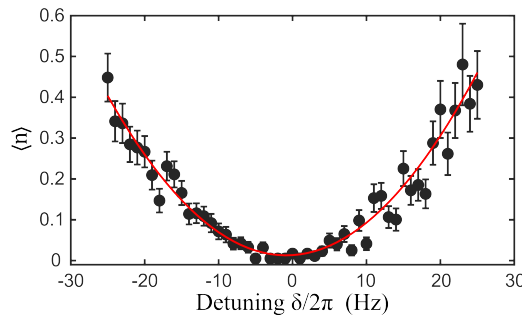


Figure 9.3: Optimizing the probe frequency ω_p for the super-resolution protocol. The mean phonon number $\langle n \rangle$ is measured while scanning the probe detuning δ as the QFS protocol is applied. The optimal probe frequency ω_p is determined by minimizing $\langle n \rangle$. The solid line is the theoretical fit to the data, which gives an uncertainty $\sigma_{\omega_p} \approx 2\pi \cdot 500$ mHz.

9.2.1 Optimizing Probe Frequency

The probe frequency ω_p is optimized by scanning the probe detuning δ as the QFS protocol is applied. ω_p is optimized when the measured $\langle n \rangle$ is minimized, as shown in Fig. 9.3. However, suppose the applied ω_p deviates from the optimal value by some frequency offset ω_{offset} . The measured mean phonon number $\langle n \rangle$ is then:

$$\langle n \rangle = \frac{\Omega_{\text{eff}}^2}{8} \left(\frac{\Delta\omega^2 T^4}{4} + \omega_{\text{offset}}^2 T^4 - \frac{\Delta\omega^2 \omega_{\text{offset}}^2 T^6}{4} \right) \quad (9.24)$$

For small ω_{offset} , i.e. $\omega_{\text{offset}}^2 T^2 \ll 1$, this expression simplifies to

$$\langle n \rangle = \frac{\Omega_{\text{eff}}^2}{8} \left(\frac{\Delta\omega^2 T^4}{4} + \omega_{\text{offset}}^2 T^4 \right). \quad (9.25)$$

Compared to the standard expression (Eq. 9.16), this frequency error introduces an additional offset independent of $\Delta\omega$. To correct for this offset, a two-point measurement can be performed, measuring $\langle n \rangle$ using our protocol by shifting ω_{offset} by a known $\Delta\omega$. By jointly analyzing the two measurements, nonzero ω_{offset} can be compensated. In our experiment, the value of ω_{offset} can be controlled to $\approx 2\pi \cdot 500$ mHz, as demonstrated in Fig. 9.3. However, the presence of a nonzero offset increases the uncertainty in $\langle n \rangle$ during an Allan deviation, artificially enlarging $\Delta\omega$ and degrading the measurement resolution.

9.2.2 Effective Rabi Frequency

The effective Rabi frequency Ω_{eff} determines the strength of the sensing protocol and as such is a value we want to accurately characterize. The straightforward approach is to simply measure the peak mean phonon number $\langle n \rangle$ of the spectroscopic lineshape with the probe frequency set to either $\omega_\mu \pm \omega_o$. However, this method is susceptible to drifts in ω_o . To mitigate these errors, a more robust method extracts Ω_{eff} by integrating the total lineshape, illustrated in the inset of Fig. 9.4. The shaded band represents the 68% prediction band. A function

$$\begin{aligned} \text{Area} &= \Omega_{\text{eff}}^2 \int_{-1/T}^{1/T} \langle |\alpha|^2 \rangle_{\Delta\varphi} df_p \\ &\approx 1.8 T \Omega_{\text{eff}}^2 \end{aligned} \quad (9.26)$$

is used for the fit, where the interrogation time T is set directly in hardware, and $f_p = \frac{\omega_p}{2\pi}$. This expression is accurate to within 2% as $\frac{T \Delta\omega}{2\pi} \leq 0.4$. Ω_{eff} , as used in Fig. 9.8, is then extracted via a calibration curve based on the measured phonon number, shown in Fig. 9.4. An interrogation time $T = 5$ ms is used. The resulting estimate of Ω_{eff} has a standard error of approximately 4%, as defined by the measured prediction band.

9.3 Results

9.3.1 Phase Dependence

As a first test, the super-resolution protocol is applied for $\Delta\varphi$ fixed at several values (0, 0.9π , and 1.9π). The measured $\langle n \rangle$ for $\omega_\mu = 2\pi \cdot 86$ MHz and $T = 1$ ms is shown in Fig. 9.5 (a) as a function of $\Delta\omega$, where lines depict the analytical result given by Eq. 9.15. All parameters in Eq. 9.15 are either calibrated experimentally or set accurately in hardware. To account for instability of the QHO frequency between measurements, we add a uniform offset when fitting Eq. 9.15 as the only free parameter. Fig. 9.5(b) further shows the measured $\langle n \rangle$ as a function of $\Delta\varphi$ at fixed $\Delta\omega = 2\pi \cdot 50$ Hz. The excellent agreement of experimental results with the theory indicates that, with only straightforward analysis, $\Delta\omega$ can be measured orders of magnitude below the traditional linewidth limit ($\sim 1/T = 1$ kHz) for fixed $\Delta\varphi$.

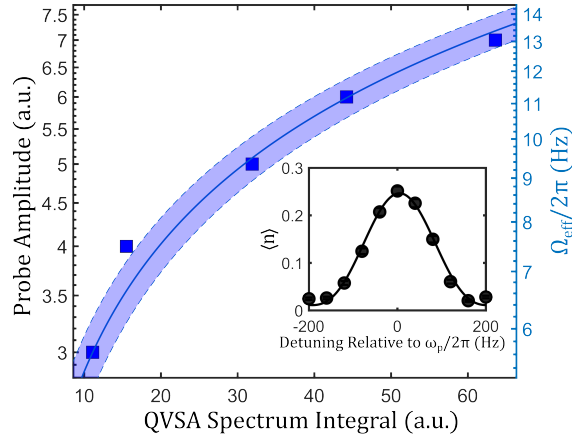


Figure 9.4: Robust calibration of the effective Rabi frequency Ω_{eff} . Ω_{eff} is measured by integrating the total lineshape, obtained via conventional spectroscopy. Data (blue squares) are fitted to a function (blue line) $\text{Area} \approx 1.8 T \Omega_{\text{eff}}^2$. The shaded band indicates the 68% prediction band, which corresponds to a 4% prediction error in Ω_{eff} . The inset displays a sample spectrum used to calculate an individual data point, here shown for an interrogation time $T = 5$ ms. Error bars in the inset are too small to be observed.

This phase relationship indicates that the protocol can be adapted to correlation spectroscopy to enable arbitrary resolution of $\Delta\omega$ [BCZ17,SGS17].

9.3.2 Wigner Reconstruction

The cancellation of excess shot noise, which enables the technique, can be understood by examining the evolution of the Wigner function in phase space. Fig. 9.6 shows *measured* Wigner functions of the ion motional state during its evolution under several variations of parameters. For these measurements, the QHO is initialized in the vacuum state $|0\rangle$, the displacement $\hat{D}(\gamma)$ is applied, and the Wigner functions recorded using the characteristic function tomography method of Ref. [FH20]. Each constituent Wigner function was experimentally measured on a 26×26 square grid using ≈ 100 averages for each of the real and imaginary parts, resulting in $\approx 1.3 \times 10^5$ data points per image. For $\Delta\omega = 0$, when the probe phase shifts by π at $T/2$, the motional trajectory retraces itself and returns to the origin, thereby canceling the $\Delta\omega$ -independent displacement. However, for nonzero $\Delta\omega$ the cancel-

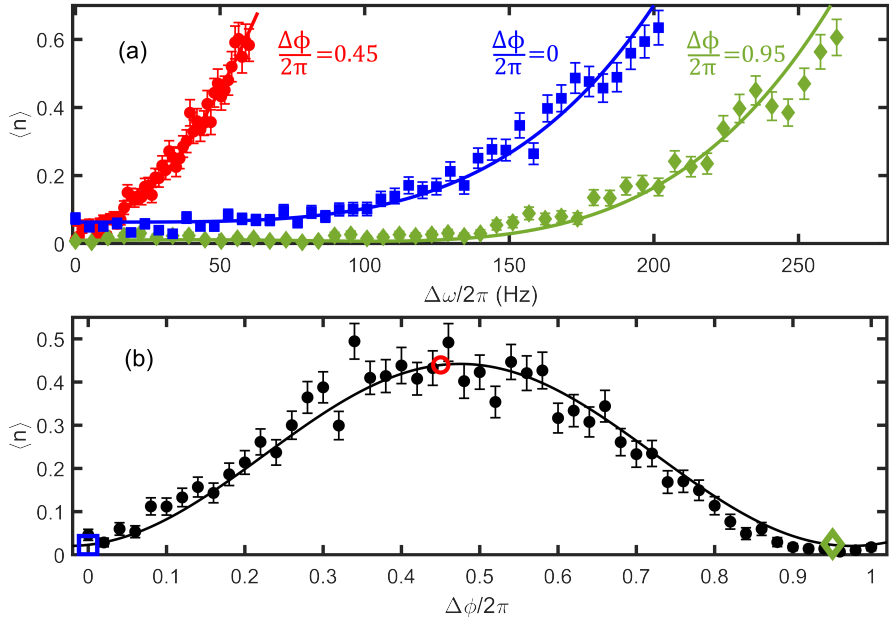


Figure 9.5: Dependence of the QFS protocol on the relative phase $\Delta\varphi$ between two signal tones $\omega_1 \approx \omega_2 \approx 2\pi \cdot 86$ MHz (a) Super-resolution spectrum of $\Delta\omega = \omega_2 - \omega_1$ for $\Delta\varphi = 0.9\pi$ (red, circle), $\Delta\varphi = 1.9\pi$ (green, diamond), and $\Delta\varphi = 0$ (blue, square). (b) Measured $\langle n \rangle$ as a function of $\Delta\varphi$ at $\Delta\omega/2\pi = 50$ Hz. Experimental data (dotted points) are in excellent agreement with fits to Eq. 9.15 (solid lines). Almost all parameters used in the fit are independently measured or controlled (e.g. Ω_{eff} and T), with a uniform vertical offset being the only fitting parameter. No additional vertical or horizontal scaling is applied. (a-b) A total interrogation time $T = 1$ ms was used. Error bars represent one standard error. Each point is averaged over $N = 600$ repetitions.

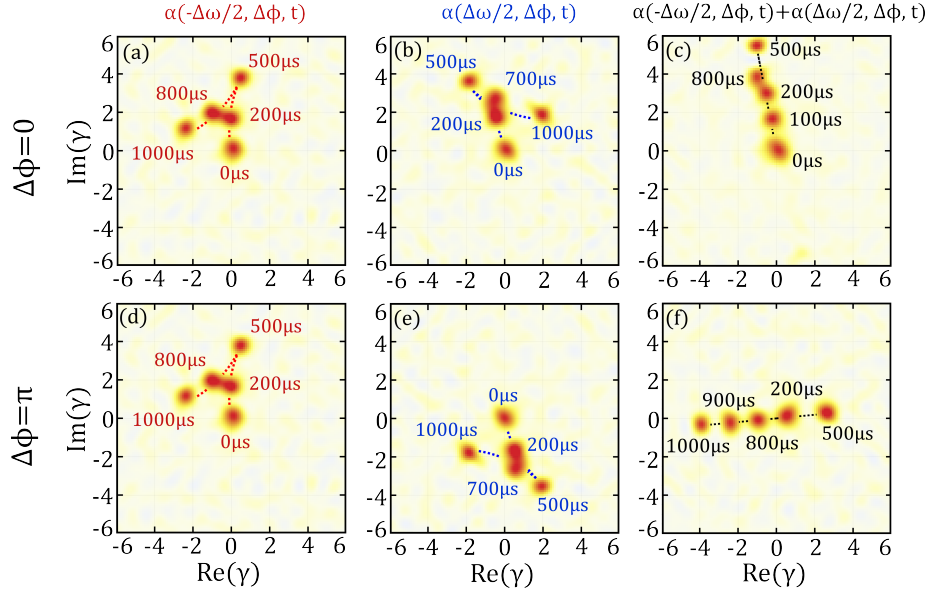


Figure 9.6: Phase space evolution of the QHO Wigner function under the QFS protocol for $\Delta\varphi = 0$ (a, b, c) and $\Delta\varphi = \pi$ (d, e, f) at $\Delta\omega/2\pi = 200$ Hz. The Wigner function is experimentally measured using motional state tomography. The blue (red) dashed lines represent the analytical phase-space trajectory due to evolution under a single tone. The black dashed lines show the resultant trajectory when both tones are applied. (c,f) Following phase reversal at $T = 500\mu s$, the motional trajectory retraces itself to remove the $\Delta\omega$ -independent displacement, leaving only a $\Delta\omega$ -dependent displacement.

lation becomes incomplete due to the differing phase accumulation $\pm\Delta\omega T/2$ during the evolution for the displacements arising from ω_1 and ω_2 . This behavior can be seen in Fig. 9.6(a–b) for $\Delta\varphi = 0$ and (c–d) for $\Delta\varphi = \pi$, with the first two columns showing the displacement of the individual signal tones and the last column showing the total displacement $\hat{D}(\alpha_{\text{SR}})$.

9.3.3 Ultimate Resolution

Next, to test a more general scenario where the two signal tones are incoherent, $\Delta\varphi$ is varied randomly between each measurement over a uniform distribution on $[0, 2\pi)$. Using the phase-averaged estimator, the relationship between $\Delta\omega$ and the mean phonon number $\langle n \rangle$ for two incoherent signal tones is characterized, shown in Fig. 9.7(a). Here, Ω_{eff} is independently measured and used in the fit. The solid line represents a fit of the data to the inversion function of Eq. 9.16 with a uniform horizontal offset as the sole free parameter. The shaded regions present the 68% prediction band for $\Delta\omega$ from the data. These narrow prediction bands, as shown in Fig. 9.1(f), provide clear evidence that this protocol can be used to achieve super-resolution on even incoherent signals.

Following this demonstration, an Allan deviation is performed at six different values of $\Delta\omega$ to evaluate the ultimate achievable resolution for $\Delta\omega$ with known Ω_{eff} from pre-calibration, presented in Fig. 9.7(b–d). Here, the dashed lines indicate the expected $1/\sqrt{N}$ scaling. Deviations from this trend result from systematic sources of error, and are chiefly due to instability in the natural frequency of the QHO. After $N = 8160$ repetitions, the uncertainty in $\Delta\omega$ falls below the single-Hz level, and is nearly identical across all values of $\Delta\omega$, consistent with the theoretical prediction that, in the super-resolved measurement regime, the uncertainty is independent of $\Delta\omega$. To resolve signals separated by 5 Hz, our QFS protocol requires a measurement time of 110 s. By comparison, spectroscopy using a conventional protocol would require a measurement time of roughly $3 \cdot 10^7$ s to achieve a similar resolution - this is a result that would be practically unachievable at any level without even factoring in systematic sources of error such as long-term drifts.

As a final demonstration to characterize the precision and accuracy of our super-

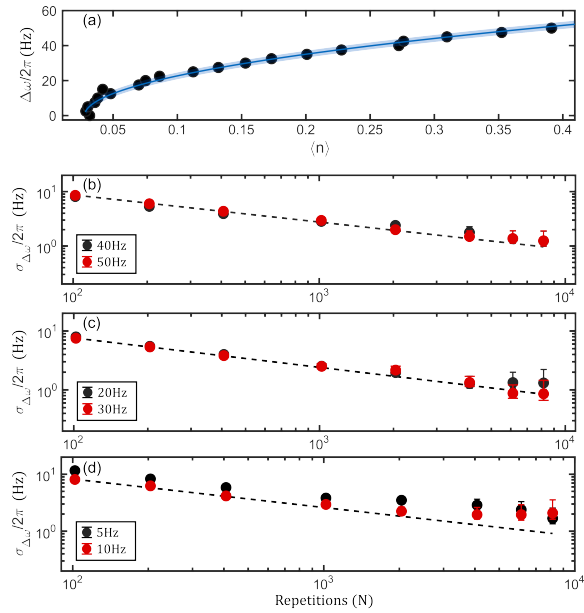


Figure 9.7: Characterizing ultimate resolution of $\Delta\omega$ with two incoherent signal tones centered at $2\pi \cdot 86$ MHz. (a) Calibration curve used to convert the measured $\langle n \rangle$ to the parameter $\Delta\omega$. Experimental data is plotted as dotted points and fitted to an inversion of the analytical model (solid lines) in Eq. 9.16 with only a constant horizontal offset as the sole fit parameter. The colored band indicates the 68% prediction band. Each point is averaged over $N = 5000$ measurements. There is no additional rescaling. (b-d) Overlapping Allan deviations of $\Delta\omega$ at different frequency separations $\Delta\omega$ with fixed “known” Rabi frequency Ω_{eff} . (b) $\Delta\omega/2\pi = 40, 50$ Hz. (c) $\Delta\omega/2\pi = 20, 30$ Hz. (d) $\Delta\omega/2\pi = 5, 10$ Hz. (a-d) Data was taken using a total interrogation time $T = 1$ ms. To simulate incoherence, the relative phase between the two signal tones $\Delta\varphi$ was varied randomly between each measurement from a uniform distribution on $[0, 2\pi)$. Error bars represent one standard error. The dashed lines represent a $1/\sqrt{N}$ trend. Each repetition takes ≈ 13 ms.

resolution protocol, we apply and sense two unknown, incoherent signals. We first optimize the probe frequency ω_p by scanning the probe detuning δ with our QFS protocol and minimizing the measured mean phonon number $\langle n \rangle$. The effective Rabi frequency Ω_{eff} is extracted by performing conventional spectroscopy, with a resulting prediction error $(\sigma_{\Omega_{\text{eff}}}) \approx 4\%$ based on the measured phonon number. Next, we conduct a super-resolution measurement with interrogation time $T = 1$ ms at the optimized ω_p to determine $\Delta\omega$. To suppress the effect of drifts in ω_o , a reference signal with known $\Delta\omega$ is concurrently measured. The distribution of the measured $\omega_\mu \pm \Delta\omega/2$ is shown in Fig. 9.8(a) for $\Delta\omega = 2\pi \cdot 30$ Hz (see Appendix I in SI for further data at different $\Delta\omega$). As expected, two incoherent spectral components (blue and red) are clearly resolved at frequencies $\omega_\mu \pm \Delta\omega/2$, in contrast to the conventional method, shown in purple. This process is repeated at several values of $\Delta\omega$ to compare the measured values $\Delta\omega$ to their applied true values in Fig. 9.8(b). The excellent direct correspondence of our measurements to the true values in Fig. 9.8(b) demonstrates the accuracy and reliability of our super-resolution protocol.

