

**Ramsey-Bordé matter-wave interferometry with a thermal
calcium beam for optical frequency stabilization $\leq 10^{-16}$**

by

Judith Olson

B.S., Ithaca College, 2011

B.M., Ithaca College, 2011

M.M., University of Colorado at Boulder, 2013

M.S., University of Colorado at Boulder, 2016

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written by Judith Olson
has been approved for the Department of Physics

Prof. Scott Diddams

Andrew D. Ludlow

Date _____

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Olson, Judith (Ph.D., Physics)

Ramsey-Bordé matter-wave interferometry with a thermal calcium beam for optical frequency stabilization $\leq 10^{-16}$

Thesis directed by Prof. Scott Diddams

This dissertation details the developments of a calcium thermal beam atom interferometer designed to act as an ultra-stable frequency reference over a broad range of timescales. The obtained fractional frequency instabilities of the final apparatus reached $\leq 10^{-16}$ after 10's of seconds of averaging without the aid of vibration isolation or significant apparatus temperature stabilization. The vastly superior stability performance of this calcium system compared to other thermal atomic references was facilitated by the enormous atom numbers from the thermal beam, which can be efficiently addressed with Bordé-Ramsey spectroscopy on the well-studied $(4s^2)^1S_0$ - $(4s4p)^3P_1$ ($m=0$) clock transition in neutral ^{40}Ca . This method yields interrogation laser frequency dependent interference fringes with a linewidth of 1.6 kHz. This measurement is further enhanced with an electron-shelving detection scheme used for state detection after the interferometry process, causing each excited state atom to emit many hundreds of photons in the detection region.

The exceptional instability results from this atom interferometer highlight an underutilized realm of precision measurement science based on relatively simple thermal ensembles. An intuitive understanding of the Bordé-Ramsey process is presented and relevant experimental techniques explained such that this dissertation should prove useful to a broad readership.

Dedication

There are people in this world without whom I could not imagine having made it to where I am today. No words could fully convey their importance to me, though here I will try.

To the strong women in my life, who encouraged me to pursue my passions, however unconventional, with reckless abandon. To my mother, who embraced my enthusiasm for understanding the world around me and never showed doubt that I could accomplish nearly anything I set my mind to. And to her mother, Ginny, who did the same for her. I am eternally grateful for all of the sacrifices they made for my betterment. Though they'll never read these words, they knew them in their hearts long ago.

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Lastly, I owe a great deal of my development to the wonderful physics department at Ithaca College, where I spent my formative undergraduate years. The support and help of professors like my adviser, Bruce Thompson, was absolutely invaluable. The positive environment created by the Ithaca College physics department (including but not limited to Matthew C. Sullivan, Dan Briotta, Matthew Price, Andrew Crouse, Beth Ellen-Clark-Joseph, Jennifer Mellot, Luke Keller, and Bodhi Rogers) taught me what kind of teacher, researcher, and person I want to be.

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Preface

It is not merely the results, but rather *how* they were arrived at that provides the most useful information for an experimental physicist. While not quite conventional, I thus intend this dissertation to also provide laboratory guidance for any who might endeavor to replicate this or similar experiments regardless of their initial skill level in the field. While these discussions may seem tedious to some readers, I find such examples to be educational and informative. However, for those desiring more straight-forward result churning, I point them to the second-to-last chapter, where I have attempted to be brief and straight-forward.

This calcium experiment was undertaken with a very small group of people, with originally only myself and NIST staffer Richard Fox regularly working in the lab before it largely became my sole endeavor. With the calcium project being isolated from the main laboratory group and relying on methods that had largely been abandoned decades earlier, I wish I had a resource similar to this dissertation to guide my efforts in designing, building, and testing the Ramsey-Bordé experiments of this dissertation. While journal articles sometimes subtly hint at potential pitfalls or failed attempts, I try to be very frank here and derive useful information from *why* and *how* some experimental choices were abandoned. Thus, I intend this document to help those in similar situations to myself seven years ago, i.e. those who are perhaps newer to frequency metrology and forced to work more independently than is prudent. May it serve as a learning tool and guide for those looking to pursue similar lines of research or learn more about methods used in experimental AMO physics.

Chapter 1

Introduction

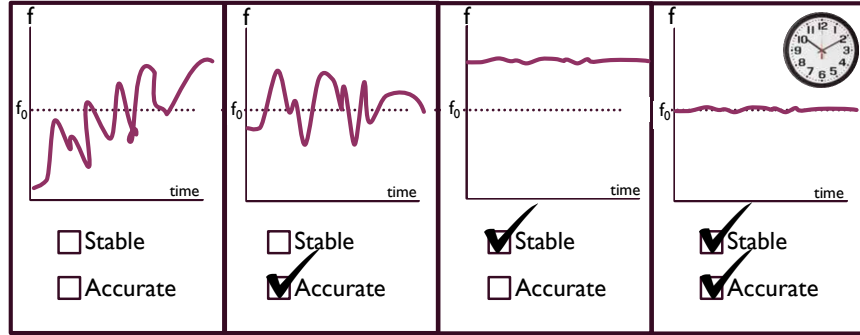
This thesis details how Ramsey-Bordé atom interferometry, also called simply Bordé interferometry, can be used to stabilize a laser with fractional frequency instabilities never before recorded from a thermal atom ensemble. I demonstrate here instabilities $\leq 10^{-15}$ at < 1 s and $\leq 10^{-16}$ at tens of seconds. Though frequency stability at this level typically requires either high-Q Fabry-Perot cavities or complex optical lattice clocks, here we explore how Bordé spectroscopy makes it possible for a relatively simple experimental apparatus to yield similarly excellent frequency stability. Thus, in this dissertation, the relevant physical theories and experimental design considerations are viewed through the lens of frequency metrology, and this introductory chapter centers upon the world of stable frequency references, including field-specific terminology, relevant history, timekeeping contribution, and varied research applications. Then the specific advantages of Ramsey-Bordé for ultra-stable optical frequency generation is covered, followed by atomic properties of calcium that make it well-suited for Ramsey-Bordé laser stabilization via a thermal beam. An overview of this dissertation is then given.

1.1 Stable frequency reference and clock basics

Clocks are essentially a combination of two components: an oscillator and a counter [5]. The oscillator provides the ‘ticking’ of the clock, while the counter keeps track of the number of ticks that have passed to provide timing information. Often the oscillating portion of a clock is referred to as the frequency reference or local oscillator (LO). Clocks are typically characterized

two frequency parameters: stability (i.e. precision) and accuracy (i.e. uncertainty). The difference between these two properties are displayed in Figure 1.1. Stability depends on how much the frequency varies between measurements while accuracy depends on how the values vary around the anticipated frequency (f_0 in Fig. 1.1).