To contextualize the frequency resolution achievable with our super-resolution protocol, we compare our resolution limits against those of the conventional protocol, depicted in Fig. 9.8(c). The $\sigma_{\Delta\omega} = \Delta\omega$ line demarcates the resolvable from the unresolvable regions. The uncertainty using the conventional protocol crosses this bound at $\Delta\omega \approx 2\pi \cdot 66$ Hz using an interrogation time $T = 1$ ms and $N = 8160$ repetitions. In contrast, the QFS limit is $\sigma_{\Delta\omega}^{(\text{SR})} = 2\pi \cdot 0.65$ Hz for the same parameters, indicating that the spectrally resolvable frequency range can be extended by more than two orders of magnitude. The QFS limit corresponds to the lowest possible value set by both the corresponding quantum Cramér–Rao limit and standard error propagation estimate. In practice, however, our super-resolution protocol has extended the resolvable frequency range by approximately $\approx 14\times$. Deviations of the measured uncertainty from $\sigma_{\Delta\omega}^{(\text{SR})}$ arise from a combination of unsaturated estimators [WMH25], uncertainty in Ω_{eff} and ω_μ , and systematic errors due to drifts in ω_o when applying the QFS protocol. Notably, at $\Delta\omega/2\pi = 5$ Hz our protocol achieves a resolution that requires $> 10^5$ fewer experimental repetitions than the conventional approach, without considering even systematic sources of error in the latter. This enhanced resolution demonstrated by our protocol allows determination

of $\Delta\omega$ at levels 200 times beyond the spectral linewidth limit ($\sim 1/T = 1$ kHz). We have also demonstrated broadband capability by resolving two signals separated by $\Delta\omega/2\pi = 20$ Hz across a range of order 10 MHz to the GHz regime; please see Appendix L for further details.

In order to show broadband capability, we performed additional measurements to demonstrate that, even up to the GHz range, our protocol is still able to resolve two tones separated by a factor of 50 below the system linewidth (~ 1 kHz). Fig. ?? highlights the broadband capability of our protocol. The lowest frequency at which we measure is 30 MHz, set by the 30 MHz cutoff frequency of the high-pass filters in our system used to suppress noise near the ion's secular frequency (≈ 1 MHz). This low frequency limit is set by trap electronics and is easily extended to the kHz regime with more judicious signal conditioning. The highest frequency we measure is 934 MHz, which is set by the available power of our current electronics. This high frequency limit is easily extendable with better amplifiers.

As for the theoretical frequency range of our method, we can understand this frequency performance through the quantum Fisher information, $\mathcal{F}(\Delta\omega) \leq \frac{(\Omega_{\text{eff}} T^2)^2}{8}$, where the precision of $\Delta\omega$ is determined by T and $\Omega_{\text{eff}} = \frac{\Omega_d \Omega_p \omega_0}{(\omega_{1/2}^2 - \omega_0^2)}$. Wideband performance introduces the condition $\omega_{1/2} \gg \omega_0$. The precision in $\Delta\omega$ thus scales with $\omega_{1/2}^2$, the same as the resolvable $\Delta\omega$. The ideal resolution is shown by the blue curve in Fig. 9.10, calculated using a probe power of 40 W. Under these ideal conditions, our protocol is able to resolve two tones separated by 10 Hz at even GHz frequencies — i.e. a factor $\times 100$ smaller than the 1 kHz linewidth. In practice, however, practical power constraints and anharmonicity of the ion trap limit the maximum coherent displacement and therefore the usable Ω_{eff} . For $T = 1$ ms, we find empirically that $\Omega_{\text{eff}}/2\pi < 1.7$ kHz is required to maintain high QFS fidelity. Consequently, the resolvable $\Delta\omega$ remains roughly constant across a wide frequency range and increases only when the applied power reaches this limit at 700 MHz, shown by the red trace in Fig. 9.10.

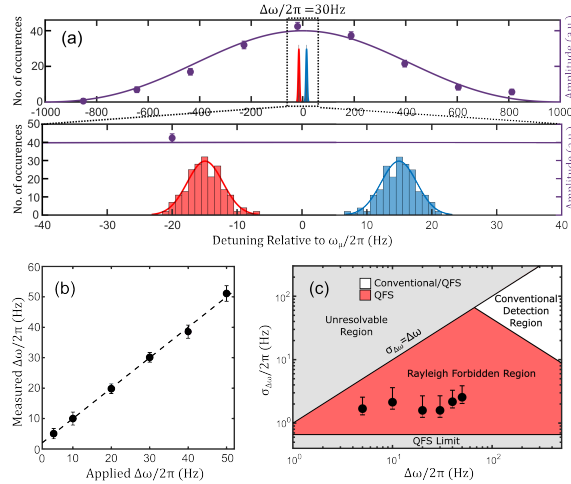


Figure 9.8: Measurement of “unknown” $\Delta\omega$ with two incoherent signal tones at $\omega_\mu = 2\pi \cdot 86$ MHz with all sources of error accounted for. (a) Distribution of measurements of $\omega_\mu \pm \Delta\omega/2$ for $\Delta\omega/2\pi = 30$ Hz with QFS protocol (red and blue). For comparison, measurements (purple circles) and a corresponding fit (purple line) are shown for a conventional protocol. The solid red and blue lines depict Poisson fits to the measured distributions. Each occurrence is averaged over $N = 102$ repetitions. (b) Measured values of $\Delta\omega$ compared against the true values. The dashed line represents an ideal 1:1 correspondence between the applied and measured values. (c) Comparison of resolvable regions between conventional and QFS protocols. The conventional detection region indicates the region resolvable by both conventional and QFS protocols. The QFS protocol extends the resolvable region into the Rayleigh-forbidden region—beyond the traditional limit imposed by linewidth—reaching the bound set by lowest possible value of the quantum Cramér–Rao limit, referred to here as the QFS limit. Beyond $\sigma_{\Delta\omega} = \Delta\omega$, $\Delta\omega$ becomes unresolvable (grey). At $\Delta\omega/2\pi = 5$ Hz, the QFS protocol requires 10^5 fewer repetitions than a conventional protocol to achieve the same uncertainty. (b-c) An interrogation time of $T = 1$ ms is used with $N = 8160$ repetitions. Error bars represent one standard error.

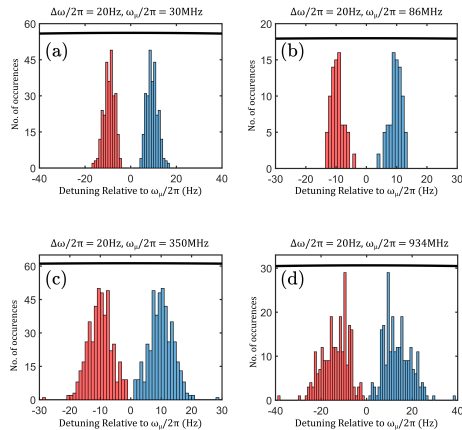


Figure 9.9: Summary of histograms of super-resolution for different signal frequencies(ω_μ) with $\Delta\omega/2\pi = 20$ Hz, which is a factor 50 lower than Fourier-transform-limited linewidth (~ 1 kHz). The purple solid line is the result of conventional QVSA measurement. The red/blue solid lines are fits to the data assuming a Poisson distribution. Each subfigure is averaged over M repetitions. (a) $M=102$, $\omega_\mu = (\omega_1 + \omega_2)/2 = 2\pi \cdot 30$ MHz; (b) $M=255$, $\omega_\mu = 2\pi \cdot 86$ MHz; (c) $M=51$, $\omega_\mu = 2\pi \cdot 350$ MHz; (d) $M=102$, $\omega_\mu = 2\pi \cdot 934$ MHz; $T = 1$ ms.

9.4 Further Discussion

9.4.1 Higher-Order Phase Reversal

In the main text, the super-resolution demonstration is implemented using a single π phase switch. In principle, our QHO displacement protocol can be extended to include multiple phase reversals. Figure 9.12 plots the resulting mean phonon number for these higher-order implementations (i.e., with more phase reversals). We consider only odd orders as only these achieve full suppression of excess shot noise from $\Delta\omega$ -independent contributions. For clarity, we use standard error propagation to illustrate the relative efficiency of different orders. The uncertainty in estimating $\Delta\omega$ is

$$\sigma_{\Delta\omega} = \frac{\Delta n}{\frac{\partial n}{\partial \Delta\omega}}. \quad (9.27)$$

If the resulting mean phonon number $\bar{n}$ is fixed, then the shot noise Δn is also fixed for all orders. In this case, a larger slope $\frac{\partial n}{\partial \Delta\omega}$ corresponds to a smaller uncertainty. As shown in Fig. 9.12, the first-order protocol provides the steepest slope and therefore

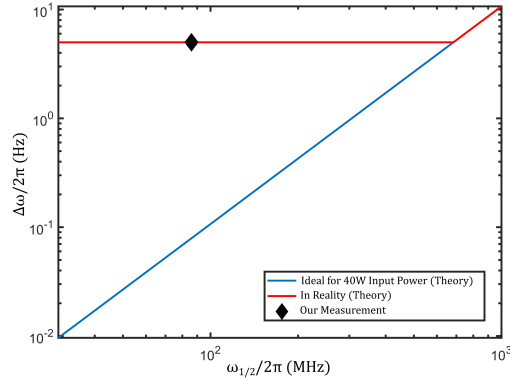


Figure 9.10: Resolvable $\Delta\omega$ as a function of the signal frequency $\omega_{1/2}^2$. (Blue curve) Resolution in the ideal case, calculated with a probe power of 40W. As our super-resolution protocol relies on a motional Raman interaction, the resolution (Ω_{eff}) scales inversely with $\omega_{1/2}^2$. In practice, power constraints and trap anharmonicity limit the maximum coherent displacement and the concomitant protocol fidelity, which in turn limits the maximum Ω_{eff} . In our system, we find empirically that $\Omega_{\text{eff}}/2\pi < 1.7$ kHz is required to maintain high fidelity for the QFS protocol. (Red curve) Predicted achievable resolution when accounting for power and anharmonicity constraints, i.e. $\Omega_{\text{eff}}/2\pi < 1.7$ kHz. The resolution degrades at ≈ 700 MHz due to power constraints. (Black diamonds) Experimental results for the best-case resolution at $\omega_{1/2}/2\pi = 86$ MHz using an interrogation time $T = 1$ ms.

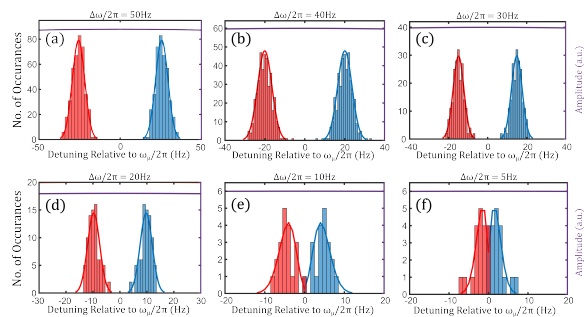


Figure 9.11: Summary of histograms of super-resolution for different $\Delta\omega$ measurements. The purple solid line is the result of conventional QVSA measurement. The red/blue solid lines are fits to the data assuming a Poisson distribution. Each subfigure is averaged over M repetitions. (a-b) $M=51$; (c) $M=102$; (d) $M=255$; (e) $M=510$; (f) $M=1020$

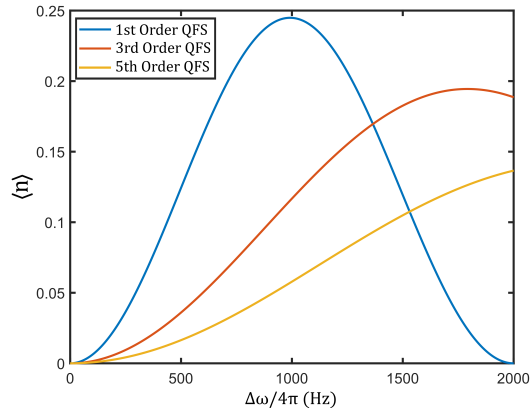


Figure 9.12: Resultant mean phonon number calculated as a function of the signal frequency separation $\Delta\omega$ under different QFS orders (number of phase reversals). Only odd QFS orders are shown as only these achieve full suppression of excess shot noise from $\Delta\omega$ -independent contributions. Calculations are done using identical Ω_{eff} and total pulse times $T = 1$ ms for all depicted cases.

the smallest uncertainty. For this reason, we employed only the first-order sequence in our experiment.

9.4.2 Amplitude Imbalance

When the amplitudes of the two signal tones are unequal, the spectroscopic peak shifts from the midpoint $\frac{\omega_1 + \omega_2}{2}$ to a weighted average:

$$\omega_{\text{peak}} = \frac{\Omega_1 \omega_1 + \Omega_2 \omega_2}{\Omega_1 + \Omega_2}, \quad (9.28)$$

where Ω_1 and Ω_2 are the respective effective Rabi frequencies. The integrated line-shape is thus

$$\text{Area} \approx 0.9T (\Omega_1^2 + \Omega_2^2). \quad (9.29)$$

Similarly, the mean phonon number when the relative phase $\Delta\varphi$ is randomized is:

$$\langle |\alpha|^2 \rangle_{\Delta\varphi} \approx \frac{1}{8} \left(\frac{\Omega_1 \Omega_2}{\Omega_1 + \Omega_2} \right)^2 \Delta\omega^2 T^4. \quad (9.30)$$

Measurement of the ratio $R = \Omega_1/\Omega_2$ is thus required to determine $\Delta\omega$. Three approaches can be used to determine the amplitude ratio R . The first is a classical, self-homodyne technique that uses typical laboratory test and measurement equipment (e.g. an oscilloscope, a spectrum analyzer). The input signals can be expressed

as

$$f(t) = a_1 \cos(\omega_1 t) + a_2 \cos(\omega_2 t) \quad (9.31)$$

The inputs are split with equal amplitude using an RF splitter and fed into the LO and RF ports of a mixer, producing an output

$$f(t) = \frac{a_1^2 + a_2^2}{2} + \frac{a_1^2}{2} \cos(2\omega_1 t) + \frac{a_2^2}{2} \cos(2\omega_2 t) + a_1 a_2 (\cos((\omega_1 - \omega_2)t) + \cos((\omega_1 + \omega_2)t)). \quad (9.32)$$

The DC component $\frac{a_1^2 + a_2^2}{2}$ can be isolated by applying a low-pass filter to the output and measuring the signal on an oscilloscope. Similarly, in the super-resolution regime where $2\omega_1 \approx 2\omega_2 \approx \omega_1 + \omega_2$, the $a_1^2 + a_2^2 + 2a_1 a_2$ component can be measured it on a spectrum analyzer. Together, these measurements can be processed to yield the desired ratio $R = \frac{a_1}{a_2} = \frac{\Omega_1}{\Omega_2}$.

The second approach combines quantum (ion-based) measurements with classical techniques. A key challenge with quantum systems is that the Schrödinger equation is inherently linear, making it difficult to obtain nonlinear cross-terms, e.g. $a_1 a_2$. The requisite nonlinearity can instead be introduced classically by, e.g., applying the inputs to an RF mixer, as in the classical method. However, instead of using traditional test and measurement equipment for detection, the mixed outputs can be detected using the quantum system at hand; here, the trapped ion. Detection of the mixed signal using the technique in Fig. 9.4 now yields $\Omega_1^2 + \Omega_2^2 + 2\Omega_1 \Omega_2$, which can be combined with the direct measurement of Eq. 9.30 to yield the amplitude ratio R .

Finally, the amplitude ratio can be detected using only the QHO system at hand. Excitation of a nonlinear, second-order subharmonic generates an intermodulation term proportional to $\Omega_1 \Omega_2$, effectively functioning like the RF mixer in the classical method [WMH25]. Specifically, integration of the lineshape obtained by a scan of ω_p about the second-order subharmonic gives:

$$\text{Area}_2 \propto \Omega_1^2 + \Omega_2^2 + 2\Omega_1 \Omega_2, \quad (9.33)$$

where proportionality factors can be derived from the relevant theory. This complements the measurement in Eq. 9.28 to allow exact calculation of the amplitude ratio R .

CHAPTER 10

CAT Interferometry

10.1 CAT Interferometry

Implementing CAT states for enhancing a displacement measurement has been done in [C 13, WDI26, C M96]. Our goal is to extend it into applications in mass spectrometry since mass spectrometry is an essential tool in modern science and technology, where it is used to enable tests of fundamental physics and chemistry [HC13], identify biological samples in hospital and forensic laboratories, and discover and produce pharmaceuticals. Previous theoretical discussions have discussed implementing quantum enhancement into mass spec [Rod11, AC24, SP04], but so far a dedicated implementation hasn't been demonstrated. To confirm the mass of the dark ion that we load for EGGs is truly H^{35}Cl^+ , we decided to implement a quantum mass spec technique using CAT states.

10.1.1 Theory

In order to generate a spin-motional CAT state, we used bichromatic tones at the motional sidebands of the quadrupole transition [Hem14]. Similar to the calculation performed to get Eqn. 3.39, the full Hamiltonian will be represented by

$$\begin{aligned} \frac{H}{\hbar} = & \omega_0 \hat{a}^\dagger \hat{a} + \sum_{i=0}^N \frac{\Delta}{2} \sigma_z^{(i)} + \Omega_b \sigma_x^{(i)} ((\cos(\omega_b t - \varphi_b) + \eta(\hat{a} + \hat{a}^\dagger) \sin(\omega_b t - \varphi_b)) \\ & + \Omega_r \sigma_x^{(i)} ((\cos(\omega_r t - \varphi_r) + \eta(\hat{a} + \hat{a}^\dagger) \sin(\omega_r t - \varphi_r)) \end{aligned} \quad (10.1)$$

Moving into two different interaction pictures will simplify this Hamiltonian, with the first step transforming into the interaction picture of the atomic energy level. In this frame, the operators $\sigma_\pm$ transform to $\sigma_\pm e^{\pm i\Delta t}$, such that this interaction Hamiltonian

is

$$\begin{aligned} \frac{H_I}{\hbar} = \omega_0 \hat{a}^\dagger \hat{a} + \sum_{i=0}^N (\sigma_+^{(i)} e^{i\Delta t} + \sigma_-^{(i)} e^{-i\Delta t}) (\Omega_b ((\cos(\omega_b t - \varphi_b) + \eta(\hat{a} + \hat{a}^\dagger) \sin(\omega_b t - \varphi_b)) \\ + \Omega_r ((\cos(\omega_r t - \varphi_r) + \eta(\hat{a} + \hat{a}^\dagger) \sin(\omega_r t - \varphi_r))). \end{aligned} \quad (10.2)$$

The second interaction picture involves moving into the ion frame. In this case the operator $\hat{a}$ and $\hat{a}^\dagger$ transform into $\hat{a}e^{-i\omega_0 t}$ and $\hat{a}^\dagger e^{i\omega_0 t}$, respectively. Additionally, the 0^{th} order term in the Lamb-Dicke approximation will drop out, resulting in

$$\begin{aligned} \frac{H_I}{\hbar} = \sum_{i=0}^N (\sigma_+^{(i)} e^{i\Delta t} + \sigma_-^{(i)} e^{-i\Delta t}) (\Omega_b (\eta(\hat{a}e^{-i\omega_0 t} + \hat{a}^\dagger e^{i\omega_0 t}) \sin(\omega_b t - \varphi_b)) \\ + \Omega_r (\eta(\hat{a}e^{-i\omega_0 t} + \hat{a}^\dagger e^{i\omega_0 t}) \sin(\omega_r t - \varphi_r))). \end{aligned} \quad (10.3)$$

Once in the interaction picture, applying the necessary rotating wave approximations results in non-vanishing terms related to the smallest detuning, i.e. $\omega_b - (\Delta + \omega_0)$ and $\omega_r - (\Delta - \omega_0)$. Since ω_b , ω_r , and Δ are all optical frequencies, it should be clear that we must have the difference between them. The secular frequency of the trap, ω_0 , is on the order of MHz, whereas the smallest detunings we care about keeping are on the order of kHz or smaller, which motivates subtracting ω_0 from ω_b , and adding ω_0 to ω_r . After performing the approximations, the resulting Hamiltonian is

$$\begin{aligned} \frac{H_I}{\hbar} = \sum_{i=0}^N \left(\frac{\Omega_b \eta}{2i} (\sigma_-^{(i)} \hat{a} e^{i((\omega_b - \Delta - \omega_0)t - \varphi_b)} - \sigma_+^{(i)} \hat{a}^\dagger e^{-i((\omega_b - \Delta - \omega_0)t - \varphi_b)}) \right. \\ \left. + \frac{\Omega_r \eta}{2i} (\sigma_-^{(i)} \hat{a}^\dagger e^{i((\omega_r - \Delta + \omega_0)t - \varphi_r)} - \sigma_+^{(i)} \hat{a} e^{-i((\omega_r - \Delta + \omega_0)t - \varphi_r)}) \right). \end{aligned} \quad (10.4)$$

For the BSB tone, there is a detuning $\delta_b = \omega_b - \Delta - \omega_0$ and likewise for the RSB tone, the detuning is $\delta_r = \omega_r - \Delta + \omega_0$, such that

$$\frac{H_I}{\hbar} = \sum_{i=0}^N \left(\frac{\Omega_b \eta}{2i} (\sigma_-^{(i)} \hat{a} e^{i(\delta_b t - \varphi_b)} - \sigma_+^{(i)} \hat{a}^\dagger e^{-i(\delta_b t - \varphi_b)}) + \frac{\Omega_r \eta}{2i} (\sigma_-^{(i)} \hat{a}^\dagger e^{i(\delta_r t - \varphi_r)} - \sigma_+^{(i)} \hat{a} e^{-i(\delta_r t - \varphi_r)}) \right). \quad (10.5)$$

This definition of detunings will be short-lived as I redefine the detunings into the components from a carrier detuning and a secular frequency detuning, i.e. $\delta_b = \delta_c + \delta_s$ and $\delta_r = \delta_c - \delta_s$. We can similarly break down the phases into a symmetric and asymmetric component, $\varphi_b = \varphi_+ + \varphi_-$ and $\varphi_r = \varphi_+ - \varphi_-$. Lastly, for ease, we will assume the strength of each laser is the same $\Omega_b = \Omega_r = \Omega$,

$$\frac{H_I}{\hbar} = \sum_{i=0}^N \frac{\Omega \eta}{2i} (\sigma_-^{(i)} e^{i(\delta_c t - \varphi_+)} - \sigma_+^{(i)} e^{-i(\delta_c t - \varphi_+)}) (\hat{a}^\dagger e^{-i(\delta_s t - \varphi_-)} + \hat{a} e^{i(\delta_s t - \varphi_-)}) \quad (10.6)$$

Rewriting the complex value in the denominator as an exponential ($\imath = e^{\imath\frac{\pi}{2}}$), this Hamiltonian can be further simplified to

$$\frac{H_I}{\hbar} = \sum_{i=0}^N \frac{\Omega\eta}{2} (\sigma_-^{(i)} e^{\imath(\delta_c t - \varphi_+ - \frac{\pi}{2})} + \sigma_+^{(i)} e^{-\imath(\delta_c t - \varphi_+ - \frac{\pi}{2})}) (\hat{a}^\dagger e^{-\imath(\delta_s t - \varphi_-)} + \hat{a} e^{\imath(\delta_s t - \varphi_-)}) \quad (10.7)$$

or, when using

$$\sigma_\varphi = \sigma_+ e^{-\imath\varphi} + \sigma_- e^{\imath\varphi} = \cos(\varphi)\sigma_x + \sin(\varphi)\sigma_y \quad (10.8)$$

$$\frac{H_I}{\hbar} = \sum_{i=0}^N \frac{\Omega\eta}{2} \sigma_{\varphi'}^{(i)} (\hat{a}^\dagger e^{-\imath(\delta_s t - \varphi_-)} + \hat{a} e^{\imath(\delta_s t - \varphi_-)}) \quad (10.9)$$

with $\varphi' = \delta_c t - \varphi_+ - \frac{\pi}{2}$.

For the application in mass spectrometry, we are interested in the following sequence: CAT, displace, Anti-CAT. For simplicity we will use the assumption that we are on resonance and there are no detunings. For our CAT pulse, the phases φ_+ and φ_- are 0, and for the Anti-CAT pulse, $\varphi_- = \pi$. This difference in phase will end up changing the phase of the displacement, hence why its for the Anti-CAT pulse,

$$\frac{H_{cat}}{\hbar} = \sum_{i=0}^N -\frac{\Omega\eta}{2} \sigma_y^{(i)} (\hat{a}^\dagger + \hat{a}). \quad (10.10)$$

Generating the time evolution operator for this Hamiltonian allows us to define the displacement produced by the bichromatic light. For ease, this will be calculated for a single ion interacting with the lasers

$$U_{cat} = e^{-\imath \frac{H_{cat}}{\hbar} t} = e^{\frac{\imath\eta\Omega t}{2} \sigma_y (\hat{a}^\dagger + \hat{a})}. \quad (10.11)$$

If we ignore the spin operator σ_y , it is obvious that this would create a displacement operator $\hat{D}(\alpha)$ with $\alpha = \alpha_r + \imath\alpha_i$, where α_r and α_i are the displacements in the real and imaginary quadratures, respectively. Intuitively, it should make sense that the total displacement α can be cleanly broken into its real and imaginary components, but for completeness

$$\begin{aligned} \hat{D}(\alpha = \alpha_r + \imath\alpha_i) &= e^{\alpha\hat{a}^\dagger - \alpha^*\hat{a}} = e^{\alpha_r(\hat{a}^\dagger - \hat{a}) + \imath\alpha_i(\hat{a}^\dagger + \hat{a})} \\ &= e^{-\frac{\alpha_r(\imath\alpha_i)^* - \alpha_r^*\imath\alpha_i}{2}} \hat{D}(\alpha_r) \hat{D}(\imath\alpha_i) = \hat{D}(\alpha_r) \hat{D}(\imath\alpha_i) \end{aligned} \quad (10.12)$$

Returning to our unitary 10.11, if the spin is ignored, then it would create a displacement with a value $\alpha_{cat} = \eta \frac{\Omega t}{2}$. It's trivial to show that for the Anti-CAT case, when $\varphi_- = \pi$, then the displacement is $-\alpha_{cat}$.

However, ignoring the spin dependence that exists in Eqn. 10.11 would be incorrect, and as such, by expanding the unitary exponential for some σ_φ ,

$$\begin{aligned}
U &= e^{(\alpha \hat{a}^\dagger - \alpha^* \hat{a}) \sigma_\varphi} = \cosh(\alpha \hat{a}^\dagger - \alpha^* \hat{a}) \mathbb{1} + \sinh(\alpha \hat{a}^\dagger - \alpha^* \hat{a}) \sigma_\varphi \\
&= \frac{e^{(\alpha \hat{a}^\dagger - \alpha^* \hat{a})} + e^{-(\alpha \hat{a}^\dagger - \alpha^* \hat{a})}}{2} \mathbb{1} + \frac{e^{(\alpha \hat{a}^\dagger - \alpha^* \hat{a})} - e^{-(\alpha \hat{a}^\dagger - \alpha^* \hat{a})}}{2} \sigma_\varphi \\
&= \frac{\hat{D}(\alpha) + \hat{D}(-\alpha)}{2} \mathbb{1} + \frac{\hat{D}(\alpha) - \hat{D}(-\alpha)}{2} \sigma_\varphi \\
&= \frac{\hat{D}(\alpha)}{2} (\mathbb{1} + \sigma_\varphi) + \frac{\hat{D}(-\alpha)}{2} (\mathbb{1} - \sigma_\varphi) \\
&= \frac{\hat{D}(\alpha)}{\sqrt{2}} e^{i \frac{\pi}{4} \sigma_\varphi} + \frac{\hat{D}(-\alpha)}{\sqrt{2}} e^{-i \frac{\pi}{4} \sigma_\varphi}
\end{aligned} \tag{10.13}$$

we are able to introduce the unitary as two displacement operators dependent on the spin matrix σ_φ . This can be rewritten for some combination of the Pauli matrices $\bar{\sigma}$ as

$$U = \frac{\hat{D}(\alpha)}{\sqrt{2}} e^{i \frac{\pi}{4} \hat{n} \cdot \bar{\sigma}} + \frac{\hat{D}(-\alpha)}{\sqrt{2}} e^{-i \frac{\pi}{4} \hat{n} \cdot \bar{\sigma}}. \tag{10.14}$$

Using Eqn. 10.11, the spin dependence is of the form σ_y for a single Ca^+ ion initially prepared in the $|\downarrow\rangle = \frac{1}{\sqrt{2}}(|+_y\rangle - |-_y\rangle)$, which results in

$$U_{cat} |\downarrow\rangle = \frac{1}{i\sqrt{2}} (\hat{D}(\alpha) |+_y\rangle - \hat{D}(-\alpha) |-_y\rangle) \tag{10.15}$$

To perform our mass spec, the displacement can be generated by either a two-tone motional Raman scan or by a tickle. In either case, the math will be identical; two-tone motional Raman was chosen for ease of implementation as a new ARTIQ experiment, and because we gotta rep (cite) our motional Raman technique, whereas it was later replaced with a tickle, since the high voltages started to generate strange lineshapes without implementing the CAT pulses. To describe the displacement generated by the two-tone motional Raman excitation, γ , we have

$$\gamma = \frac{g}{\delta} e^{-i\varphi} (e^{i\delta t} - e^{i\delta t_0}), \tag{10.16}$$

so that the total pulse sequence can be visualized most simply as $\hat{D}(-\alpha) \hat{D}(\gamma) \hat{D}(\alpha) =$

$e^{\gamma\alpha^* - \gamma^*\alpha} \hat{D}(\gamma)$, where $\Theta \equiv \frac{\gamma\alpha^\dagger - \gamma^\dagger\alpha}{i}$ is the phase that is picked up by the CAT sandwich,

$$\begin{aligned}\Theta &= \frac{g}{i\delta} e^{-i\varphi} (e^{i\delta t} - e^{i\delta t_0}) \left(-i\eta \frac{\Omega t}{2}\right) - \frac{g}{\delta} e^{i\varphi} (e^{-i\delta t} - e^{-i\delta t_0}) \left(i\eta \frac{\Omega t}{2}\right) \\ &= -2 \frac{g}{i\delta} \alpha (\cos(\delta t - \varphi) - \cos(\varphi) - \cos(\delta t_0 - \varphi) + \cos(\varphi))\end{aligned}\quad (10.17)$$

The terms $\cos(\varphi)$ were included to make the following trig multiplication identity step more transparent.

$$\begin{aligned}\Theta &= 4 \frac{g}{i\delta} \alpha \left(\sin\left(\frac{\delta t}{2}\right) \sin\left(\frac{\delta t}{2} - \varphi\right) - \sin\left(\frac{\delta t_0}{2}\right) \sin\left(\frac{\delta t_0}{2} - \varphi\right) \right) \\ &= 2g|\alpha| \left(t \text{sinc}\left(\frac{\delta t}{2}\right) \sin\left(\frac{\delta t}{2} - \varphi\right) - t_0 \text{sinc}\left(\frac{\delta t_0}{2}\right) \sin\left(\frac{\delta t_0}{2} - \varphi\right) \right)\end{aligned}\quad (10.18)$$

where Θ is the geometric phase (Berry phase) of this operation. This is related to the area enclosed by the two paths taken, $\mathcal{A}$, by the $|+_y\rangle$ and $|-_y\rangle$ by $\Theta = -2\mathcal{A}$ [CSS87]. The final spin state after the entire operation is given by $|\psi_f\rangle = \frac{1}{i\sqrt{2}}(e^{i\Theta} |+_y\rangle - e^{-i\Theta} |-_y\rangle)$. The value of i in the denominator is a global phase associated with splitting $|\downarrow\rangle$ into the eigenvectors of the σ_y operation. Recall that the motional state will be given by the RF induced displacement $\hat{D}(\gamma) |\downarrow\rangle = |\gamma\rangle$.

10.1.2 Results: Ca^+

The initial calibrations for this scheme are performed using a single Ca^+ ion. In order to perform this experiment, a displacement is generated using motional Raman at $2\pi \cdot 50\text{MHz}$, where we apply a dipole tone at $2\pi \cdot 50\text{MHz}$ and a quadrupole tone at the axial blue sideband. In this case, we had $\omega_0 = 2\pi \cdot 702\text{kHz}$. Prior to applying the CAT amplification pulses, we perform RAP readout of the ion to ensure we know the strength of the displacement given by γ . Additionally, the CAT/Anti-CAT and CAT/displacement phases are calibrated using a motional Ramsey. In the former, we produced a CAT state, then waited 100us (the typical displacement time) before applying the Anti-CAT pulse. In the latter, the motional Raman displacement is applied first, followed by a laser-based pulse of comparable displacement strength. A laser-based displacement pulse differs from a CAT pulse only in that the spin is prepared in the $|+_y\rangle$ state first, which creates only a single displacement. An optional quench 854nm pulse can be used to prepare the spin state back to the $S_{\frac{1}{2}} \Leftrightarrow P_{\frac{1}{2}}$ manifold, i.e. the $|\downarrow\rangle$ state. After the displacement's phase is determined

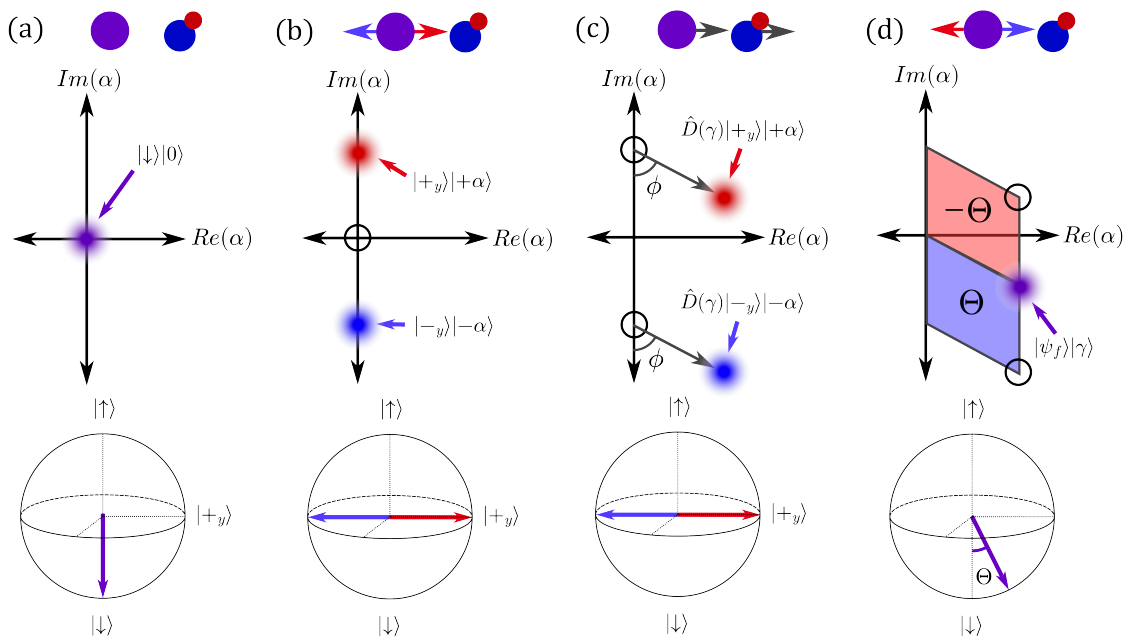


Figure 10.1: In the simplest implementation, a spin-motional CAT uses a single Ca^+ . The internal state of the atom is prepared in $|\downarrow\rangle$ after sideband cooling the motional modes to the vacuum state. Since $|\downarrow\rangle$ is composed of $|\pm_y\rangle$, when a bichromatic pulse is applied, the initial wavepacket will split in two, via a spin-motional CAT operation. A displacement is applied that affects both wavepackets equally, with a phase ϕ relative to the CAT displacement. A final Anti-CAT pulse will recombine the wavepackets, where each path has now accumulated a geometric phase given by Θ , which is read out using the fluorescence of the ion.

relative to the CAT phase, it is shifted by $\frac{\pi}{2}$ so that it will be orthogonal to the CAT/Anti-CAT pulses.