Figure 1.1: Visualization of stability and accuracy characteristics for measurements of a frequency with values relative to f_0 plotted over time. Primary clocks ultimately need stability and accuracy, but it is obvious that a purely stable frequency source can be offset to act as an excellent timekeeper.



A ‘good’ clock possesses both exceptional stability and accuracy, i.e. it ticks very regularly and the rate at which it ticks is extremely well known. Because clocks (specifically in this dissertation, atomic clocks) cannot be perfectly isolated from external perturbations that shift their frequencies, most clocks obtain their accuracy at least in part from exceptionally well controlled environments and carefully calibrated sensitivities. Some clocks tick very repeatedly but their frequency shifts are difficult to calculate, such as inhomogeneous effects caused by atomic velocity distributions. As we shall see, this is the case for our Ramsey-Bordé system thermal atomic beam system, which has inherent uncertainties that limit its accuracy above 10^{-15} . However, such systems can still function extremely well as frequency references for a great number of applications. If lower uncertainty is needed, it must be derived from comparison with a more accurate frequency reference.

A good example of clock reliance on multiple sources of accuracy and stability is the cesium frequency standard defining the International System (SI) second. A cesium frequency standard consists

of a microwave oscillator whose frequency is locked to that of a 9.19 GHz hyperfine transition in atomic cesium. The cesium clock at NIST is now accurate to near a part in 10^{16} , but its frequency data must be averaged for many days or weeks to obtain this accuracy. For shorter times, the hydrogen maser bank serves as the local oscillator and a long-term steering correction is added based on the well-averaged cesium frequency. In fact, a calibrated and predictable frequency reference can provide much of the utility that accurate clocks do. For instance, both can be used to tell time, measure distances, or assess properties of other frequency sources. In fact, the maser bank is so reliable these days, that even after 500 days of running, the differences between the cesium-steered maser frequency and un-steered maser frequency time predictions differed by only 6.3 ns total with an average time deviation of 1.6×10^{-16} over the entire interval [196]. Very little difference was observed between these two timescales and the International Atomic Time (TAI). This example shows just how powerful highly stable frequency sources can be, and also highlights an exciting future application for Ramsey-Bordé thermal beam systems to play the role similar to a maser in a future optical timescale.

1.1.1 Quantum projection noise and atomic clocks

A generic frequency reference can be parameterized by two properties: the quality factor (Q) of the resonance and the signal-to-noise (S/N) of the resonance's measurement. The Q is given by the resonance width ($\Delta\nu$) relative to its nominal frequency (ν_0) such that $Q = \nu_0/\Delta\nu$ and therefore Q is dimensionless. For quantum absorbers, this is given by the spectroscopic properties of the transition resonance being utilized. Meanwhile, the S/N depends on a variety of factors, but, for a single measurement of a two-level quantum system, the maximum value is given by the quantum projection noise (QPN) [135] of the ensemble $S/N = \sqrt{N_{atom}}$, where N_{atom} is the number of atoms contributing to the resonance signal in each frequency measurement. For such uncorrelated white noise characterizing QPN, the fractional frequency instability (σ) of such an oscillator after an averaging time (τ) is approximated as $\sigma(\tau) = \frac{\Delta\nu}{\nu} \sqrt{\frac{1}{N\tau}}$. However, many atomic clocks are based on passive frequency stabilization, where the atom is sequentially probed and its response determines

a frequency correction. Thus, there is some finite time for each frequency correction, which takes a total cycle time T_c , and thus the minimum time over while frequency corrections are considered is for $\tau \geq T_c$. Additionally, specific spectroscopic signals often require an additional scaling factor, κ , such that $0 < \kappa \leq 1$. This term often includes calculations of the signal contrast¹ and lineshape factors, such that

$$\sigma(\tau) \propto \frac{\kappa \Delta \nu}{\nu} \sqrt{\frac{T_c}{N \tau}}, \quad (1.1)$$

where it should be obvious then that this equation only applies for $\tau \geq T_c$. This formula is easily applied to the main categories of quantum-absorber frequency references available today, and it would be helpful now to give a few examples of its use to better understand the potential strengths of a thermal beam frequency reference.

For instance, state-of-the-art optical clock systems are pushing to achieve linewidths of order one Hz or less [42, 168], so $\delta \nu / \nu \approx (1 \text{ Hz}) / (500 \text{ THz}) = 2 \times 10^{-15}$. Resolving such a narrow spectral feature (thus measuring a very precise energy) requires a finite amount of time by the energy-time uncertainty principle ($\Delta E \Delta t \geq \hbar/2$); therefore, in this case, a measurement requires a finite time, of order $\delta t \approx \pi \text{ Hz}^{-1}$, or roughly one second. Thus, most modern-day atomic clocks utilizing such narrow transitions only make measurements of the atomic state about every second, i.e. $T_{\text{cycle}} \approx 1$ s. Thus an optical lattice clock with a reasonable number of atoms, say $N = 5000$, could in theory support 1 s instability of 3×10^{-17} if only limited by quantum-projection noise.

In contrast to the previous optical lattice clock example, the thermal beam ensemble utilized in this dissertation work has a linewidth that is about 1000 times larger, with $\delta \nu / \nu \approx 3.5 \times 10^{-12}$. However, a broader linewidth transition requires less time to resolve, and thus here $T_{\text{cycle}} \approx 10$ ms. In that 10 ms, a billion atoms are interrogated, with $N = 10^9$ being a conservative estimate. Thus, in principle, a quantum-projection noise near 1×10^{-17} could be obtained in a second.

¹ A contrast term is common in interferometric systems utilizing, for example, Ramsey fringes and the Ramsey-Bordé system of this dissertation. In these cases, the fringe amplitude often does not extend entirely from the maximum to minimum atomic response due to imperfections in the interrogation process or ensemble inhomogeneities, thus requiring the additional modification to the quantum projection noise formula.

Furthermore, the thermal beam system is far less complex, utilizing fewer lasers, less stringent vacuum requirements, and takes less space and electricity, making it an attractive choice for atom-based ultra-stable frequency generation.

Of course, optical lattice clocks are developed for their frequency accuracy as well, though low instability is also employed to realize reach desired accuracy targets in reasonable measurement times. However, many common atomic clock applications can be provided without the need for cutting-edge accuracy at a few parts in 10^{-18} , or even accuracies many orders of magnitude above this. This is because a great deal of information can be accurately related to the relative frequency shift as opposed to the absolute frequency shift of a measured variable over certain timescales. Moreover, frequency references can be easily calibrated to provide reasonable accuracy between ultra-accurate frequency updates acting as steering corrections, such as is done with a maser bank and the cesium atomic clocks of NIST to maintain the timescale.