Readout through laser-induced fluorescence determines what population was in the $|S_{\frac{1}{2}}\rangle = |\downarrow\rangle$ or in the $|D_{\frac{5}{2}}\rangle = |\uparrow\rangle$. The probabilities are given by

$$P_{\downarrow} = \langle\psi_f|\downarrow\rangle\langle\downarrow|\psi_f\rangle = |\langle\downarrow|\psi_f\rangle|^2 = \cos^2(\Theta) \quad (10.19)$$

$$P_{\uparrow} = \langle\psi_f|\uparrow\rangle\langle\uparrow|\psi_f\rangle = |\langle\uparrow|\psi_f\rangle|^2 = \sin^2(\Theta) \quad (10.20)$$

for each state. Since there are only two possible outcomes for fluorescence, the system is given by a binomial distribution and the Fisher information of the geometric phase is

$$\mathbb{F}_{\Theta} = \frac{1}{P_{\downarrow}(1 - P_{\downarrow})} \left(\frac{\partial P_{\downarrow}}{\partial \Theta} \right)^2. \quad (10.21)$$

For mass spectroscopy, the relevant quantity is the detuning and hence the Fisher information will be

$$\mathbb{F}_{\delta} = \frac{1}{P_{\downarrow}(1 - P_{\downarrow})} \left(\frac{\partial P_{\downarrow}}{\partial \delta} \right)^2 = \mathbb{F}_{\Theta} \left(\frac{\partial \Theta}{\partial \delta} \right)^2. \quad (10.22)$$

So far, decoherence has been ignored. In the fits, decoherence is introduced as a loss of contrast, which is easiest to include by using the double angle identity and writing the populations as

$$P_{\downarrow} = \frac{1}{2} + \frac{1 - \varepsilon}{2} \cos(2\Theta) \quad (10.23)$$

$$P_{\uparrow} = 1 - P_{\downarrow} = \frac{1}{2} - \frac{1 - \varepsilon}{2} \cos(2\Theta). \quad (10.24)$$

10.1.3 Results: One Bright and One Dark

When a dark ion is loaded into a chain with the single Ca^+ , the additional mass will change the secular frequencies. Provided that an initial calibration was performed on the single Ca^+ prior to loading, numerically solving for the energy potentials described in section 2.1.2 allows us to determine the mass of an unknown ion based on the secular frequencies. Measuring the unknown particle's mass through this method faces complications related to both loading and ion orientation. The loading process for our dark ion of choice H^{35}Cl^+ , as mentioned in section 5.2.2, involves a fair amount

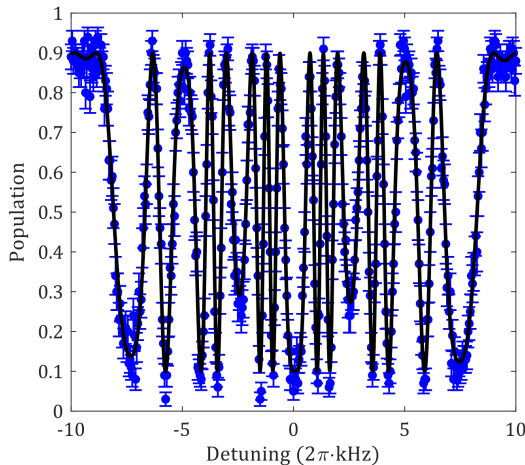


Figure 10.2: Probability of measuring a single Ca^+ in the $S_{1/2}$ state after the CAT interferometer sequence. The blue data points were taken using $100\mu\text{s}$ on both the CAT pulse and a displacement pulse. In this test the displacement is generated using motional Raman, but all future displacements were done with a tickle instead, since it requires less power. The black curve is the fit using equations 10.23 and 10.17. Without the CAT pulses, the displacement produces $\bar{n}_\gamma = 0.55$. Based on the fit, the CAT displacement was $\alpha \approx 3.2$.

of 243nm light to be sent through our trap electrodes. Scattering off the electrodes is unavoidable and the UV light is easily able to charge the electrodes, resulting in stray fields that can shift the secular frequencies of the ion chain. Furthermore, the electrodes will slowly discharge some of the charges that accumulate, which means that there is additional time dependence on the stray fields. Ion orientation also poses a challenge to implementing this method. Due to trap anharmonicities, there are slight changes in the secular frequencies depending on where the chain is {bright, dark} or {dark, bright}, which can be on the order of $\sim 1\text{kHz}$ or $\sim 10^{-3}$ in terms of frequency resolution. In order to overcome these limitations, a scheme described in [DWZ00] was implemented which uses an additional bright ion to form an ion balance. A similar scheme that does not require the second calcium ion can be done using the ROCKING mode, but as will be discussed, the second bright ion will improve our sensitivity in a couple different ways.

10.2 Ion Balance

However, for practical considerations, this method is challenging to implement due to the loading procedure we have for the analyte ion. As will be described, the dark ion loading involves shining pulsed UV light through our trap and leaking gas into our chamber. This procedure will change our ion's position within the trap, indicating that the secular frequencies are drifting during the loading of the molecule.

To overcome this limitation, a different approach was chosen, leveraging the symmetry of a three ion chain with the analyte ion between two logic ions. The dynamics for this approach have been investigated before for quantum logic applications [DWZ00] with the results of the mode eigenfrequencies being

$$\beta_1 = \sqrt{\frac{13}{10} + \frac{1}{10\mu}(21 - \sqrt{441 - 34\mu + 169\mu^2})} \quad (10.25)$$

$$\beta_2 = \sqrt{3} \quad (10.26)$$

$$\beta_3 = \sqrt{\frac{13}{10} + \frac{1}{10\mu}(21 + \sqrt{441 - 34\mu + 169\mu^2})} \quad (10.27)$$

for the COM, ROCKING, and Zig-Zag modes, respectively, each normalized to the axial secular frequency of a single logic ion. The eigenvectors of these modes are then given by

$$\bar{v}_1 = N_1(1, \frac{\sqrt{\mu}}{8}(13 - 5\beta_1^2), 1) \quad (10.28)$$

$$\bar{v}_2 = N_2(1, 0, -1) \quad (10.29)$$

$$\bar{v}_3 = N_3(1, \frac{\sqrt{\mu}}{8}(13 - 5\beta_3^2), 1) \quad (10.30)$$

with N_1, N_2 , and N_3 being the normalization values of each mode. By taking a measurement of two of these modes, the ratio is used to give us the ratio of the masses, μ . We specifically choose the COM and Zigzag modes for the ratio. The ROCKING mode is not chosen because when we drive the CAT displacement, the motion of each logic ion is going in opposite directions to one another, making it significantly more difficult to drive. An additional benefit to using the axial COM and Zig-Zag modes is that they are the smallest and largest frequencies of our chain, which limits coupling to the other modes.

Utilizing the normalized eigenfrequencies, the mass ratio can be analytically writ-

ten as

$$\mu = \frac{m_{\text{analyte}}}{m_{\text{logic}}} = \left(-\frac{256}{338} + \frac{145}{338} \left(\left(\frac{\omega_1}{\omega_2} \right)^2 + \left(\frac{\omega_2}{\omega_1} \right)^2 \right) \right) \mp \sqrt{-\frac{298116}{338^2} + \left(-\frac{256}{338} + \frac{145}{338} \left(\left(\frac{\omega_1}{\omega_2} \right)^2 + \left(\frac{\omega_2}{\omega_1} \right)^2 \right) \right)^2}, \quad (10.31)$$

where the positive term of the quadratic formula will be dropped based on the other measured mode frequencies indicating the analyte mass is close to 36, not 116. Determine the uncertainty of the mass from error propagation results in a long but simple analytic form. A simplified form can be written as

$$\left(\frac{\delta_m}{m} \right)^2 = f(m_0, \omega_1, \omega_2) \left(\left(\frac{\delta_1}{\omega_1} \right)^2 + \left(\frac{\delta_2}{\omega_2} \right)^2 \right) \quad (10.32)$$

where m is the mass of the analyte ion, m_0 is the mass of the logic ion, and $f(m_0, \omega_1, \omega_2)$ is an analytical function dependent on the mass of the logic ion and the two measured frequencies. Because we use lasers to address transitions within the logic ion, we are certain that it is Ca^+ , allowing m_0 to be a known value.

To get an overall improvement in the precision of the mass, the precision of each mode needs to be measured below the SQL. However, even a measurement of a single mode beyond the SQL can result in a reduction in the mass uncertainty. Specifically, the COM mode will have a dominant effect, owing from the fact that it is roughly $2/5^{\text{th}}$ of the Zig-Zag mode.

10.2.1 Theory

Previous discussions related to the Fisher information have been limited to binomial statistics due to the single Ca^+ ion that is used for readout. In order to implement the ion balance, a total of three ions are required, with the outer two in the chain being of the same mass. While in principle, the outer two ions could be dark ions, this orientation does not make practical sense; using two Ca^+ ions and a single dark ion is the easier to configure and has additional benefits. There are twice as many coolant ions for cooling the motional modes of the chain and the mass of these ions is certainly $^{40}\text{Ca}^+$ since the atomic transitions are highly selective, even for isotopes [DS04]. Introduction of the second Ca^+ ion also results in additional

complexity for readout since there are now three possible results for state readout: both dark, one bright/one dark, and both bright, with populations given by $P_{\uparrow\uparrow}$, $P_{\uparrow\downarrow}$, and $P_{\downarrow\downarrow}$ respectively. Fig. 10.3 illustrates how the second Ca^+ will contribute to the geometric phase, due to how each component of the wavepacket interacts with the bichromatic light. If we start by considering the Hamiltonian for a two Ca^+ ion chain undergoing Rabi flopping we can immediately see how the dynamics of the system may change. In the simple case of carrier Rabi flopping, there is no spin motional coupling but the ions will have different laser intensities and, as such, will have different Rabi rates, resulting in a beat note. If we instead drive the system to measure Rabi flopping on the blue sideband, we introduce spin-motional coupling on both ions described by the following Hamiltonian

$$\frac{H_{BSB}}{\hbar} = \sum_{i=0}^N \frac{\Omega\eta}{2i} (\sigma_-^{(i)} \hat{a} - \sigma_+^{(i)} \hat{a}^\dagger) \quad (10.33)$$

which we can get from setting $\Omega_r = 0$ and $\omega_b = \Delta + \omega_0$ in Eqn. 10.5. The resulting time evolution applied to $|\downarrow\downarrow, n\rangle$ is

$$\begin{aligned} U |\downarrow\downarrow, n\rangle &= \left(1 - \frac{n+1}{2(n+1)+1} \left(1 - \cos\left(\sqrt{2(2(n+1)+1)} \frac{\Omega\eta}{2} t\right)\right)\right) |\downarrow\downarrow, n\rangle \\ &+ \sqrt{\frac{n+1}{2(n+1)+1}} \sin\left(\sqrt{2(2(n+1)+1)} \frac{\Omega\eta}{2} t\right) \left(|\uparrow\downarrow, n+1\rangle + |\downarrow\uparrow, n+1\rangle\right) \\ &+ \frac{\sqrt{(n+1)(n+2)}}{2(n+1)+1} \left(1 - \cos\left(\sqrt{2(2(n+1)+1)} \frac{\Omega\eta}{2} t\right)\right) |\uparrow\uparrow, n+2\rangle. \end{aligned} \quad (10.34)$$

The Hamiltonian given by Eqn. 10.10 for an $N = 2$ ion chain is given by

$$\frac{H_I}{\hbar} = \sum_{i=0}^{N=2} \frac{\Omega\eta}{2} \sigma_y^{(i)} (\hat{a}^\dagger + \hat{a}) = \frac{\Omega\eta}{2} (\sigma_y^{(1)} + \sigma_y^{(2)}) (\hat{a}^\dagger + \hat{a}), \quad (10.35)$$

the unitary of which would be analyzed using a similar process as Eqn. 10.13 resulting in

$$U = e^{2i|\gamma|\alpha(\sigma_y^{(1)} + \sigma_y^{(2)})} D(\gamma) \quad (10.36)$$

where $\alpha = \eta \frac{\Omega t}{2}$. Assuming the initial state is $|\downarrow\downarrow\rangle$, the state at some time later is given by

$$U |\downarrow\downarrow, 0\rangle = \cos^2(\Theta) |\downarrow\downarrow, \gamma\rangle + 2 \sin(\Theta) \cos(\Theta) \frac{|\uparrow\downarrow, \gamma\rangle + |\downarrow\uparrow, \gamma\rangle}{\sqrt{2}} + \sin^2(\Theta) |\uparrow\uparrow, \gamma\rangle. \quad (10.37)$$

To determine the effects of decoherence, we must first consider what the density matrix is for $|\psi_f\rangle$. The density matrix of a pure state is given by

$$\rho_p = U |\downarrow\downarrow\rangle \langle\downarrow\downarrow| U^\dagger = \rho_m + \sum_{i \neq j} c_i c_j^* |\varphi_i\rangle \langle\varphi_j|, \quad (10.38)$$

with the density matrix of the mixed state given by

$$\rho_m = \sum_j |c_j|^2 |\varphi_j\rangle \langle\varphi_j| = |c_0|^2 |\downarrow\downarrow\rangle \langle\downarrow\downarrow| + |c_1|^2 |\uparrow\uparrow\rangle \langle\uparrow\uparrow| + |c_2|^2 \frac{|\uparrow\downarrow\rangle \langle\uparrow\downarrow| + |\downarrow\uparrow\rangle \langle\downarrow\uparrow|}{\sqrt{2}}. \quad (10.39)$$

As a pure state decoheres, it approaches the mixed state. In the case we are looking at with two bright ions

$$\rho_m = \cos^4(\Theta) |\downarrow\downarrow\rangle \langle\downarrow\downarrow| + \sin^4(\Theta) |\uparrow\uparrow\rangle \langle\uparrow\uparrow| + 2 \sin^2(\Theta) \cos^2(\Theta) \frac{|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle}{\sqrt{2}} \frac{\langle\uparrow\downarrow| + \langle\downarrow\uparrow|}{\sqrt{2}}. \quad (10.40)$$

When $\Theta = \frac{\pi}{4}$, the resulting mixed state is what we'd expect for equal chance of each state

$$\rho_m = \frac{1}{4} |\downarrow\downarrow\rangle \langle\downarrow\downarrow| + \frac{1}{4} |\uparrow\uparrow\rangle \langle\uparrow\uparrow| + \frac{1}{2} \frac{|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle}{\sqrt{2}} \frac{\langle\uparrow\downarrow| + \langle\downarrow\uparrow|}{\sqrt{2}}. \quad (10.41)$$

When we read out the bright/bright population it will be 0.25, as will dark/dark. The probability of a single bright ion will be 0.5. Similarly to the single Ca^+ case, we will use the double angle trig identity to introduce the loss of contrast term ε .

$$P_{\downarrow\downarrow} = \left(\frac{1}{2} + \frac{1-\varepsilon}{2} \cos(2\Theta) \right)^2 \quad (10.42)$$

$$P_{\uparrow\uparrow} = \left(\frac{1}{2} - \frac{1-\varepsilon}{2} \cos(2\Theta) \right)^2 \quad (10.43)$$

$$P_{\uparrow\downarrow} = 1 - P_{\uparrow\uparrow} - P_{\downarrow\downarrow} = 2 \left(\frac{1}{2} + \frac{1-\varepsilon}{2} \cos(2\Theta) \right) \left(\frac{1}{2} - \frac{1-\varepsilon}{2} \cos(2\Theta) \right). \quad (10.44)$$

When decoherence is negligible $\varepsilon \rightarrow 0$, the state populations are given by

$$P_{\downarrow\downarrow} = \cos^4(\Theta) \quad (10.45)$$

$$P_{\uparrow\uparrow} = \sin^4(\Theta) \quad (10.46)$$

$$P_{\uparrow\downarrow} = 2 \sin^2(\Theta) \cos^2(\Theta). \quad (10.47)$$

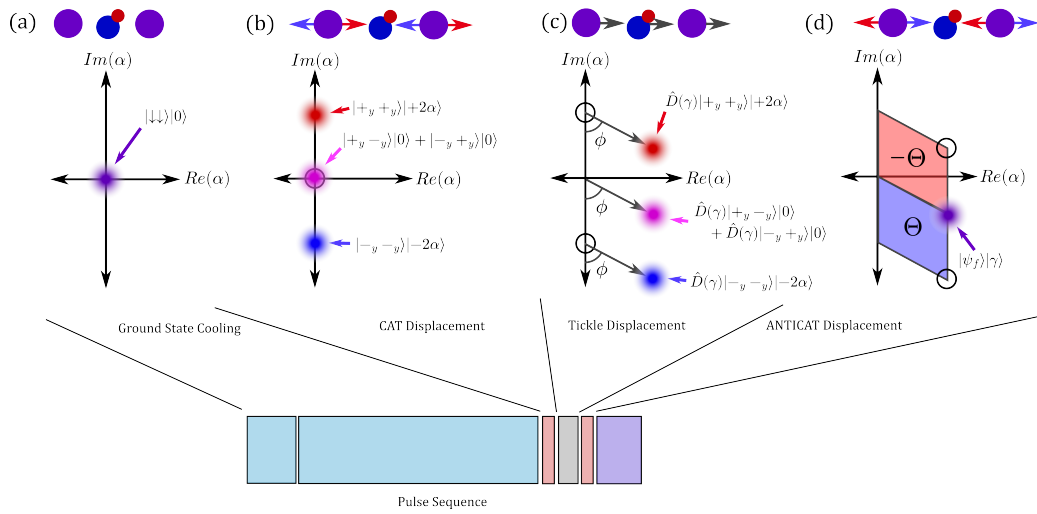


Figure 10.3: Illustration of the CAT interferometer ion balance. The state is initially prepared in $|\downarrow\downarrow\rangle$, which can be decomposed into its σ_y -basis terms, to better understand what parts of the wavepackets get displaced by the CAT operations. Half of the state is made up of $|+y-y\rangle + |-y+y\rangle$ which will not undergo any displacement, whereas the states $|\pm_y\pm_y\rangle$ get displaced by $\pm 2\alpha$ since each atom added $\pm\alpha$ to the displacement. When recombined, only the states that were displaced by the CAT pulse contribute to the geometric phase.

10.2.2 Trinomial Statistics

The Fisher information of a parameter x is given by

$$\mathbb{F}_x = \sum_k \frac{1}{P_k} \left(\frac{\partial P_k(x)}{\partial x} \right)^2 \quad (10.48)$$

for some observable random variable k , with probability P_k that is dependent on the parameter x . In this case, the parameter x could be Θ or a parameter that makes up Θ such as δ , and the observable variable k is the spin state of the system i.e. $k = \uparrow\uparrow, \uparrow\downarrow, \downarrow\downarrow$ for the three different outputs. Noticeably, due to the presence of the second Ca^+ ion, we have trinomial statistics instead of binomial. As a result, we are able to take the three different probability lineshapes to produce more Fisher information than if only one was used, as in the binomial case. Using the probabilities given in Eqn. 10.47, the Fisher information for Θ is

$$\mathbb{F}_\Theta = \frac{1}{P_{\uparrow\uparrow}} \left(\frac{\partial P_{\uparrow\uparrow}}{\partial \Theta} \right)^2 + \frac{1}{P_{\uparrow\downarrow}} \left(\frac{\partial P_{\uparrow\downarrow}}{\partial \Theta} \right)^2 + \frac{1}{P_{\downarrow\downarrow}} \left(\frac{\partial P_{\downarrow\downarrow}}{\partial \Theta} \right)^2 = 8, \quad (10.49)$$

which means that the same Fisher information is achievable regardless of what geometric phase is used. However, this is not the case if we choose the Fisher parameter to be the detuning, which instead has

$$\mathbb{F}_\delta = \mathbb{F}_\Theta \left(\frac{\partial \Theta}{\partial \delta} \right)^2 \quad (10.50)$$

as we've done before. The uncertainty of Θ will follow the SQL $\frac{1}{\sqrt{N}}$ behavior, where N is the number of measurements,

$$\sigma_\Theta = \frac{1}{\sqrt{N} \mathbb{F}_\Theta}. \quad (10.51)$$

10.2.3 Results and Analysis

Mass calculations are dependent on highly sensitive measurements of the secular frequency of two different modes. Each mode frequency stems from an Allan deviation measurement from over 40,000 datapoints, taken on the order of 10-15 minutes. Data collection via Allan deviations results in a high precision measurement of the secular frequency of a given mode. Prior to performing the Allan deviation, an initial linescan is performed in order to determine fitting parameters and to determine what detunings will have the greatest Fisher information. Each population is fitted with

equations 10.44 and 10.18, for the free variables of $|\alpha|$, t , t_0 , δ , φ , and ε . Following this linescan, thousands of data points are taken using the two detunings determined by the linescan. In order to analyze the results of our Allan deviation measurement, we use the Maximum Likelihood Estimator (MLE) method to determine the detuning from our linescan fit.

Given that there are three possible outcomes for fluorescence, the likelihood function for a given parameter x is

$$L(N_{\uparrow\uparrow}, N_{\uparrow\downarrow}, N_{\downarrow\downarrow}, x) = \frac{N!}{N_{\uparrow\uparrow}!N_{\uparrow\downarrow}!N_{\downarrow\downarrow}!} P_{\uparrow\uparrow}(x)^{N_{\uparrow\uparrow}} P_{\uparrow\downarrow}(x)^{N_{\uparrow\downarrow}} P_{\downarrow\downarrow}(x)^{N_{\downarrow\downarrow}}. \quad (10.52)$$

Maximizing the log-likelihood function gives

$$\ln L = C + N_{\uparrow\uparrow} \ln P_{\uparrow\uparrow}(x) + N_{\uparrow\downarrow} \ln P_{\uparrow\downarrow}(x) + N_{\downarrow\downarrow} \ln P_{\downarrow\downarrow}(x) \quad (10.53)$$

where C is some constant that will disappear once we solve for the maximum via

$$\left. \frac{\partial \ln L}{\partial x} \right|_{x=x_e} = 0 \quad (10.54)$$

$$\Rightarrow \frac{N_{\uparrow\uparrow}}{P_{\uparrow\uparrow}(x)} \frac{\partial P_{\uparrow\uparrow}(x)}{\partial x} + \frac{N_{\uparrow\downarrow}}{P_{\uparrow\downarrow}(x)} \frac{\partial P_{\uparrow\downarrow}(x)}{\partial x} + \frac{N_{\downarrow\downarrow}}{P_{\downarrow\downarrow}(x)} \frac{\partial P_{\downarrow\downarrow}(x)}{\partial x} \bigg|_{x=x_e} = 0. \quad (10.55)$$

The value x_e is the MLE for the parameter x , which results in an observed Fisher information given by

$$\mathbb{F}_{x_e} = - \left. \frac{\partial^2 \ln L}{\partial x^2} \right|_{x=x_e}. \quad (10.56)$$

In our case, working with the geometric phase as our parameter, results in a MLE Θ_e which can then be converted into a detuning. To start we use Eqn. 10.44 to define the probabilities, resulting in

$$\Theta_e = \frac{1}{2} \arccos \left(\frac{1}{1 - \varepsilon} \frac{N_{\downarrow\downarrow} N_{\uparrow\uparrow}}{N_{\downarrow\downarrow} + N_{\uparrow\uparrow} + N_{\uparrow\downarrow}} \right), \quad (10.57)$$

which can be converted to δ_e using Eqn. 10.18. Since the solution is many valued, we need to limit the analysis to a small range around the expected detuning from the fit. In practice this procedure is a reasonable approach since the drift in the system is small over the period of data collection; when there is a large drift, this procedure fails to be appropriate.

Prior to introducing the dark ion into the ion trap, a single Ca^+ is used to characterize the system, ensuring that the single ion secular frequencies are measured

for initial mass estimation. Since various types of dark ions can be loaded during the HCl^+ loading sequence, working with a single Ca^+ enables a larger variation between the single ion COM modes and the chain COM modes. Additionally, by limiting the chain to only two ions, reduces detection of other modes that may complicate dark ion mass estimation. Provided that the dark ion mass appears to be mass 36amu or mass 37amu (recall that mass 37amu will undergo photodissociation to mass 36amu from the cooling light), a second Ca^+ is introduced into the chain via the standard Ca^+ loading procedure in section 5.2.2. Once the second bright ion is loaded, the dark ion may not be oriented in the center of the chain, which is a requirement for the ion balance. The most reliable method we've tried so far, has been to blue detune the cooling laser until the chain starts to melt, using low enough power that the ions are not ejected from the trap. While the blue-detuned light will melt the crystal, introducing far red-detuned light by opening the aperture on the double pass AOM setup will recrystallize the chain. This procedure is repeated until the dark ion is in the center of the chain; reorienting the chain typically takes on the order of $\sim 30\text{s}$. Once the dark ion is in the center, it will typically keep that orientation for on the order of ~ 1 hour, dependent on collisions with background gas. Since a crystal has more energy in it than a single ion, the chain melting is typically much more common than the single ion crystal melting. Additionally, we have seen that the laser power for laser-induced fluorescence may need to be reduced from the single ion case.

As mentioned earlier, in order to cool to the ground motional state, we cool all modes in the chain. The modes of interest for the ion balance procedure are the axial COM mode and the axial Zig-Zag mode. For our typical trapping parameters, a single Ca^+ will have $\{1600\text{kHz}, 1300\text{kHz}, 710\text{kHz}\}$ for the $\{\text{EGGs}, \text{RF}, \text{AXIAL}\}$ mode, respectively. With a three ion chain and the dark ion in the center, the axial COM is around 721kHz and the axial Zig-Zag is around 1770kHz. Due to different mode parameters and secular frequencies, various values need to be calculated for each mode separately. The first step is to determine the strength of the electronically induced displacement γ . For this data set, this displacement is generated using a resonant tickle applied onto one of the trapping endcap electrodes. A custom bias tee was built for us by Peter so that the endcap voltage did not need to be reduced; the highest DC voltage we could find in an off-the-shelf model was 100V, and we

apply 200-300 volts typically on the endcaps. The overall displacement strength will be determined by the sum of the mode parameters of each ion in the chain: the COM mode will be the largest since all mode parameters are positive, whereas the other modes will be dramatically reduced. For example, with a dark ion of mass 36amu, we need to apply nearly $\times 60$ more voltage on the Zig-Zag mode than on the COM mode. In principle, the ROCKING mode will never be displaced in this way since only the calcium ions will move, but in opposite directions, and the Zig-Zag mode will get more difficult to measure as the ions become more similar in mass. To readout the strength of the tickle, we use the RAP method, which is used to convert $|\downarrow\downarrow\rangle|\alpha\rangle$ into $|\downarrow\downarrow\rangle|0\rangle$ via an operation that is essentially $\mathbb{I} \otimes \mathbb{I} \otimes |n=0\rangle\langle n=0| + (\sigma_+^{(1)} + \sigma_+^{(2)})\hat{a} + \sigma_+^{(1)}\sigma_+^{(2)}\hat{a}^2$. Only the first term in this sum will be relevant for our readout detection, which is determined with basic thresholding of states.

An additional consideration for the three ion chain, is that the secular frequencies for the COM mode and the Zig-Zag mode are around 1 MHz separated and will have a different mode parameter b_i based on the normalization factor in Eqn. 10.30. Both result in a smaller Lamb-Dicke parameter for the Zig-zag mode compared to the COM mode. The frequency difference further reduces the overall CAT strength since the bichromatic light is generated via a single pass AOM, experiencing a frequency dependence in the light transmitting the AOM, shown in Fig. 10.4. To balance the two tones, the AOM is centered at the carrier by detecting power on a photodiode, and fine adjustment via the power of the RF signal going into the AOM.

In order to perform the mass comparison, a frequency linescan is performed immediately prior to a two point measurement used for an Allan deviation measurement. Two different conditions were used: a CAT pulse 100 μ s long, and a pulse 200 μ s long, the latter being used in order to try higher CAT strengths on the Zig-Zag mode since, as outlined earlier, this mode needs approximately 16 times more 729nm power to create the same strength displacement as the COM mode. Using the results shown in figures 10.6 and 10.7, we estimate the mass of the dark ion to be 36.0158(5) compared to the actual mass of H^{35}Cl^+ which is 35.97555342. The inaccuracy in this measurement is due to shifts in the normal mode frequencies, which for mixed ion species [Hom13b], typically are associated with systematic drifts, dif-

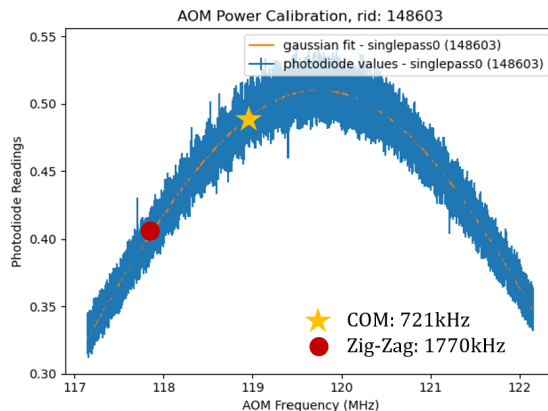


Figure 10.4: Photodiode power measurement of 729nm light transmitted through the single pass AOM used to produce bichromatic light. The peak output of the Gaussian peak appears at 119.74MHz, with the locations of the RSB’s of the COM and Zig-Zag motional modes being depicted with a star and a circle respectively. This accounts for a 15% decrease in power for 729nm light at the Zig-Zag mode compared to the COM mode.

ferential forces on the ions due to a gradient in the pseudopotential dependent on the position within the ion chain, a different radiation force affecting bright ions compared to dark ions, and anharmonicities within the trapping potential. Several datasets were taken during the day for both a $100\mu\text{s}$ CAT and for a $200\mu\text{s}$ CAT pulse duration, with several hours between repeat conditions. Comparing these datasets, we see that the standard deviation of the COM mode was $\pm 17\text{Hz}$ and the Zig-Zag mode was $\pm 30\text{Hz}$, both of which would not correspond to this discrepancy. Instead, we believe that the discrepancy likely comes from residual micromotion similar to the measurements performed in [Jam13], although trap anharmonicities are another potential candidate. Since this trap uses geometries based off of [Gug17], we believe 1V on a compensation electrode will produce about 2.5V/m electric field at the center of our trap. Comparing to [Jam13], the discrepancy would likely come from the Zig-Zag mode frequency and comes from a $\sim 10\text{V/m}$ offset in our shimming, as shown in Fig. 10.5. Measurements of both modes on the previous day indicated about -20Hz in the COM mode and about $+100\text{Hz}$ change on the Zig-Zag, which is on the order of the shifts needed to account for this inaccuracy (albeit in the wrong direction). During the tickle scan there are no lasers illuminating the ions so the

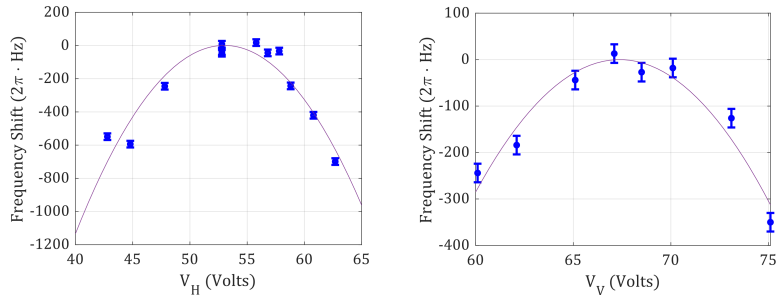


Figure 10.5: To determine whether the inaccuracy is due to being off the micromotion null, we applied voltages onto the compensation electrodes to see if the secular frequency shifted. The left image shows adjusting just the horizontal shimming voltage and the right shows adjusting the vertical shimming voltage, after setting the horizontal voltage to the compensated value. The y-axis is frequency offset relative to some arbitrary frequency center.

radiation pressure forces can be ignored.

In order to determine if the shimming led to the mass inaccuracy, we took measurements of the secular frequency of the Zig-Zag mode at different shimming parameters, shown in Fig. 10.5. The secular frequencies were taken without implementing the CAT steps, and for motional state readout with implemented rapid adiabatic passage. This was a quick option for measuring the secular frequency. The results from this measurement shows that for the measurements shown in figures 10.6 and 10.7 the shimming could have been off by $\sim 5\text{V}$ on our compensation electrodes. Unlike the Zig-Zag mode, when we tried doing the same procedure on the COM mode there was no noticeable change to the secular frequency over 20V. This agrees with the trend shown in [Jam13, Hom13a], that the COM mode is much less sensitive compared to a ROCKING mode. To model this behavior, a stray field component needs to be included in the DC potential term prior to solving for the motional modes. The coupling between the axial and radial dimensions will come from the off-diagonal terms in the Hessian curvature matrix, outlined in [Hom13a].

To compare the precision of this measurement to a classical limit, we plot the Allan deviation result to a classical Ramsey uncertainty. The Ramsey pattern is generated by a pair of displacements with a delay between them, whereby the second displacement is applied to perfectly antidisplace back to the ground state when on

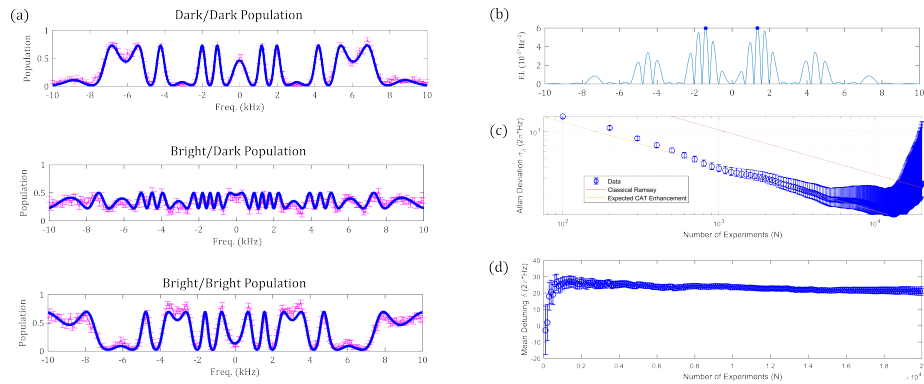


Figure 10.6: COM mode for ion balance to sense the mass of a dark ion. For an unentangled ion chain, the probabilities (a) are given by equations 10.47, with the RAW data shown in pink, and the fits shown in blue. By fitting each of the probabilities, we take a weighted average of the fit parameters to determine the Fisher information (b). To perform an Allan deviation measurement, we take 20000 points at the two blue markers in (b), which represent the highest Fisher information points. Processed Allan deviation is shown in (c), with the detuning from the set point shown in (d). This data is taken with a $100\mu\text{s}$ CAT time, a $100\mu\text{s}$ tickle time, a tickle displacement $\gamma = 1.16\text{s}$, and a CAT displacement of 3.98.

resonance. Residual displacement will occur at different detunings, given by

$$|\alpha| = 2\Omega_d t_F \text{sinc}\left(\frac{\delta t_F}{2}\right) \sin\left(\frac{\delta(T + t_f)}{2}\right) \quad (10.58)$$

as shown in the supplementary information of [WSH19], where t_F is the duration of the tickle pulse, T is the duration of the delay, and $\Omega_d = \frac{eEx_0}{2\hbar}$ is the tickle strength defined earlier in this work. Displacements are provided by a tickle given by γ with tickle time $t_F = t_{cat}$ and the delay is given by $T = t_{tickle}$. In this way, we have kept constant the amount of time each pulse in the sequence is as well as the tickle strength. The Fisher information is then calculated for different detunings

$$\mathbb{F}_\delta = 4 \left(\frac{\partial |\alpha|}{\partial \delta} \right)^2 \quad (10.59)$$

where we have taken the SQL for a displacement measurement $\mathbb{F}_\alpha = 4$, and the classical Ramsey limit corresponds to the detunings where the Fisher information was maximized.

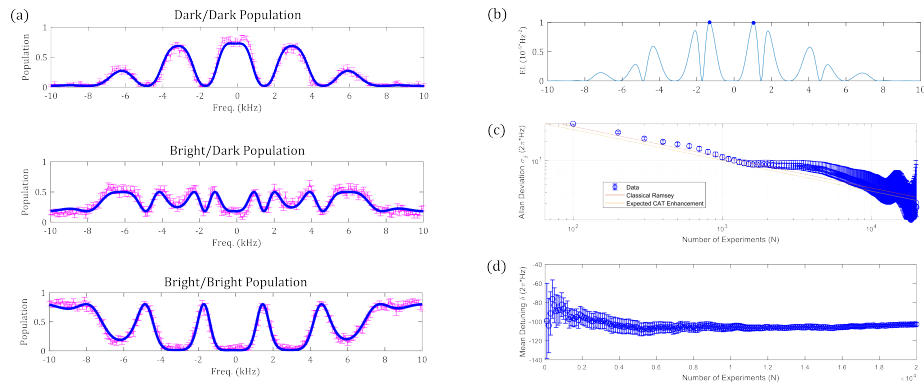


Figure 10.7: Zig-Zag mode for ion balance to sense the mass of a dark ion. For an unentangled ion chain, the probabilities (a) are given by equations 10.47, with the RAW data shown in pink, and the fits shown in blue. By fitting each of the probabilities, we take a weighted average of the fit parameters to determine the Fisher information (b). To perform an Allan deviation measurement, we take 20000 points at the two blue markers in (b), which represent the highest Fisher information points. Processed Allan deviation is shown in (c), with the detuning from the set point shown in (d). This data is taken with a $100\mu\text{s}$ CAT time, a $100\mu\text{s}$ tickle time, a tickle displacement $\gamma = 1.08\text{s}$, and a CAT displacement of 0.56.

10.3 Entangled Ion Balance

Starting in $|\downarrow\downarrow\rangle$ results in three different wavepackets after a bichromatic pulse creates a spin-motional CAT state, evident when the initial state is written in the σ_y basis,

$$|\downarrow\downarrow\rangle|0\rangle = -\frac{1}{2}\left(|+_y+_y\rangle|0\rangle + |-_y-_y\rangle|0\rangle - |+_y-_y\rangle|0\rangle - |-_y+_y\rangle|0\rangle\right) \quad (10.60)$$

as depicted in Fig. 10.3. The final pair of terms in this equation will not enclose any area in phase space during the CAT interferometry sequence since the CAT pulse will not lead to a net displacement. We can think about each ion's displacement from the laser: $|\pm_y\rangle$ will be displaced by $\pm\alpha$, so $|\pm_y\pm_y\rangle$ will be displaced by $\pm 2\alpha$, but both terms in $|\pm_y\mp_y\rangle$ will destructively interfere and lead to no net displacement. By starting in $\frac{1}{\sqrt{2}}\left(|+_y+_y\rangle|0\rangle \pm |-_y-_y\rangle|0\rangle\right)$ we can increase the overall Fisher information by a factor of two by removing any population in the states that do not generate a geometric phase, since the average geometric phase is now doubled. This phase space picture is illustrated in Fig. 10.8.