It is implicit in this discussion that optical atomic clock systems require laser frequency pre-stabilization, as initial laser linewidths relative to the spectral feature widths are often an important factor in determining whether the atom-laser process will be viable. Over time, technological improvements to laser pre-stabilization and control have enabled a variety optical atomic clocks with previously unfathomable performance. Of course, when any technical problem is “solved” with an atomic clock, it has really just been minimized such that a different limiting factor becomes the roadblock to improved stability or accuracy. It is this relentless push for better performance that has led atomic timekeeping from its cesium origins to ever more precise and accurate frequency references.

Atoms first became central to timekeeping in 1967, when the International Bureau of Weights and Measures (BIPM) redefined the second in terms of a hyperfine transition in cesium [81]. Since then, atomic clock methods have continued to improve, with recent cesium primary frequency standards showing uncertainties approaching 10^{-16} [114, 139]. However, other atomic species have been utilized to create novel types of optical atomic clocks capable of accuracies far better than those of the best cesium measurements. In some ways, these high-uncertainty frequency measurements

defy the very definition of the second because absolute frequency metrology is defined only in terms of the less accurate cesium transition. Two such examples include novel optical lattice clocks described earlier and specialized ion traps, both of which have demonstrated frequency uncertainties approaching 10^{-18} [189, 181, 134].

The inherent measurement advantage of optical over microwave frequencies in quantum projection noise is perhaps too large for microwave standards like cesium to ever reasonably overcome. Indeed, it is expected that eventually the second will be redefined [217]. Of course, work is actively undergoing to pursue improved control of even higher frequency radiation like x-rays, which could excite nuclear transitions in atoms and molecules [198, 46]. Such transitions would have even more shielding from environmental effects than the electronic transitions in the optical domain demonstrated here. One day they may very well replace the optical clocks we hold in such high regard now.

Another trend in pursuit of narrower quantum-absorber optical frequency references that has been alluded to is the cooling, slowing, and trapping of these quantum absorbers. This can reduce the first- and second-order Doppler effects to manageable levels, but it comes at the cost of the number of atoms. Without the ability to control the position or speed of the quantum absorbers within an ensemble, velocity selection [110, 203, 205, 149, 51] was one of the only viable options for reducing second-order Doppler shift uncertainties. Velocity selection allows only a small, selected portion of the total ensemble to interact with the laser field. While the shifts were more straight-forward, the signal was comprised of far fewer atoms. Later methods of cooling [204, 269] and trapping [182, 210] were able to be more efficient. With neutral atoms these methods could create far denser quantum ensembles than were previously possible and rely on many thousands or millions of atoms. However, with the increased density came problems including frequency shifts and increased collisions and correlations that hadn't been seen before. Nonetheless, cooling, trapping, and similar methods are extremely powerful and have been essential to the development of ultra-accurate frequency standards through improved Q and reduced frequency shifts. However, this trend has also led to quantum-absorber systems being associated with the longer-cycle times

needed to both to prepare (cool and trap) the ensemble and then interrogate an ultra-narrow transitions.

Though atomic systems have steadily come to be associated with low-bandwidth frequency corrections as they pursued higher Q , atomic systems can also be utilized for fast and high fidelity frequency discrimination. We achieve this with the Bordé interferometer through instead emphasizing a comparatively enormous N value from a thermal beam while accepting the inherent accuracy limitations. For the Ramsey-Bordé calcium thermal beam system, the accuracy is limited by exactitude of both the second-order Doppler shift when averaged over the ensemble velocities and the phase of the Bordé oscillatory spectroscopic signal. Both of these shifts are hard to characterize and calculate to better than perhaps a few tens to hundreds of Hz with specialized techniques, but their uncertainty would be difficult to improve beyond perhaps a few parts in 10^{-14} . Though, once calibrated (and if well isolated from environment fluctuations), our system shows potential to maintain instabilities near 10^{-16} for extended periods of time, during which measurements could be made with similar levels of accuracy provided the system’s nominal frequency was previously well measured.

1.1.2 Cavities and combs

Despite the potential for high- N ultra-stable quantum absorber sources, another technology has largely filled this optical frequency stabilization niche for timescales ≤ 1 s. This technology is the widely used Fabry-Perot cavity. Fabry-Perot cavities rely on two highly reflecting surfaces fixed by a spacer such that the optical path length experienced by a laser beam coupled into the cavity remains steady. Advanced techniques like phase modulation of the laser light coupled to the cavity allow for extremely high S/N frequency corrections to the lasing source, such as through Pound-Drever-Hall stabilization [75, 29]. Meanwhile, electro-optics allow for very high bandwidth for these corrections. Though extremely successful and a cornerstone of laser spectroscopy, cavity frequency stabilization performance is nearing a rather insurmountable barrier to future improvements in the form of thermal noise. Dubbed the ‘thermal noise problem’ by many in the field, this issue stems

from the ever so small motion of the atoms and molecules comprising the reflecting surfaces [190]. So long as these particles are at a finite temperature, their Brownian motion limits the stability of the optical path length, thus the ultimate laser frequency stability.

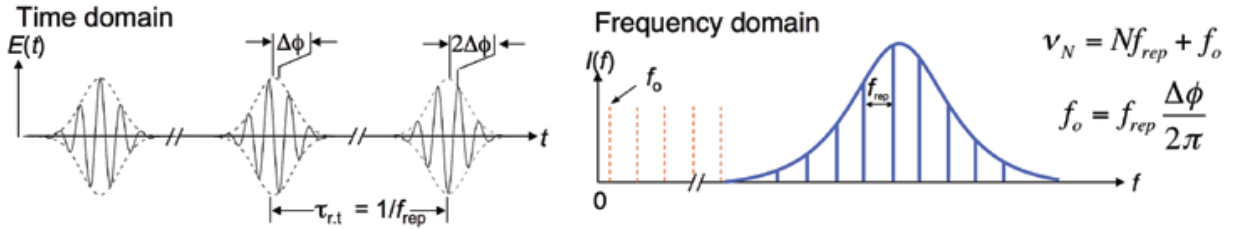
Of course, cavity specialists have found ways to reduce the impact of thermal noise on the resulting frequency instability. These methods include specialized crystalline reflecting surfaces [57], cavity lengths nearing the limits of manufacturing processes [107], and operation of the cavity at cryogenic temperatures [220]. Even with these advances, the measured and predicted thermal noise limits for optical cavities are at or above the earlier predicted quantum-projection noise of the Bordé interferometer around which this dissertation centers. In the future, stabilities beyond the thermal noise limitations of Fabry-Perot systems will most likely be desired, and quantum-absorber systems based on high-flux thermal beams may be the most practical way forward. In fact, the necessary infrastructure is already in place to use and transfer such low 10^{-18} stabilities throughout the electromagnetic spectrum thanks to continued development of cutting-edge frequency combs.