10.3.1 Theory

Starting in the state $|\downarrow\downarrow\rangle|0\rangle$, we will apply a Mølmer-Sørensen gate [GB09] to produce the state

$$|\psi_0\rangle = e^{i\Omega t} \left(\cos(\Omega t) |\downarrow\downarrow\rangle|0\rangle - ie^{2i\varphi} \sin(\Omega t) |\uparrow\uparrow\rangle|0\rangle \right) \quad (10.61)$$

where Ω is the Rabi rate of the pulse, t is the pulse duration, and φ is the phase of the red sideband and blue sideband laser phase. To get equal population of $|\downarrow\downarrow\rangle$ and $|\uparrow\uparrow\rangle$, the MS pulse needs to have $\Omega t = \frac{\pi}{4}$ such that the state becomes

$$\begin{aligned} |\psi_0\rangle &= \frac{1}{\sqrt{2}} e^{i\varphi} \left(e^{-i(\varphi - \frac{\pi}{4})} |\downarrow\downarrow\rangle|0\rangle + e^{i(\varphi - \frac{\pi}{4})} |\uparrow\uparrow\rangle|0\rangle \right) \\ &= \frac{1}{\sqrt{2}} e^{i\varphi} \left(i \sin\left(\varphi - \frac{\pi}{4}\right) (|+_y+_y\rangle + |-_y-_y\rangle) + \cos\left(\varphi - \frac{\pi}{4}\right) (|+_y-_y\rangle + |-_y+_y\rangle) \right) |0\rangle. \end{aligned} \quad (10.62)$$

To create the desired state, we choose $\varphi = \frac{3\pi}{4}$, so that

$$|\psi_0\rangle = \frac{1}{\sqrt{2}} e^{i\frac{5\pi}{4}} \left(|+_y+_y\rangle + |-_y-_y\rangle \right) |0\rangle. \quad (10.63)$$

where there is some global phase of $\frac{5\pi}{4}$ picked up at the end of the MS pulse. Repeating the time evolution from the CAT interferometer sequence results in the following populations:

$$P_{\downarrow\downarrow} = \frac{1}{2} \cos^2(2\Theta) \quad (10.64)$$

$$P_{\uparrow\downarrow} = \sin^2(2\Theta) \quad (10.65)$$

$$P_{\uparrow\uparrow} = \frac{1}{2} \cos^2(2\Theta) \quad (10.66)$$

where the geometric phase Θ is still given by 10.18.

10.3.2 Results

An initial test of an entangled CAT was performed using two Ca^+ ions. After ground state cooling, they experience an entangling pulse from a pair of bichromatic tones detuned from resonance, approximately $100\mu\text{s}$ long. To determine the fidelity of the operation, the standard [GB09] MS population and parity measurements are performed, with an entanglement fidelity of

$$F = \frac{P_{\downarrow\downarrow} + P_{\uparrow\uparrow} + A}{2} \quad (10.67)$$

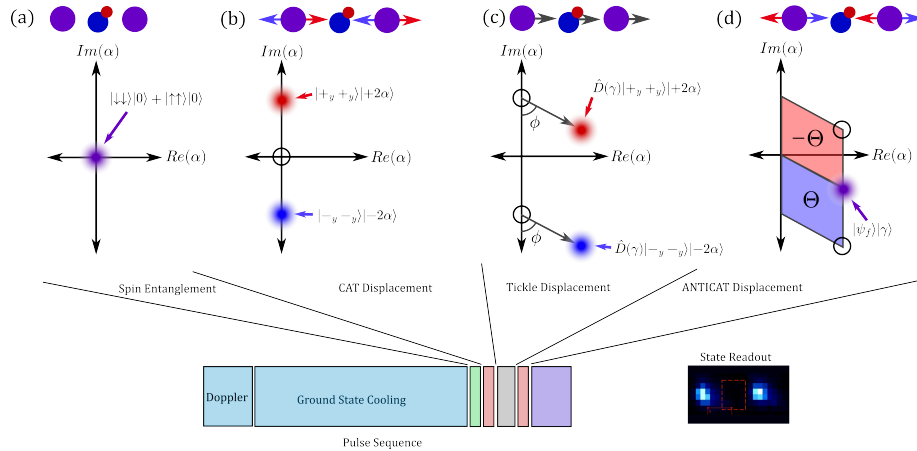


Figure 10.8: An entangled ion balance requires an additional entangling step during state preparation, but otherwise follows the same procedure as the unentangled case. By including the entanglement step, the CAT procedure creates two wavepackets that are displaced by $\pm 2\alpha$, removing the additional population that is not displaced during the unentangled case. This ends up doubling the Fisher information, for greater sensitivity.

where A is the contrast of the parity pulse. After entangling the CAT interferometry sequence continues as before. Data for this test is shown in Fig. 10.10 for the COM mode of a two Ca^+ chain. For a CAT of $\alpha = 1.59$, and tickle of $\gamma = 0.72$, we are able to measure 18.7dB below an equivalent classical Ramsey measurement. The classical Ramsey is uses the same tickle strength as the CAT interferometer tickle, with the tickle time of the CAT pulse, and a delay of the tickle in the CAT interferometer.

We’ve repeated the measurement with a single H^{35}Cl^+ between the two bright ions. The fidelity of this entanglement is noticeably worse, due to the parity pulse. As of writing this thesis, we are in the middle of taking higher fidelity data and repeating entangled measurements with a molecular ion. Similarly, we are investigating the possible sources for the inaccuracy of our mass measurement to improve the system for practical applications in mass spectroscopy.

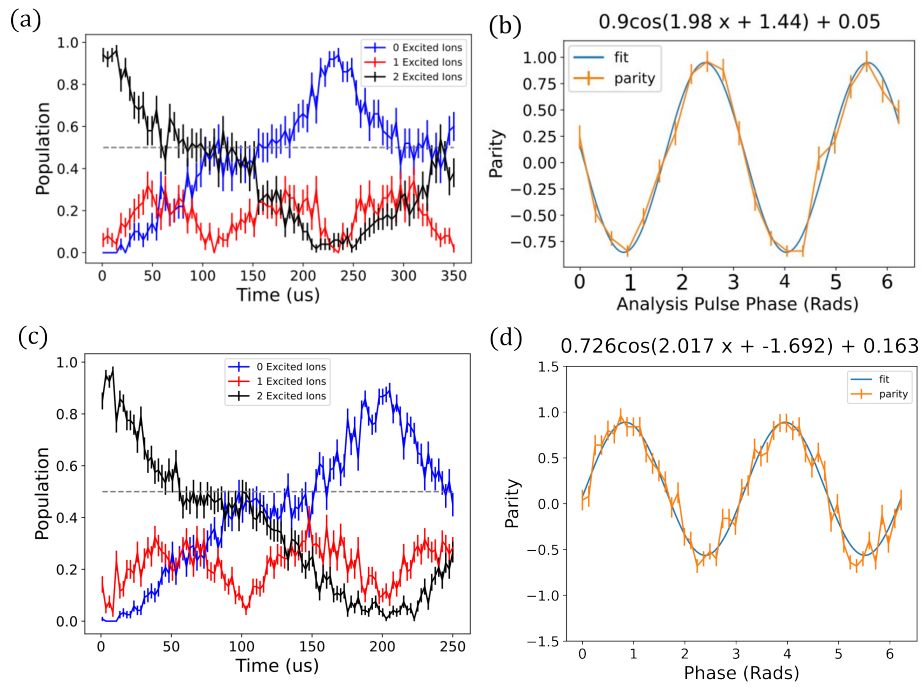


Figure 10.9: Measurement of the entanglement fidelity for two calcium ions (a)/(b) and for a three ion chain (c)/(d) with H^{35}Cl^+ trapped between two Ca^+ ions. The populations of the different bright/dark states are shown in (a)/(c), with the parity pulses shown in (b)/(d). The fidelity of the two Ca^+ is greater than 90%, whereas the fidelity of the three ion chain is greater than 80%.

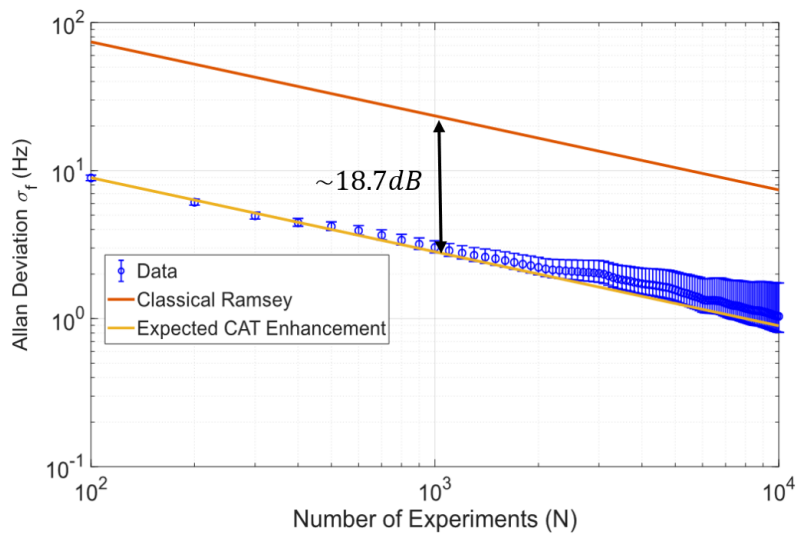


Figure 10.10: As a test of the entanglement step, the Ca^+ were loaded without a dark ion. As shown in Fig. 10.9, the ions are entangled with moderate fidelity prior to the CAT interferometer sequence. A test of the COM frequency sensitivity was performed in this way, which produced a $\sim 18.7\text{dB}$ gain over the classical Ramsey limit. The data was taken using the following parameters $\alpha = 1.59$, $\gamma = 0.72$, $t_{\text{cat}} = 100\mu\text{s}$, and $t_{\text{tickle}} = 50\mu\text{s}$.

CHAPTER 11

Conclusion

Encompassed within this work has been an overview of theory and experimental methods implemented during my graduate research experience towards our goal of achieving molecular quantum logic via electric field gradients. Despite not having definitive confirmation that this scheme has been successfully demonstrated, this experience has been fruitful in establishing new theory and techniques related to the field of trapped ion quantum sensing. A phrase my colleague Clayton came up with that fits our experiment so well is “Those that can’t quantum compute, quantum sense”. While this can be interpreted as a jab, in reality it is meant to remind us that we have made an incredibly sensitive system that is reacting to fields and systematics that we never considered pertinent. Not only that, we then went on to determine and understand the physical phenomena that are at play. For this reason, I am happy that we went down the rabbit holes that were motional Raman, subharmonic sensing, super-resolution, and quantum mass spec. Unfortunately, I would have loved to be the first to demonstrate a transition driven in a molecule using EGGs, but this will need to be left to the future researcher on this experiment. If Josh demonstrates this within the first six months after my leaving lab, I’ll probably cry (mix of joy and sadness).

As this experiment matures, it has the capabilities to accomplish many of the goals set out in [\[HC21\]](#) as well as other proposals that Eric and Wes have come up with, in addition to the various ideas that we already have lined up. Molecular ions, while very challenging to control, offer so many unique avenues and our greater understanding of the dynamics of quantum harmonic oscillators has introduced its own possibilities. I hope to see some of these options explored in the future, and I hope that some of what was included in this thesis will be useful to other graduate students.

And dibs on the trap once it's retired!

APPENDIX A

Wigner-Eckart Formalism for Molecules

Tensors behave differently depending on the reference frame. The typical notation has the reference frame of the molecule denoted by “q” and has the lab reference frame denoted by “p”. Different momenta will be referred to in tensor notation. To switch between reference frames we rely on a rotation operator $D_{pq}^{(k)}(\omega)$, which rotates from ‘p’ to ‘q’ about ω , where $\omega = (\varphi, \vartheta, \chi)$ represents the Euler angles associated with the transformation from the space-fixed axes to the molecule-fixed axes.

$$T_q^k(\mathbf{A}) = \sum_p D_{pq}^{(k)}(\omega) T_p^k(\mathbf{A})$$

and the inverse is

$$T_p^k(\mathbf{A}) = \sum_q D_{pq}^{(k)}(\omega)^* T_q^k(\mathbf{A})$$

In this illustration of the Wigner-Eckart properties I cover the spin term of the Zeeman Hamiltonian for a Hund’s case (a) molecule, such as HCl^+ . The eigenstates of this type of molecule are

$$|\Psi\rangle = |\eta, A; S, \Sigma; J, \Omega; I, F, m_F\rangle. \quad (\text{A.1})$$

For ease, I have written only a single nuclear spin term and hyperfine F. While the notation may differ from other sources, I am writing it as such to illustrate least complex to most complex momentum terms. The Hamiltonian term we are going to look at is

$$\frac{H_{spin}}{\hbar} = g_S \mu_B B_z T_{p=0}^1(\mathbf{S}) + g'_L \mu_B B_z \sum_{q=\pm 1} e^{-2iq\varphi} D_{0,-q}^{(1)}(\omega)^* T_q^1(\mathbf{S}). \quad (\text{A.2})$$

We’ll start by examining the term $\langle \Psi | T_{p=0}^1(\mathbf{S}) | \Psi \rangle$. For analysis, I’m going to introduce the Wigner-Eckart equations relevant to breaking it down, starting with

$$\langle j, m | T_p^k(\mathbf{A}) | j', m' \rangle = (-1)^{j-m} \begin{pmatrix} j & k & j' \\ -m & p & m' \end{pmatrix} \langle j || T^k(\mathbf{A}) || j' \rangle \quad (\text{A.3})$$

To break down a reduced matrix element that is a sum of multiple momentum terms $j_1 + j_2 = j_{12}$, such as $\mathbf{F} = \mathbf{J} + \mathbf{I}$, use the property

$$\langle j_1, j_2, j_{12} | T^{k_1}(\mathbf{A}_1) | j'_1, j'_2, j'_{12} \rangle = \delta_{j_2 j'_2} (-1)^{j'_{12} + j_1 + k_1 + j_2} \sqrt{(2j_{12} + 1)(2j'_{12} + 1)} \begin{Bmatrix} j'_1 & j'_{12} & j_2 \\ j_{12} & j_1 & k_1 \end{Bmatrix} \langle j_1 | T^{k_1}(\mathbf{A}_1) | j'_1 \rangle \quad (\text{A.4})$$

For a Hund's case (a) molecule, the reduced matrix for the rotation term will behave as the following

$$\langle J, \Omega | D_q^{(1)}(\omega)^* | J', \Omega' \rangle = (-1)^{J - \Omega} \begin{pmatrix} J & 1 & J' \\ -\Omega & q & \Omega' \end{pmatrix} \sqrt{(2J + 1)(2J' + 1)} \quad (\text{A.5})$$

The dot in the subscript indicates the matrix element has been reduced as far as possible within the lab reference frame p, but not with the molecule's q. The goal is to be able to remove all reduced matrix elements. The key to this is get to

$$\langle S | T^1(\mathbf{S}) | S' \rangle = \sqrt{(2S + 1)(S + 1)S} \delta_{SS'} \quad (\text{A.6})$$

Now that we've outlined the tools that we need to use to solve the inner product, it is time to solve each of the Zeeman terms in the Hamiltonian

To start, we will look at the spin component of the Zeeman Hamiltonian. Using equation A.3 and the state in A.1, we obtain

$$\begin{aligned} \langle \Psi | T_{p=0}^1(\mathbf{S}) | \Psi' \rangle &= \langle S, \Sigma; \eta, \Lambda; J, \Omega; I; F, m_F | T_{p=0}^1(\mathbf{S}) | S', \Sigma'; \eta', \Lambda'; J', \Omega'; I'; F', m'_F \rangle \\ &= (-1)^{F - m_F} \begin{pmatrix} F & 1 & F' \\ -m_F & 0 & m'_F \end{pmatrix} \langle F | T^1(\mathbf{S}) | F' \rangle \quad (\text{A.7}) \end{aligned}$$

Although the subscript p has been removed, it's important to keep in the back of your mind that this is still in the lab's frame. Next we'll use A.4 on the reduced matrix element that we generated using $\mathbf{F} = \mathbf{J} + \mathbf{I}$

$$\langle F | T^1(\mathbf{S}) | F' \rangle = \delta_{II'} (-1)^{F' + J + 1 + I} \sqrt{(2F + 1)(2F' + 1)} \begin{Bmatrix} J' & F' & I \\ F & J & 1 \end{Bmatrix} \langle J | T^1(\mathbf{S}) | J' \rangle \quad (\text{A.8})$$

Next, we'll swap from the lab frame to the molecular frame.

$$\langle J | T^1(\mathbf{S}) | J' \rangle = \sum_q \langle J | D_q^{(1)}(\omega)^* T_q^1(\mathbf{S}) | J' \rangle = \sum_q \langle J | D_q^{(1)}(\omega)^* | J' \rangle \langle J | T_q^1(\mathbf{S}) | J' \rangle \quad (\text{A.9})$$

There is a little subtlety here that has tripped me up, so I'm including it for clarification.

$$\begin{aligned}\langle J || T^1(\mathbf{S}) || J' \rangle &= \sum_p \langle J || T_p^1(\mathbf{S}) || J' \rangle = \sum_p \sum_q \langle J || D_{pq}^{(1)}(\omega)^* T_q^1(\mathbf{S}) || J' \rangle \\ &= \sum_q \langle J || D_q^{(1)}(\omega)^* T_q^1(\mathbf{S}) || J' \rangle \quad (\text{A.10})\end{aligned}$$

Now, going back to the two reduced matrix elements we are left with

$$\langle J || D_q^{(1)}(\omega)^* || J' \rangle = (-1)^{J-\Omega} \begin{pmatrix} J & 1 & J' \\ -\Omega & q & \Omega' \end{pmatrix} \sqrt{(2J+1)(2J'+1)} \quad (\text{A.11})$$

For the rotation, J is a good quantum number. When we move to the molecule's frame, we can't break $\mathbf{J}$ into $\mathbf{L}$ and $\mathbf{S}$ like we did with $\mathbf{F} = \mathbf{J} + \mathbf{I}$ since $\mathbf{L}$ is not a good quantum number. Instead, since $\mathbf{S}$ is a good quantum number, we can replace $\mathbf{J}$ with it. If we were calculating the orbital momentum component, then we could replace it with $\mathbf{L}$.

$$\begin{aligned}\langle J || T_q^1(\mathbf{S}) || J' \rangle &= (-1)^{S-\Sigma} \begin{pmatrix} S & 1 & S' \\ -\Sigma & q & \Sigma \end{pmatrix} \langle S || T^1(\mathbf{S}) || S' \rangle \\ &= (-1)^{S-\Sigma} \begin{pmatrix} S & 1 & S' \\ -\Sigma & q & \Sigma \end{pmatrix} \sqrt{(2S+1)(S+1)S} \delta_{SS'} \quad (\text{A.12})\end{aligned}$$

By combining [A.7](#), [A.8](#), [A.9](#), [A.11](#), and [A.12](#), we get the final result for the Zeeman matrix elements for the electronic spin Hamiltonian.

Moving on to the rotational component $\mathbf{J}$, it is worth noting that

$$\begin{aligned}& \sum_p (-1)^p \langle J, \Omega, M || T_p^1(\mathbf{J}) D_{-pq}^1(\omega)^* || J', \Omega', M' \rangle \\ &= \sum_p \sum_{J'' \Omega'' M''} (-1)^p \langle J, \Omega, M || T_p^1(\mathbf{J}) || J'', \Omega'', M'' \rangle \langle J'', \Omega'', M'' || D_{-pq}^1(\omega)^* || J', \Omega', M' \rangle,\end{aligned}$$

which will produce nonzero values when $J'' = J$, $\Omega'' = \Omega$, and $M'' = M - p$.

For the orbital Zeeman splitting, we have

$$\begin{aligned}
& \langle \Psi | T_p^1(\mathbf{L}) \cdot T_p^1(\mathbf{B}) | \Psi' \rangle \\
&= \langle S, \Sigma; \eta, \Lambda; J, \Omega; I; F, m_F | B_z T_{p=0}^1(\mathbf{L}) | S', \Sigma'; \eta', \Lambda'; J', \Omega'; I'; F', m'_F \rangle \\
&= \langle S, \Sigma; \eta, \Lambda; J, \Omega; I; F, m_F | B_z \sum_q D_{0q}^{(1)}(\omega)^* T_q^1(\mathbf{L}) | S', \Sigma'; \eta', \Lambda'; J', \Omega'; I'; F', m'_F \rangle \\
&= B_z \sum_q \sum_{\Psi''} \langle \Psi | D_{0q}^{(1)}(\omega)^* | \Psi'' \rangle \langle \Psi'' | T_q^1(\mathbf{L}) | \Psi' \rangle \quad (\text{A.13})
\end{aligned}$$

Following a similar procedure as with the spin, the matrix elements of the orbital tensor product can be decomposed into

$$\begin{aligned}
\langle \Psi'' | T_q^1(\mathbf{L}) | \Psi' \rangle &= (-1)^{F''-m_F''} \begin{pmatrix} F'' & 1 & F' \\ -m_F'' & 0 & m_F' \end{pmatrix} \langle F'' | T^1(\mathbf{L}) | F' \rangle \\
&= (-1)^{F''-m_F''+F'+J''+I''+1} \sqrt{(2F''+1)(2F'+1)} \begin{Bmatrix} J' & F' & I'' \\ F'' & J'' & 1 \end{Bmatrix} \\
&\quad \begin{pmatrix} F'' & 1 & F' \\ -m_F'' & 0 & m_F' \end{pmatrix} \langle J'' | T^1(\mathbf{L}) | J' \rangle \quad (\text{A.14})
\end{aligned}$$

and

$$\begin{aligned}
\langle \Psi | D_{0q}^{(1)}(\omega)^* | \Psi'' \rangle &= (-1)^{J-m_j} \begin{pmatrix} J & 1 & J'' \\ -m_j & 0 & m_j'' \end{pmatrix} \langle J, \Omega | D_{0q}^{(1)}(\omega)^* | J'', \Omega'' \rangle \\
&= (-1)^{J+J-m_j-\Omega} \begin{pmatrix} J & 1 & J'' \\ -m_j & 0 & m_j'' \end{pmatrix} \begin{pmatrix} J & 1 & J'' \\ -\Omega & q & \Omega'' \end{pmatrix} \sqrt{(2J+1)(2J''+1)}. \quad (\text{A.15})
\end{aligned}$$

The remaining term in A.18 can also be seen in

$$\langle J, m_j | T_q^1(\mathbf{L}) | J', m_j' \rangle = (-1)^{J-m_j} \begin{pmatrix} J & 1 & J' \\ -m_j & q & m_j' \end{pmatrix} \langle J | T^1(\mathbf{L}) | J' \rangle \quad (\text{A.16})$$

To substitute in this result, it is instructive to rewrite it as

$$\begin{aligned}
\begin{pmatrix} J & 1 & J' \\ -m_j & 0 & m_j' \end{pmatrix} &= \frac{\langle \eta, \Lambda | T_0^1(L) | \eta', \Lambda' \rangle}{\langle \eta, \Lambda | T_0^1(L) | \eta', \Lambda' \rangle} (-1)^{m_j-J} \\
&= \frac{\Lambda}{\langle \eta, \Lambda | T_0^1(L) | \eta', \Lambda' \rangle} (-1)^{m_j-J}
\end{aligned}$$

where we constrain $q = 0$. Using this constraint and combining the terms gives us the final result

$$\langle \Psi | T_p^1(\mathbf{B}) \cdot T_p^1(\mathbf{L}) | \Psi' \rangle = B_z \Lambda (-1)^{F-m_F+F'+2J+I+1-\Omega} \begin{Bmatrix} J & F & I \\ F' & J & 1 \end{Bmatrix} \begin{pmatrix} F & 1 & F' \\ -m_F & 0 & m_F \end{pmatrix} \begin{pmatrix} J & 1 & J \\ -\Omega & 0 & \Omega \end{pmatrix} \sqrt{(2F+1)(2F'+1)(2J+1)(2J+1)}. \quad (\text{A.17})$$

As we move to the rotational term, it becomes easier to solve,

$$\langle \Psi'' | T_q^1(\mathbf{J}) | \Psi' \rangle = (-1)^{F''-m_F''+F'+J''+I''+1} \sqrt{(2F''+1)(2F'+1)} \begin{Bmatrix} J' & F' & I'' \\ F'' & J'' & 1 \end{Bmatrix} \begin{pmatrix} F'' & 1 & F' \\ -m_F'' & 0 & m_F' \end{pmatrix} \langle J'' | T^1(\mathbf{J}) | J' \rangle \quad (\text{A.18})$$

with the last term being replaced with

$$\langle J | T^1(\mathbf{J}'') | J' \rangle = \sqrt{(2J''+1)(J''+1)J''} \delta_{J''J'} \quad (\text{A.19})$$

The nuclear spins term follow nearly procedure the same as the rotational term, just swapping $\mathbf{J}/J$ for $\mathbf{I}/I$.

APPENDIX B

Magnus Expansion

The Magnus expansion is a method to produce approximate solutions to the time-dependent Schrödinger equation, hence why it is sometimes referred to as the time-dependent exponential perturbation theory [SR09]. Starting from the Schrödinger equation, the time evolution operator $U(t)$ of a time-dependent Hamiltonian $H(t)$ is defined by

$$i\hbar \frac{\partial}{\partial t} U(t) = H(t)U(t). \quad (\text{B.1})$$

The Magnus expansion creates approximate solutions to this equation via a power series expansion given by

$$U(t) = e^{\Omega(t)} \quad (\text{B.2})$$

where $\Omega(t)$ is a sum of integrals. The first several results of $\Omega(t)$ are given by

$$\begin{aligned} \Omega_1(t) &= - \int_{t_0}^t \frac{i}{\hbar} H(t_1) dt_1 \\ \Omega_2(t) &= - \frac{1}{2\hbar^2} \int_{t_0}^t dt_1 \int_{t_0}^{t_1} [H(t_1), H(t_2)] dt_2 \\ \Omega_3(t) &= \frac{i}{6\hbar^3} \int_{t_0}^t dt_1 \int_{t_0}^{t_1} dt_2 \int_{t_0}^{t_2} ([H(t_1), [H(t_2), H(t_3)]] + [H(t_3), [H(t_2), H(t_1)]]) dt_3 \end{aligned} \quad (\text{B.3})$$

which become progressively more complex. Physically interpretation of these terms may not be apparent but it has a profound effect when considering the dynamics of a system. For example, it is typical to limit the sum to the first order term, which produces the form of the time evolution operator most recognized

$$U(t) = e^{-\frac{iHt}{\hbar}}. \quad (\text{B.4})$$

The correct physical interpretation of this term is the first-order effect of the Hamiltonian. Working with the operators of a quantum harmonic oscillator, $\hat{a}$ and $\hat{a}^\dagger$, we can consider a first-order effect as being an interaction with a single phonon. This

limits the dynamics to the absorption or emission of a single phonon. If we consider the second order term in the Magnus Expansion, then these will be viewed as a two-phonon process, the results being the following $\hat{a}_1\hat{a}_2$, $\hat{a}_1\hat{a}_2^\dagger$, $\hat{a}_1^\dagger\hat{a}_2$, $\hat{a}_1^\dagger\hat{a}_2^\dagger$, etc. A total of two interactions will be experienced, one involving phonon 1 and one with phonon 2. Interesting results start to emerge at this order when multiple terms exist in the Hamiltonian, such as with the QVSA Hamiltonian, see SI of [\[WMH25\]](#).

APPENDIX C

Floquet Theory

In this section, we derive the dynamics of motional excitation on a subharmonic resonance within the framework of multimode Floquet theory [ESM05, WLS22]. Consider an unknown signal tone ω_s to be detected, which is coupled to the trapping electrodes in a quadrupole configuration. Additionally, a probe tone ω_p is applied in a combination of dipole and quadrupole configurations as shown in Fig. 8.5. The resulting Hamiltonian representing the motional state of the harmonic oscillator and electric field coupling can be is:

$$\begin{aligned} \frac{\hat{H}}{\hbar} = & \omega_{r/z} \hat{a}^\dagger \hat{a} + \Omega_q (\hat{a} + \hat{a}^\dagger)^2 \cos(\omega_p t) \\ & + \Omega_d (\hat{a} + \hat{a}^\dagger) \cos(\omega_p t) \\ & + \Omega_s (\hat{a} + \hat{a}^\dagger)^2 \cos(\omega_s t + \varphi_s), \end{aligned} \quad (\text{C.1})$$

where $\omega_p = \omega_s + \delta + 2\omega_{r/z}/K$, $\Omega_q = eV_q \eta^2 / \hbar$, $\Omega_d = eV_d \eta / \hbar$, $\Omega_s = eV_s \eta^2 / \hbar$, $\eta = x_0 / r_0$ is effective Lamb-Dicke parameter, r_0 is the ion-electrode distance, $x_0 = \sqrt{\hbar / (2m\omega_{r/z})}$ is the zero-point wavefunction of the QHO, e is the electron charge, and δ is the detuning. Without loss of generality, we ignore the phase of the probe tones and use ω to represent $\omega_{r/z}$.

Unlike previous schemes [SKD00, ZRS02, CD00, TPA03] which have exclusively employed quadrupole tones, the present approach applies an additional probe tone in the dipole configuration for several reasons:

- The effective Lamb-Dicke parameter η associated with a dipole tone is of a lower order than that of quadrupole tones, which reduces the power threshold required to excite higher-order subharmonics.
- Application of a dipole tone induces motional excitation that generates a coherent state rather than a squeezed state. Coherent states operate within a

linear regime compared to squeezed states, making them more straightforward to manipulate compared to a squeezed state.

- Measurement of a coherent state provides higher Fisher information, leading to enhanced sensitivity in parameter estimation, such as for frequency measurements.

Transforming into the interaction picture with respect to $\omega\hat{a}^\dagger\hat{a}$, the Hamiltonian C.1 becomes:

$$\begin{aligned}
\frac{\hat{H}_I}{\hbar} = & + \Omega_q/2 \left(\hat{a}^2 e^{-i(2\omega+\omega_p)t} + \hat{a}^{\dagger 2} e^{i(2\omega+\omega_p)t} \right) \\
& + \Omega_q/2 \left(\hat{a}^2 e^{i(\omega_p-2\omega)t} + \hat{a}^{\dagger 2} e^{-i(\omega_p-2\omega)t} \right) \\
& + \Omega_q/2 \left(2\hat{a}^\dagger\hat{a} + 1 \right) \left(e^{i\omega_p t} + e^{-i\omega_p t} \right) \\
& + \Omega_d/2 \left(\hat{a} e^{-i(\omega+\omega_p)t} + \hat{a}^\dagger e^{i(\omega+\omega_p)t} \right) \\
& + \Omega_d/2 \left(\hat{a} e^{i(\omega_p-\omega)t} + \hat{a}^\dagger e^{-i(\omega_p-\omega)t} \right) \\
& + \Omega_s/2 \left(\hat{a}^2 e^{-i(2\omega+\omega_s+\varphi_s)t} + (\hat{a}^\dagger)^2 e^{i(2\omega+\omega_s+\varphi_s)t} \right) \\
& + \Omega_s/2 \left(\hat{a}^2 e^{i(\omega_s-2\omega+\varphi_s)t} + \hat{a}^{\dagger 2} e^{-i(\omega_s-2\omega+\varphi_s)t} \right) \\
& + \Omega_s/2 \left(2\hat{a}^\dagger\hat{a} + 1 \right) \left(e^{i(\omega_s t+\varphi_s)} + e^{-i(\omega_s t+\varphi_s)} \right)
\end{aligned} \tag{C.2}$$

Within the framework of multimode Floquet theory [ESM05, WLS22], the effective Hamiltonian $\hat{H}_I$ can be represented as an eight-mode Floquet Hamiltonian:

$$\bar{H}_I(t) = \sum_{\langle c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8 \rangle} \left(H_{(1)}^{(c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8)} + H_{(2)}^{(c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8)} + \right. \tag{C.3}$$

$$\left. \dots + H_{(K)}^{(c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8)} + \dots \right) \cdot e^{-i(\sum_{k=1}^8 c_k \omega_k)t - i(c_6 + c_7 + c_8)\varphi_s}, \tag{C.4}$$

where for $H_{(K)}^{(c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8)}$, the subscript K denotes the corresponding order of subharmonic excitation and the number of tones involved in the nonlinear process, while the superscript values $(c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8)$ represent the eight selectable tones – $\omega_1 = \omega_p + 2\omega$, $\omega_2 = \omega_p - 2\omega$, $\omega_3 = \omega_p$, $\omega_4 = \omega_p + \omega$, $\omega_5 = \omega_p - \omega$, $\omega_6 = \omega_s + 2\omega$, $\omega_7 = \omega_s - 2\omega$, and $\omega_8 = \omega_s$.

The Fourier components corresponding to the eight selectable tones are given as

follows:

$$\begin{aligned}
H^{(0,-1,0,0,0,0,0,0)} &= H^{(1,0,0,0,0,0,0,0)} = \Omega_q/2(\hat{a}^\dagger)^2, \\
H^{(0,1,0,0,0,0,0,0)} &= H^{(-1,0,0,0,0,0,0,0)} = \Omega_q/2(\hat{a})^2, \\
H^{(0,0,-1,0,0,0,0,0)} &= H^{(0,0,1,0,0,0,0,0)} = \Omega_q/2(2\hat{a}^\dagger\hat{a} + 1), \\
H^{(0,0,0,0,-1,0,0,0)} &= H^{(0,0,0,1,0,0,0,0)} = \Omega_d/2(\hat{a}^\dagger), \\
H^{(0,0,0,0,1,0,0,0)} &= H^{(0,0,0,-1,0,0,0,0)} = \Omega_d/2(\hat{a}), \\
H^{(0,0,0,0,0,0,-1,0)} &= H^{(0,0,0,0,0,1,0,0)} = \Omega_s/2(\hat{a}^\dagger)^2, \\
H^{(0,0,0,0,0,0,1,0)} &= H^{(0,0,0,0,0,-1,0,0)} = \Omega_s/2(\hat{a})^2, \\
H^{(0,0,0,0,0,0,0,-1)} &= H^{(0,0,0,0,0,0,0,1)} = \Omega_s/2(2\hat{a}^\dagger\hat{a} + 1), \tag{C.5}
\end{aligned}$$

When the resonance condition $\sum_{k=1}^8 c_k \omega_k = 0$ is satisfied, a higher-order subharmonic is excited, generating a motional displacement. The resonance condition can be reduced to $(\sum_{k=1}^8 c_k) \omega_s + ((\sum_{k=1}^5 c_k) 2/K + (2c_1 - 2c_2 + c_4 - c_5 + 2c_6 - 2c_7)) \omega + (\sum_{k=1}^5 c_k) \delta = 0$ to yield the constraints:

$$\begin{aligned}
c_1 + c_2 + c_3 + c_4 + c_5 + c_6 + c_7 + c_8 &= 0, \\
(2c_1 - 2c_2 + c_4 - c_5 + 2c_6 - 2c_7) &= -1, \\
c_1 + c_2 + c_3 + c_4 + c_5 &= K/2. \tag{C.6}
\end{aligned}$$

These constraints can be intuitively understood as a multi-phonon Raman transition. To generate a displacement resonant with the secular frequency ω , the number of probe tones must match the number of signal tones to cancel the carrier frequency ω_s .

Creating a general analytical expression for an arbitrary K th-order Hamiltonian that includes all parameters is nontrivial. Instead, we here derive an empirical formula.

The K th-order Hamiltonian $H_{(K)}^{(c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8)}$ involves $K-1$ commutation operations [ESM05, WLS22]. From C.5, all Fourier components $H_{(K)}^{(c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8)}$ are comprised of only first-order ($\hat{a}$, $\hat{a}^\dagger$) or second-order ($\hat{a}^2$, $\hat{a}^{\dagger 2}$, $\hat{a}^\dagger \hat{a}$) ladder operators. The commutation between ladder operators is nonvanishing only when the number of first-order ladder operators is exactly one, which occurs only if the dipole tone

contributes only one of K terms in the Hamiltonian. Together with the resonance constraints, we arrive at an empirical formula for subharmonic driving of a coherent state:

$$\alpha = \beta(\omega_s, K, \omega_{r/z}) \Omega_d \Omega_q^{(K-2)/2} \Omega_s^{K/2} e^{-i\varphi_s K/2}, \quad (\text{C.7})$$

where β is a function of the signal frequency ω_s , the secular frequency ω , and the subharmonic order K , and can be numerically calculated from QuTip simulations [JNN12, JNN13].