When it comes to actually disseminating and using optical frequencies, discovery of the frequency comb [68] around the turn of the 21st century enabled relatively straightforward transfer of optical stabilities down to electronically accessible microwave and RF frequencies [86]. Prior to the advent of frequency combs, laboratories often required entire rooms with specialized equipment dedicated to individual harmonic down-conversion chains [123, 262], which were first demonstrated in 1983 by Jennings *et al* [141]. These systems typically had to be designed for measuring a specific optical frequency, thus could only be applied to very limited ranges of the electromagnetic spectrum at any given time. In contrast, frequency combs can access broad regions of the infrared (IR) and visible spectra and allow for simultaneous measurement of many different frequencies, all within a relatively compact system.

Frequency combs now play a critical role in comparing and evaluating the performance of stable optical frequencies. In this dissertation work, access to various frequency combs generously supplied by Tara Fortier, Holly Leopardi, and other optical-frequency measurement group members at NIST became essential to most every frequency measurement made of the Ca experiment.

We use the comb to evaluate our calcium-stabilized system performance via comparisons against NIST's world-class clocks and cavities. Combs enable these measurements with their large periodic structure of frequencies. The comb tooth closest to the external frequency source to be compared is usually chosen, and the beat note between these two optical frequencies can then be measured very well with electronically accessible frequencies such as through a frequency counter. The combination of the comb beat note, the comb tooth spacing, f_{rep} , and the initial comb frequency offset, f_0 can then be used to calculate the optical frequency with very high accuracy and precision using the equations of Fig. 1.2.

Figure 1.2: The basic time and frequency domain views of a frequency comb are demonstrated, along with relevant variables characterizing the frequencies of the individual comb teeth spaced by the repetition rate frequency, f_{rep} . The common equations for utilizing combs in frequency measurements are also given, with ν_N being the frequency of the N th comb tooth that is beat against the measured optical frequency. For more details consult [194].



Frequency combs generate their tooth-like frequency spectrum using very short pulses of light, as depicted in Figure 1.2. The comb tooth spacing is extremely regular, allowing for spectral stability transfer at the 10^{-18} level through optical fibers [163]. However, open-air frequency transfer is complicated by the path length fluctuations from air currents, humidity changes, and numerous other factors that generally limit stability transfer near 10^{-16} [171], though recent work suggests specialized transfer protocols could perhaps enable improved performance in the future [236].

Of course, not all laboratories are fortunate enough to possess such stable frequency sources or comb systems for measurements. In this case, the common tool is the three-cornered-hat measurement [100]. It requires instead three frequencies to be compared rather than just the two

sources used in the previous arrangement, but it can allow stability measurements to be carried out when no clear best reference source is available.

Now that the basic terminology has been established for discussing the precision optical frequency reference at the heart of this dissertation has been covered, we can discuss the role this research plays in both the research and practical worlds.

1.2 Motivation and use for ultra-stable frequencies

Considerable research attention has been devoted to the development of stable frequency references. Historically, the role of stable frequency sources has been strongly tied to their utility in the areas of position, navigation, and timing (PNT). Mechanical oscillators were meticulously developed to be clocks, and were eventually eclipsed by the use of crystal oscillators in the early 20th century [167]. These new frequency references quickly became central to sonar systems on submarines before spreading into broadcast and communications systems throughout the globe. These advances in technology were largely allowed by the improved precision provided by crystal oscillators. Of course, the enormous amount of research performed on these oscillators also led to them being conveniently scalable to very small sizes, enabling their integration into cell phones, clocks, televisions, radios, and countless other locations.

In modern times, frequency references of numerous types are widely accessible thanks to the development of satellite systems with accurate clocks that orbit the Earth. Standardized protocols have been developed that simultaneously cater to applications requiring disparate levels of frequency stability and accuracy. This approach (e.g. GPS) has been so successful that frequency references are now central to modern navigation on earth, and they are expected to be the basis of future space navigation systems to explore our galaxy, mine asteroids, and identify potentially devastating objects passing through our solar system [62, 208]. Many security and authentication protocols for data transfer and communications rely on timing, including network authentication protocols. Future database communication systems are moving toward higher quality frequency references to allow for faster data transfer with higher degrees of certainty, such as in the finance

sector. Modern security and defense systems are also beginning to incorporate high performance local frequency references to avoid problems like GPS spoofing.

In addition to these uses for frequency references, two particularly exciting examples will be explored in the following subsections: (1) the impact of frequency references on atomic clock development, and (2) how improved frequency references and systems utilizing them (like atomic clocks) are enabling studies of exotic phenomena and rare physics.

1.2.1 Improving atomic clocks

One of the most effective means of improving atomic clock performance in the last decade has been to take advantage of narrow transitions identified in atomic species like Yb, Sr, Al^+ , Yb^+ , and others. These species are largely chosen for possessing an electronic structure that yields an accessible ultra-narrow optical transition with a linewidth of order 1 Hz or less. As previously discussed, interrogation of such a narrow spectroscopic feature usually entails cycle times of order one second, the majority of which can be spent on readying the atomic sample for spectroscopy via cooling, trapping, and state preparation. For this dark time duration, changes in the pre-stabilized laser properties such as intensity, frequency, and phase can cause systematic shifts or periodic fluctuations of the perceived spectroscopic frequency, degrading the obtainable accuracy of the clock.

One rather insidious type of frequency noise exacerbated by longer dead times within the measurement cycle is known as the Dick effect [71, 226]. In the Dick effect, high-frequency laser noise is aliased down onto lower frequencies by the clock sampling procedure. To reduce Dick effect noise, the pre-stabilization system can be further improved, thereby reducing the accumulated noise during the measurement. Alternatively, the clock system can be modified or extended so as to constantly be sampling the atoms, as in a zero-dead-time configured dual-lattice clock [229].

In addition to providing a way to reduce the Dick effect, improved laser pre-stabilization from novel optical frequency references could enable interrogation of even narrower spectroscopic features far below a Hz linewidth. These ultra-high Q resonances could enable more accurate or

stable clocks, thus considerable research attention is being devoted to resolving these ultra-narrow atomic transitions [175]. Though cutting-edge optical clocks have already established their relevance as a tool for measurements of fundamental constants, verifications of relativity and quantum-optical phenomenon, and probes of subtle higher-order effects, such clocks offer still more reasons to improve their accuracy and stability. Atomic clock performance is nearing a regime where table-top level particle physics is being considered. Historically, this regime of high-energy particle physics experiments has been limited to enormous and expensive accelerator experiments. However, the sheer sensitivity and accuracy of atomic clocks are now allowing scientists to consider testing such theories in low-energy laboratories as well. Such searches for new physics, enabled by improved frequency references, are covered in the next subsection.

Lastly, the better the clock laser pre-stabilization, the quicker the desired accuracy of a clock can be achieved. This is because clock frequency measurement processes tend to possess noise processes that “average down,” meaning the certainty of the data tends increase over time, i.e. as more measurements of the frequency are made. It once was the case that advanced clock laboratories required teams of people to run the clock for many days or weeks continuously to reach the desired accuracy. With improved pre-stabilization, modern atomic clocks are now able to make precision measurements much faster than before. The increased speed with which extremely accurate clock measurements can be made also allows them to assess improvements or changes to the apparatuses more efficiently, leading to an increased pace of optical clock improvements and development.

1.2.2 New physics

The unprecedented accuracy and precision available to scientists by advanced optical clocks and frequency references has opened a new regime of ultra-sensitive scientific study once thought to be beyond existing research capabilities [108, 254]. Frequency is one of the most precisely measurable quantities available to humanity, with values able to be accurately given at a part in 10^{18} thanks to optical frequency standards. Extremely small effects can be measured with optical

frequency metrology and related technology, such as the small length variations caused by passing gravitational waves or fluctuations in fundamental constants. Thus, the certainty and stability demonstrated by novel frequency references is of interest to a wide variety of burgeoning research topics including quantum computing, cosmology, gravitational theory, and tests of the standard model.

The extreme control over the frequency and phase of tunable laser sources enabled by ultra-precise frequency references (like our calcium interferometer) is required for some high-fidelity quantum computation and simulation [152, 15, 170, 16, 161, 166]. Moreover, the calcium system's frequency instability falls below a Hertz and could be further improved, potentially allowing it to serve as a phase reference for quantum technologies [252, 155, 30]. Additionally, atomic clocks and frequency references can make excellent sensors for exotic phenomena. For instance, by monitoring spectroscopic signals generated from very stable and phase controlled atom-field interactions, gravitational potentials could be explored with exacting precision [199, 148, 156, 222, 84]. Similar experiments also function as gravitational wave sensors [74, 97]. Other approaches leverage precision frequency measurements for tests of general and special relativity [73, 187, 214] or to measure subtle effects only on the edge of finally being experimentally resolved [11].

One particularly exciting new horizon for frequency references is that of table-top scientific studies once only feasible in the high-energy domain, i.e. with accelerator systems that tend to be large and expensive. Rather than having to move to higher energy scales where exotic phenomena may be easily visible, scientists are beginning to consider if such studies of particle physics, tests of the standard model, and searches for new physics that could instead be done at low-energy scales by leveraging the precisions of time and frequency metrology [33, 69, 250]. Similarly, the level of accuracy with which physicists can both predict and measure the energy of quantum transitions enables tests of novel physical theories. Thus, frequency references can be used as tools for confirming predictions of the standard model [34, 201], tests of fundamental physics [253, 60, 2], and consistency of fundamental constants [94, 207]. At the 2018 meeting of the BIPM standards meeting in Paris, where I was graciously allowed to sit in on a session regarding future changes

to fundamental constants, the committee remarked on how input from particle physicists and quantum-chromodynamic simulations could aid their evaluations. The precision and accuracy from some of the spectroscopic measurements the standards committee utilizes are beginning to breach into such realms, and future advances in the precision metrology will likely rely on incorporation of advanced theories and subtle effects.

Precision frequency references like the calcium system (as well as cavities and other atomic clocks) are also proposed to be potential detectors for transient dark matter or energy effects [99, 239, 111, 98, 215, 70]. In particular, axionic and dilaton scalar dark matter [63] would be fairly straightforward to detect with an array of atomic clocks or frequency references [9, 255, 45], as the electronic structure of atoms, ions, etc. is expected to be slightly shifted by the presence of an axionic dark-matter wave passing through the earth. Such a shift detected by multiple frequency references simultaneously across the globe would verify the presence of scalar fields passing through the earth or even be used similar to a telescope to predict the clumped dark matter trajectory for future observation. Of course, more complicated forms of topological dark matter or energy could also couple to spectroscopically resolvable transitions in quantum matter, allowing for tests of novel theories of gravity and searches for new forces in the universe [238]. The unique bandwidth of the calcium system of this dissertation could be particularly helpful in providing constraints on dark matter [260, 117].

In addition to frequency references measuring energy and frequency, they can also be used to measure length very accurately. Such a length measurement is the exact mechanism on which modern day gravitational wave observatories are dependent. For instance, the Laser Interferometer Gravitational-Wave Observatory (LIGO) [1], is essentially just one of humanity's most carefully calibrated measurements of length change as the gravitational waves cause the interferometer arm length to predictably fluctuate as the space-time ripple passes. A space-based gravitational wave observatory called the Laser Interferometer Space Antenna (LISA) is in development [85, 8, 258], which would then orbit around the sun in a nearly free-fall path while measuring the tiny variations in the laser-detected distance between them.

And, of course, there is the favorite catchall of all cutting edge science: applications we can't even fathom yet. New physical theories and ideas are being proposed routinely, and with further maturation and technological development, it is hard to fathom what new physics precision measurement science might uncover. The continued improvement to frequency standards will certainly help scientists along this path, and the system presented here utilizing Ramsey-Bordé spectroscopy is another step forward.

1.3 Ramsey-Bordé spectroscopy and interferometry

The experimental implementation of the Bordé method used in this dissertation largely derives from a 1984 paper simply entitled *Optical Ramsey fringes with traveling waves* [41]. In this publication, the potential for a unique laser geometry to interrogate a thermal beam of atoms was discussed, and it was suggested that the system could be an excellent frequency discriminator. This proposal was born at a time of great excitement in the optical spectroscopy world. Lasers were finally reaching stability levels that allowed for resolution of spectral features far narrower than anything seen before. Spectroscopists were eager to see if they could develop an optical atomic clock capable of higher accuracy than the existing microwave cesium standard. As such, many of the ideas that led to the success of the microwave standard were being considered in the optical domain. Most notable of these, within the scope of this dissertation, was the use of multiple interactions for Ramsey-like interrogation in the optical domain.

Ramsey-Bordé interferometry is only one of several methods to generate optical Ramsey fringes. Other methods rely on differing laser-atom interaction schemes for use with velocity selected atoms. The early optical Ramsey methods were first explored in the 1970's and 1980's by theorists including Chebotayev [49], Dubetskiui [77], Salour [223], Bordé, and many more. However, spectroscopic work with optical transitions faced new difficulties, such as increased first-order Doppler shifts. Optical Ramsey fringes were far more sensitive to the motion of atoms than the earlier microwave versions.