To validate the Floquet theory for the subharmonic excitation, we consider the case of $K=2$ as an example. The second-order term for eight-mode Floquet theory can be expressed as:

$$H_{(2)}^{(c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8)} = \quad (\text{C.8})$$

$$-\frac{1}{2} \sum_{\substack{c'_1, c'_2, c'_3, c'_4, \\ c'_5, c'_6, c'_7, c'_8}} \frac{[H^{(c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8)}, H^{(c'_1 - c_1, c'_2 - c_2, c'_3 - c_3, c'_4 - c_4, c'_5 - c_5, c'_6 - c_6, c'_7 - c_7, c'_8 - c_8)}]}{\sum_{k=1}^8 c_k \omega_k}, \quad (\text{C.9})$$

Evaluation of the sum Eqn. C.9 gives:

$$\begin{aligned} H_{(2)} &= 2 \left(H_{(2)}^{(0,0,0,1,0,-1,0,0)} + H_{(2)}^{(0,0,0,-1,0,1,0,0)} + H_{(2)}^{(0,0,0,0,1,0,0,-1)} + H_{(2)}^{(0,0,0,0,-1,0,0,1)} \right) \\ &= - \left(\frac{[H^{(0,0,0,0,0,-1,0,0)}, H^{(0,0,0,1,0,0,0,0)}]}{\omega_p + \omega} + \frac{[H^{(0,0,0,0,0,1,0,0)}, H^{(0,0,0,-1,0,0,0,0)}]}{-(\omega_p + \omega)} \right. \\ &\quad \left. + \frac{[H^{(0,0,0,0,0,0,0,-1)}, H^{(0,0,0,0,1,0,0,0)}]}{\omega_p - \omega} + \frac{[H^{(0,0,0,0,0,0,0,1)}, H^{(0,0,0,0,-1,0,0,0)}]}{-(\omega_p - \omega)} \right) \\ &= - \frac{\Omega_s \Omega_d}{4(\omega_p + \omega)} [\hat{a}^2, \hat{a}^\dagger] + \frac{\Omega_s \Omega_d}{4(\omega_p + \omega)} [\hat{a}^{\dagger 2}, \hat{a}] \\ &\quad - \frac{\Omega_s \Omega_d}{4(\omega_p - \omega)} [2\hat{a}^\dagger \hat{a}, \hat{a}] + \frac{\Omega_s \Omega_d}{4(\omega_p - \omega)} [2\hat{a}^\dagger \hat{a}, \hat{a}^\dagger] \end{aligned} \quad (\text{C.10})$$

$$= \frac{\omega \Omega_s \Omega_d}{\omega_p^2 - \omega^2} (\hat{a}^\dagger + \hat{a}), \quad (\text{C.11})$$

which is consistent with the result obtained from the Magnus expansion [WMH25].

The QHO squeezing operator $\hat{S}(r)$, generated via parametric excitation, can also be analyzed using Floquet theory. Similarly, application of a single quadrupole tone at frequency ω_p results in a Hamiltonian:

$$\frac{\hat{H}}{\hbar} = \omega \hat{a}^\dagger \hat{a} + \Omega_q (\hat{a} + \hat{a}^\dagger)^2 \cos(\omega_p t), \quad (\text{C.12})$$

where $\omega_p = 2\omega/K + \delta$ and $\Omega_q = eV_q \eta^2 / \hbar$.

Transforming into the interaction picture with respect to $\omega \hat{a}^\dagger \hat{a}$, the Hamiltonian C.12 becomes:

$$\begin{aligned} \frac{\hat{H}_I}{\hbar} = & \frac{\Omega_q}{2} (\hat{a}^2 e^{-i(2\omega+\omega_p)t} + \hat{a}^{\dagger 2} e^{i(2\omega+\omega_p)t}) \\ & + \frac{\Omega_q}{2} (\hat{a}^2 e^{i(\omega_p-2\omega)t} + \hat{a}^{\dagger 2} e^{-i(\omega_p-2\omega)t}) \\ & + \frac{\Omega_q}{2} (2\hat{a}^\dagger \hat{a} + 1) (e^{i\omega_p t} + e^{-i\omega_p t}) \end{aligned} \quad (\text{C.13})$$

The Hamiltonian can be treated as a sum of three Hermitian components, each composed of a factor Ω_i , ladder operators, and a time dependence $e^{\pm i\omega_i t}$, where $\{\Omega_1 = \Omega_q, \omega_1 = (2/K + 2)\omega\}$, $\{\Omega_2 = \Omega_q, \omega_2 = (2/K - 2)\omega\}$ and $\{\Omega_3 = \Omega_q, \omega_3 = 2\omega/K\}$.

Using Eqs. (A16) and (A17) in Ref. [WLS22], the K^{th} -order term of the Hamiltonian can be approximately expressed as $\hat{H} = \Omega^K / (\omega/2)^{K-1} (\hat{a}^2 + \hat{a}^{\dagger 2})$, creating a squeezing operator $\hat{S}(r)$ where $r \propto \Omega^K / (\omega/2)^{K-1}$.

C.1 Time-dependence of the K -th order Interaction

From Eqn. C.5, significant excitation occurs when near a motional Raman subharmonic resonance, i.e.

$$\sum_{k=1}^8 c_k \omega_k = \gamma, \quad (\text{C.14})$$

where γ is some small detuning from resonance and $\gamma \approx \delta \ll \omega \ll \omega_s, \omega_p$. This resonance condition can be expanded

$$\left(\sum_{k=1}^8 c_k \right) \omega_s + \left(\left(\sum_{k=1}^5 c_k \right) \frac{2}{K} + (2c_1 - 2c_2 + c_4 - c_5 + 2c_6 - 2c_7) \right) \omega + \left(\sum_{k=1}^5 c_k \right) \delta \quad (\text{C.15})$$

$$= \gamma, \quad (\text{C.16})$$

which in turn yields the constraints

$$\begin{aligned} c_1 + c_2 + c_3 + c_4 + c_5 + c_6 + c_7 + c_8 &= 0, \\ (2c_1 - 2c_2 + c_4 - c_5 + 2c_6 - 2c_7) &= -1, \\ c_1 + c_2 + c_3 + c_4 + c_5 &= K/2. \end{aligned} \quad (\text{C.17})$$

Collecting terms of order $O(\delta)$, the resonance condition [C.14](#) is only fulfilled when $(\sum_{k=1}^5 c_k) \delta = \gamma$ since $\gamma \approx \delta \ll \omega \ll \omega_s, \omega_p$. Use of [Eqn. C.16](#) with [Eqn. C.17](#) thus gives the result

$$\frac{K\delta}{2} = \gamma. \quad (\text{C.18})$$

The time-dependence of the K -th order Floquet Hamiltonian [C.4](#) is thus

$$e^{i\gamma t} = e^{-iK\delta t/2 - i(c_6 + c_7 + c_8)\varphi_s}, \quad (\text{C.19})$$

More generally, for near-resonant detunings, the K -th order Floquet Hamiltonian [Eqn. C.4](#) is similarly

$$\begin{aligned} \bar{H}_I(t) = & \sum_{\langle c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8 \rangle} \left(H_{(1)}^{(c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8)} + \dots + H_{(K)}^{(c_1, c_2, c_3, c_4, c_5, c_6, c_7, c_8)} + \dots \right) \\ & \cdot e^{-iK\delta t/2 - i(c_6 + c_7 + c_8)\varphi_s} + H.C., \end{aligned} \quad (\text{C.20})$$

Again, since we know the Hamiltonian can be written in a form of a displacement operator, [Eqn. C.20](#) can be simplified to:

$$\bar{H}_I(t) = C \cdot \hat{a} \cdot e^{-iK/2\delta t - i(c_6 + c_7 + c_8)\varphi_s} + C^* \cdot \hat{a}^+ \cdot e^{iK/2\delta t + i(c_6 + c_7 + c_8)\varphi_s}, \quad (\text{C.21})$$

where C is a δ -independent amplitude. Following [Eqns. \(14-20\)](#) of Ref. [\[LBD05\]](#), [Eqn. C.21](#) takes the form of an oscillating displacement where

$$\alpha = C \cdot \text{sinc}(K\delta t/4). \quad (\text{C.22})$$

Together with [Eqn. C.7](#), the displacement α can be made explicitly as

$$\alpha = \beta(\omega_s, K, \omega_r/z) \Omega_d \Omega_q^{(K-2)/2} \Omega_s^{K/2} e^{-i\varphi_s K/2} \text{sinc}(K\delta t/4), \quad (\text{C.23})$$

which is shown in [Eqn. 8.3](#) of the main text.

APPENDIX D

Cryogenic System

I want to include an appendix that covers some of the various troubles that we experienced with our cryogenic system.

In the initial Stinger line that we received, we experienced an incredibly high rate of clogs. This has been attributed to missing o-rings on the manifold, a leaking Aeroquip line, and a likely saturated filter. Within the blue housing (Fig. [D.1](#)), the Stinger line connects to tubing cooled to cryogenic temperatures through which the helium exchange gas flows via a VCR connection. The original Stinger line that was shipped to us had a filter that attaches to the tubing. However, this was a poor decision since the filter is exposed to air when the Stinger was shipped to us and any time the Stinger line was removed from the vacuum system. Exposure to air results in the filter becoming saturated with gas that will contaminate our system, leading to clogs.

Another challenge we faced were numerous cryogenic components breaking on us. We've had multiple compressors fail and need to be sent back to Sumitomo for repairs. Additionally, we have even had to resort to replacing the coldhead itself (Fig. [D.2](#)) which is very atypical for how new it was. You'll be able to pick out the sound of a damaged cryocooler, it is loud and just sounds unhappy. In our case, the bearing had started wearing off and created pile of dust within the coldhead housing. A Sumitomo technician was able to quickly come out and swap coldheads.

Related to the cryogenics but not a major problem, is when condensation forms on windows and feedthroughs. This is a known problem within some parts of the cryogenic community but wasn't relayed to me until after we already solved this problem. The solution is to heat the vacuum viewport with a couple of heater cartridges to keep it above the dew point ($\sim 18^{\circ}\text{C}$), and do a similar method with any

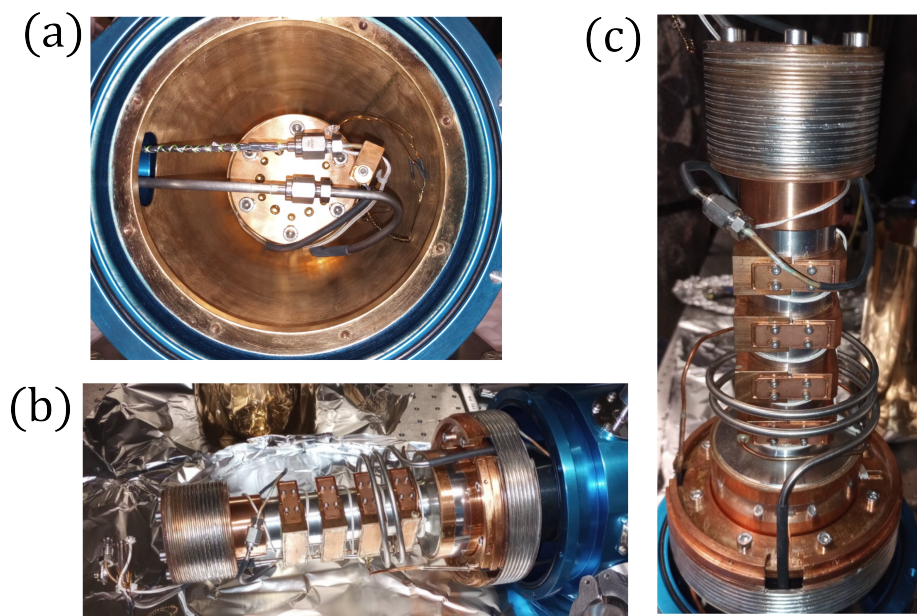


Figure D.1: What it looks like inside the blue housing. A grad student shouldn't have to know what this looks like, but I was not fortunate enough. This was opened up several times: swapping Stinger lines, adding a temperature diode to the second stage, and for cleaning the tubing.

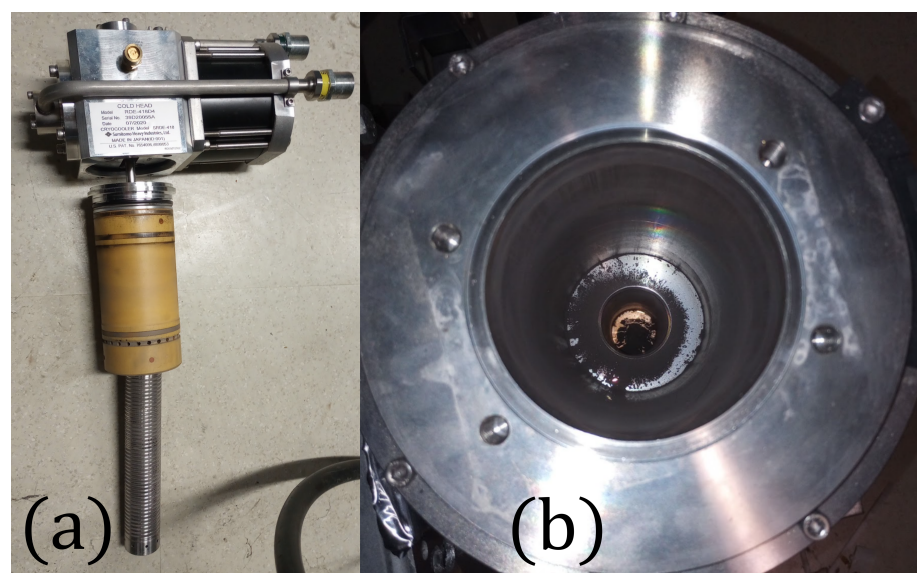


Figure D.2: Our coldhead needed to be replaced despite its relatively short lifetime.

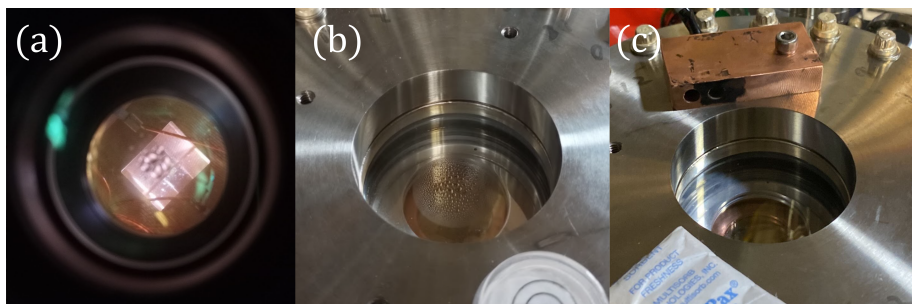


Figure D.3: Condensation formation on our reentrant viewport prior to adding heats to it (a) and (b). Our heated was epoxied inside a copper block that we mount directly onto the viewport.

feedthrough that may have this problem. In the latter case, we used wire as opposed to a heater cartridge. We also tried using desiccant pellets to absorb moisture on one of the feedthroughs. While this connection hasn't seen shorts since the pellets were installed, it's hard to say if they perform as well as a heating component.

We have also gone through several different types of filters, purifiers, and adsorbers to improve the purity of the gas in our recirculator line. Our standard is an additional HC-4E adsorber and a Supelco helium purifier.

The one time we decided to clean the tubing inside the blue housing required that we disconnect the return side of the Stinger line. We pressurized solution and sent it through the supply line including the capillary tube. Solution steps includedalconox/DI water solution, followed by deionized water, and finally isopropyl. Dry nitrogen was then sent through the line to ensure no liquids remained. This was an annoying procedure that I would not advise implementing unless absolutely necessary. For the return line we just used isopropyl and dry nitrogen; a VCR connection was required to ensure none of the alcohol splashed into the blue housing. FYI, if EHS asks this never happened.

APPENDIX E

Charcoal Getter

Cryopumping offers a powerful tool for getting UHV pressures without needing to bake a vacuum chamber for weeks, as well as lifts some material constraints such as many plastics [Eki06, Wel01, OH03]. However, cryopumping can experience limitations once the pumping surface has become coated in frozen gases, saturating it and reducing the pumping efficiency, resulting in higher pressures. Gases with high freezing points pose less of a limitation because they can form many layers before saturation, but for hydrogen gas, which freezes at 14K for atmospheric pressures, it becomes saturated with a monolayer [VF98, NIS13]. With cryogenic shielding reaching 4K, the smooth surfaces offer only a small area onto which the hydrogen gas can freeze [Boz19]. Introducing a cold surface with a large surface area provides a significant increase in pumping capacity. An established pumping source is activated charcoal, often referred to as a charcoal getter. The carbon structure is very complex and offers a very large surface area compared to its volume. It is typical that charcoal is packaged with copper mesh pressing it against another copper surface to ensure conductive cooling of the getter material; if the getter material is not able to get cold enough it will not be able to pump all gases [III22]. Other options increase using thermal epoxy or solder to fix the getter material to a copper surface.

One major challenge when working with this material is the installation and baking of it. As soon as the charcoal is exposed to air, it's going to absorb gases, and without reaching hot enough temperatures, it will not expel the gases. Additionally, the getters can become saturated and will no longer be able to absorb gas until they have been heated to remove the gas. This last step is in context to a system that is already under vacuum and is being actively pumped at room temperature. As such, the ideal charcoal getter should be mounted onto a heater, but this can be a difficult contraption to design for a cryogenic system. The heating element should ideally

only heat the getter material, not the cryogenic shielding, but the getter material must have an excellent thermal connection to the cryogenic shielding to reach the necessary temperatures to pump out gases.

We made an attempt in our experiment to introduce charcoal getters beside our ion trap. The getter material was activated coconut charcoal, the same material that was used in the cryogenic buffer gas experiment (N.I.C.E.), with the largest chunks picked out (several mm wide). About 8-10 pieces were then placed on a small copper block, with a copper mesh solders on three sides using lead-free solder, through the unsoldered side. The last side was then soldered to try to introduce a good thermal contact between the charcoal and the copper block. A single screw then mounted the copper block to the 4K shielding, which also tightened the copper mesh. Two of these pieces were assembled, and prior to installation they underwent a vacuum bake to expel any gases that they had previously absorbed. Afterwards, they were quickly installed into our vacuum chamber beside the ion trap before pumping the chamber again to minimize any additional exposure to air. No heater element was attached to the getters, with the hope being that there wouldn't be a significant amount of gas that they would be exposed to, so they shouldn't be saturated. However, either from becoming saturated or from not reaching cold enough temperatures from poor thermal conductivity, these pieces did not seem to improve our vacuum. In fact, we removed them at a later date out of concern that they were outgassing near the ion trap and ruining our ion lifetimes. Part of this concern stemmed from the fact that when we load molecules we have gas entering the 4K shield, which has relatively low vacuum conductance, so the gas would more likely be confined within this space when cryogenic.

The current method for improving our vacuum lifetime when cryopumping appears to be saturated is by doing a quick warmup (also called burping). By turning off the coldhead compressor for 5 minutes, the cryogenic shielding increased by tens of Kelvin and gas that was previously adhering to the shielding leaves the surfaces. We believe that it then is able to be pumped using the NEG/ion pump. Typically a quick warmup is done every week or every other week depending on whether we have been leaking gas to load molecules or are doing experiments on chains of ions.

Chains are more susceptible to collisions with background gas, requiring our lowest pressures for long lifetimes.

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