To overcome the problems associated with atomic velocity distributions in the optical domain,

scientists had two main paths forward. First, they could find Doppler-insensitive methods [265, 102] to reduce or remove the first-order Doppler shift. Some experiments even used velocity selection to isolate the relatively stationary atoms within an ensemble to reduce the first- and second-order Doppler shifts. However, a much more direct way forward was in development around the same time - slowing/cooling and trapping of atoms and ions. This was usually accomplished through additional lasers or fields to dissipate kinetic energy from the quantum ensemble, ultimately reducing the sample temperature, thus also reducing its motion [202, 268]. Of course, it took quite a few years before these methods became widely used. In the mean time, nonlinear optical-Ramsey methods were resolving impressively narrow features, being incorporated into atomic clocks of various species, and being used to observe physical phenomena for the first time [120, 61, 235]. By the end of the decade, aspects of both slowed atomic experiments and optical Ramsey methods had come together and were being proposed for improved fountain clocks [109].

Despite the alluringly large flux of atomic beams, the future of timekeeping clearly lay with slowed or trapped quantum absorbers. For instance, the inhomogeneous second-order Doppler shift from our calcium atomic beam is more than one kHz. For some time this was still a competitive accuracy level to have obtained, but as atomic clocks progressed further, uncertainties continued to drop, and challenges associated with the characterization of a thermal beam's relativistic Doppler shift to desired accuracy levels became clear. Bordé and other optical Ramsey methods are still heavily used, but now more commonly implemented via chopped or pulsed lasers acting on trapped (thus relatively stationary) quantum ensembles.

Then, in 1989, Bordé authored an important paper [38] that reinterpreted the 1984 Bordé spectroscopy method as a momentum-space matter-wave interferometer. It was one of the first instances of matter-wave interferometry being used to describe an atomic system using lasers as beam splitters². By 1989, matter-wave interferometry [65, 232, 177, 178, 52] was already developed with neutrons, electrons, and other light particles with deBroglie wavelengths much greater than

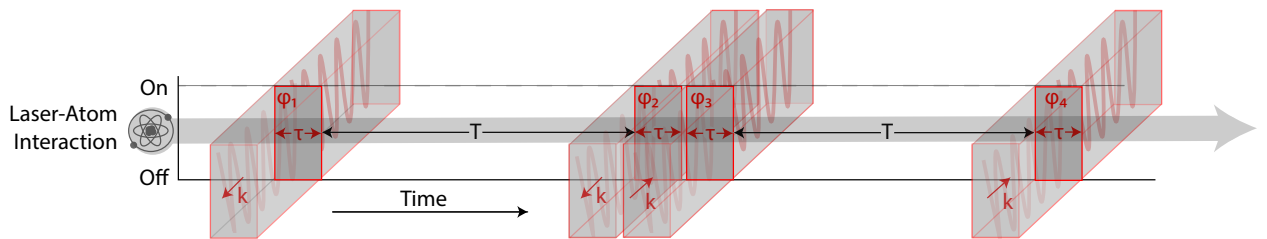
² Matter-wave interferometry was first seen in atoms in 1930 with He and H₂ diffracting off a crystalline solid [82]. Neutrons weren't discovered for another two years [48], and they were soon used to demonstrate interferometry as well [257].

thermal calcium atoms ($\lambda_{dB} \approx 17$ pm). The shorter the deBroglie wavelength, the harder it is to resolve the matter-wave interference, so it was not immediately obvious that matter-wave interference fringes might be demonstrated for massive atoms or molecules. However, once the connection was made clear by Bordé and others [53, 176, 150] around 1989, atomic physicists were quick to adopt proven techniques of electron and neutron interferometry. Today, atomic interferometers are used for various types imaging, crystal holography, scanning tunneling microscopes, and numerous other methods more often associated with condensed matter and solid state physics than atoms or optics.

Because of the solid theoretical and experimental foundation already built, atom interferometry was rapidly assimilated by spectroscopy and AMO fields and quickly became its own physics subfield. Atom interferometry is now commonly used to measure atomic properties (e.g. electric polarizability, van der Waals forces) [80, 184, 124], gravitational fields [132, 199, 148, 84], inertial motion (e.g. rotations, accelerations) [104, 142, 162], atomic recoil [263], and refraction indexes [231] to name a few.

Though potentially powerful, atom interferometry can be challenging to experimentally realize on thermal ensembles like our calcium beam. Because Ramsey-Bordé utilizes four atom-laser interactions separated in space and time as seen in Figure 1.3, the experimental design must accommodate multiple laser interactions with an extended atomic beam. Furthermore, the laser interactions must be phase coherent to produce resolvable interference fringes from the calcium ensemble.

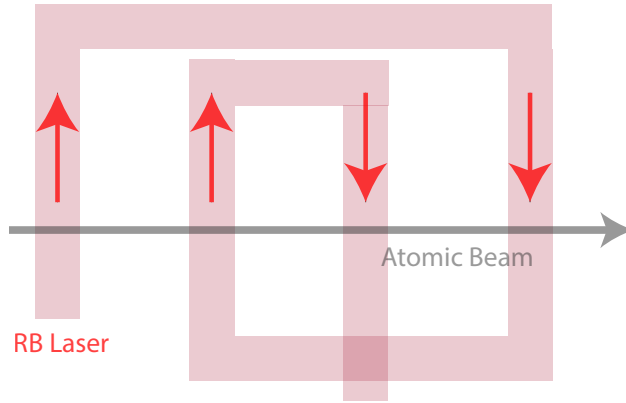
Figure 1.3: The Ramsey-Bordé method is based upon four precisely controlled laser-atom interactions. The laser wavevector reverses direction between the second and third interactions. The laser must have a definite relative phase between all four interactions for the characteristic Ramsey-Bordé interference fringes to have phase coherence across the ensemble.



The difficulties of establishing phase coherence between the laser-atom interactions originally

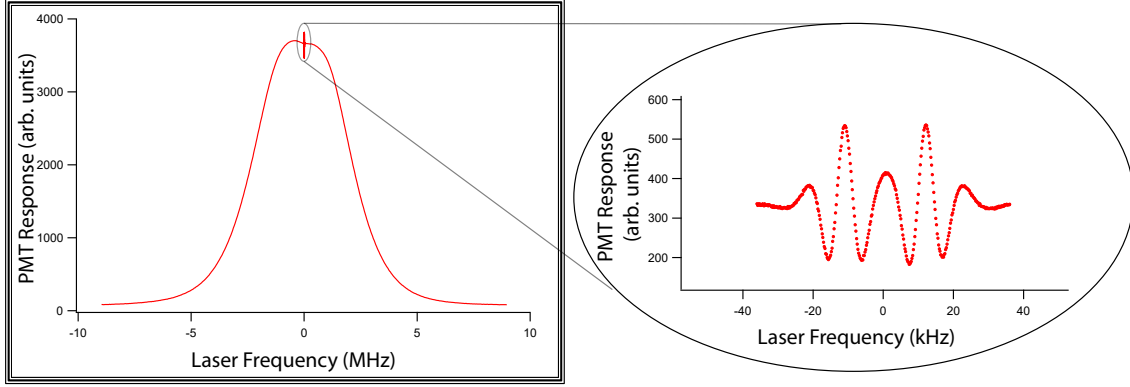
solved by utilizing a single laser source, which would then be steered along a path so as to intersect the atomic beam in multiple locations, often referred to as a traveling-wave geometry. For the case of Ramsey-Bordé, the traveling wave laser is directed to create two counter-propagating pairs of co-propagating laser paths, as depicted in Figure 1.4.

Figure 1.4: Typical folded traveling wave geometries wrap the single laser source upon itself to create the Ramsey-Bordé interaction sequence for a thermal beam of atoms. Often a double cat's eye lens system or numerous corner cubes are used instead of numerous flat reflections as pictured here.



During a Bordé interaction sequence, a portion of the calcium atoms undergo self-interference, which can be reasonably approximated by a two-level system. Self-interference results when an atom's quantum mechanical wave-packet is split then recombined such that multiple, indistinguishable quantum trajectories could have been taken before recombination. In the case of Bordé interferometry, two distinct closed interferometers exist, and the resulting interference fringes from them are encoded onto the atomic state probability. A great deal of technical details also determine the Ramsey-Bordé fringe pattern. If these other variables remain relatively stationary, then the atomic population for an interrogation laser near the fringe's optical frequency resonance also gives information about how the laser frequency has moved since the previous measurement. Thus, Ramsey-Bordé in the calcium system can very easily be used as a frequency discriminator, yielding information about the changes in the interrogation laser frequency. An example of interference fringes from our thermal calcium beam spectrometer is shown in Figure 1.5.

Figure 1.5: The image on the left shows a broad Doppler sweep of the atomic response as it crosses over the transition resonance in an optimized Ramsey-Bordé configuration. To resolve the fringes here, the sweep function had to be slowed and oversampled in the fringe region. The figure on the right represents the general spectroscopic response within 10's of kHz of the fringes. These signals are from one of the earliest versions of our Bordé interferometry apparatus, Ca1.



Due to the inherent symmetries in the Ramsey-Bordé method, two closed interferometric pathways are simultaneously formed, each shifted from the bare atomic transition resonance by the energy of a single laser-atom recoil interaction, as presented in Figure 1.5. The number of resolvable fringe cycles depends upon the velocity distribution, which ultimately causes the fringes to dephase across the ensemble away from the recoil-shifted resonance. Relatively stationary atomic ensembles, such as those in a magneto-optical trap or lattice, can be made to display many tens of cycles of the interference pattern. However, only a few fringe cycles are needed for frequency discrimination. Thus, the fringe dephasing inherent to inhomogeneously broadened thermal beams is not catastrophic, so long as the atomic beam is well enough collimated to produce a period or two of the interference fringes.

The finer details of the interferometer will be covered in more detail later, but the basic experimental variables dictating the final fringe oscillation properties are worth discussing here. The Bordé fringe oscillation period is determined by Ramsey time, which is also sometimes called the dark time or dark precession time. This is the temporal period between the 1st and 2nd or, equivalently, the 3rd and 4th laser-atom interactions of Figure 1.4, and it is often given for thermal ensembles instead by its physical length. The longer the Ramsey time, the narrower the

resulting Ramsey-Bordé fringes become and the steeper, thus more sensitive, the discriminator slope becomes for frequency corrections. The fringe linewidth can ultimately reach or even exceed the natural linewidth of the transition [191, 192], and, in our case, gets to within a factor of four. The phase of the interference fringe pattern is determined by the relative phase of the four laser interactions, and its amplitude is determined by the number of atoms that coherently participated in the given fringe's interferometer

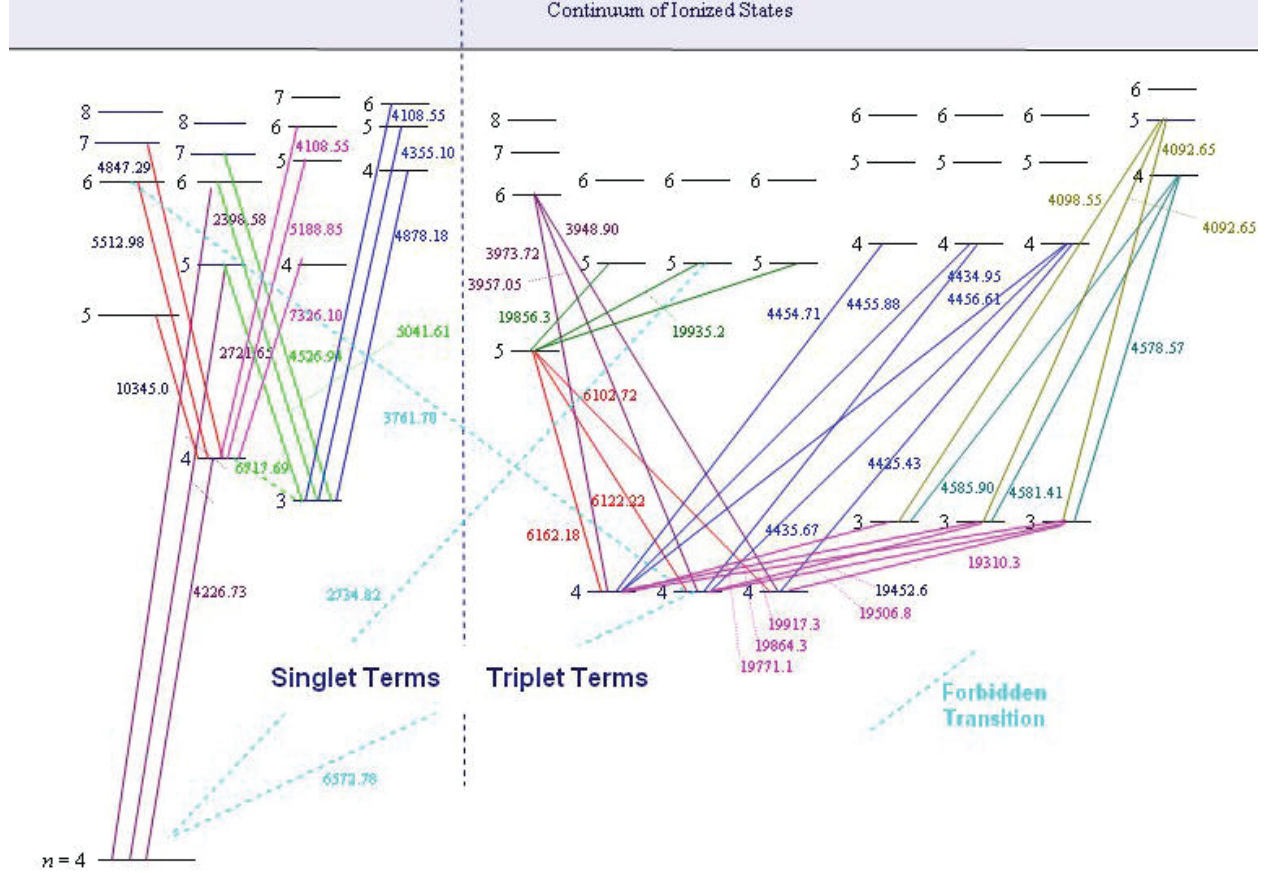
The choice in atomic species greatly impacts the capabilities of the resulting system - both its physical size and spectral resolution. Calcium, the atom used for this dissertation work, makes a good choice for Ramsey-Bordé, as discussed in the next section.

1.4 Calcium atoms

Now that the basics of Ramsey-Bordé interferometry have been explored and its historical context covered, the choice of calcium for the atomic species used here can be more fully understood. Calcium was the third species to be used for demonstration of optical Ramsey fringes, after neon and methane. In fact, optical Ramsey- fringes in calcium were first resolved at the National Institute of Standards and Technology (NIST) in Boulder [18], the same facility wherein this dissertation work was carried out. Calcium has a long history at NIST, largely owing to its relatively simple, two-electron valence structure. The interactions between these two electrons cause the alkaline-earth elements to possess two distinct symmetry classes called the singlet (para-) and triplet (ortho-) states, with the prefixes in parentheses being traditionally associated with the simplest two valence electron atom: helium. Transitions between these differing parity states are forbidden in principle, but some coupling does persist for two-electron elements more complicated than helium. Thus these types of transitions are not absolutely forbidden, but instead are called semi-forbidden or intercombination transitions. A reprinted³ Grotrian diagram of neutral calcium's energy levels is given in Fig. 1.6, with the singlet and triplet opposite states segregated.

³ Figure reprinted with pending permission from Clark Christensen[50] of Brigham Young University

Figure 1.6: The energy level structure of neutral ^{40}Ca , reprinted. The transition wavelengths are given in Ångstrom. Transitions between electronic states of opposite parity, i.e. between the singlet and triplet, are traditionally forbidden. Realistically however, Ca I possesses some weak coupling between these states, which results in a desirably long-lived excited state that can be used as an atomic clock transition.



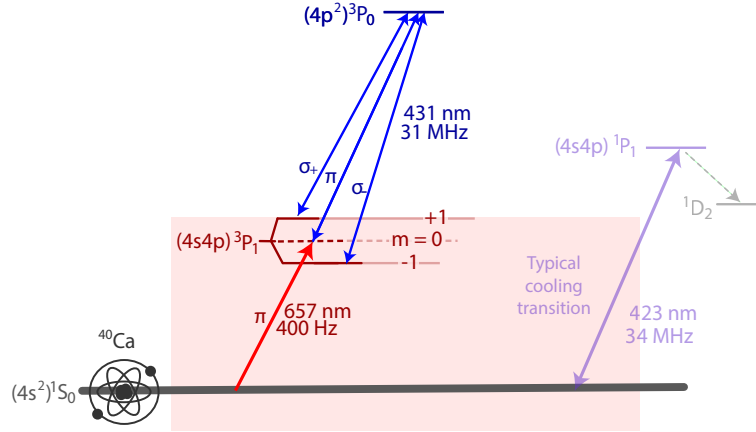
Because intercombination transitions are semi-forbidden, the excited state tends to be longer lived than for an allowed transition and is correspondingly narrower as well. These are the exact types of transitions that are typically employed in modern optical clocks. Ultra-high Q atomic clock species include many 2-electron atoms including Ca, Mg, Sr, and Yb.⁴ It is calcium's intercombination line from the singlet $^1\text{S}_0$ to the triplet $^3\text{P}_1$ level at 657 nm that we use for Ramsey-Bordé spectroscopy throughout this dissertation. Conveniently, the 657 nm transition can be sufficiently driven for the Ramsey-Bordé interrogation with reasonable diode laser powers for typical beam

⁴ Though a not an alkaline earth, the lanthanide Yb has an electronic structure that effectively mimics a two-electron valence structure: $[\text{Xe}] 4f^{14} 6s^2$.

profiles.

In addition to neutral calcium's well-studied 657 nm intercombination line, the atom also possesses transitions useful for trapping and cooling. The calcium laboratory at NIST shifted its focus to a pulsed Ramsey-Bordé interferometer acting on calcium atoms cooled in a magneto-optical trap (MOT) [210]. The calcium MOT was successful, and, for a period of time in the 1990's and early 2000's, research laboratories around the world worked with calcium to develop state-of-the-art optical clocks based on ^{40}Ca . However, calcium's electronic structure does not possess desirable transitions for optimal second-stage cooling, which limited the obtainable temperature, and thus accuracy of the calcium MOT clock. Furthermore, the optical lattice clock unveiled in 2005 [246] quickly came to dominate neutral atom optical clock methods with Sr and Yb, which possessed more favorable slowing, cooling, and trapping capabilities than calcium.

Figure 1.7: Relevant electronic levels of calcium are portrayed, including their polarization, linewidth, and average lifetimes. Transitions commonly used in calcium but not utilized in the experiment presented here are also given.



Despite calcium not being entirely compatible with novel atomic clock methods, calcium's structure is uniquely well-suited for the thermal-beam Ramsey-Bordé method used in this dissertation. The standard clock transition at 657 nm has a linewidth of 375 Hz, which is too broad for sub-Hz ultra-accurate clock studies. Here, the 375 Hz linewidth here corresponds to a lifetime of approximately 400 μs , during which an average atom moving ≈ 600 m/s travels ≈ 25 cm, which

is a convenient size for an apparatus. A distance somewhat less than this is actually used in the apparatus, as spontaneous decay otherwise degrades our signal-to-noise. To avoid loss of S/N, we typically utilize an interferometer length less than the distance traveled in the average calcium excited-state lifetime, with fringe linewidths near 1.6 kHz (about four times the atomic transition natural linewidth).. Thus, a thermal beam calcium Bordé interferometer requires a few tens of cm in its longest dimension and can be made compatible with rack-mountable housing. The system size could be further miniaturized if the ensemble velocity were reduced.

Calcium also possesses a doubly-excited state from the Ramsey-Bordé interrogation transition's excited state, via $^3P_1 - ^3P_0$. Excited by 431 nm light, both valence electrons in the calcium atom are excited ($4p^2$). Furthermore, this transition is allowed, and so has a short lifetime convenient for rapidly cycling atoms in an electron-shelving like technique. This blue laser causes each Bordé excited atom to emit many thousands of photons within the detection region, thereby offering a means for greatly improving the measurement signal-to-noise.

More details in regards to calcium, Ramsey-Bordé methods, and the physical principles that guided design and development of our improved experimental apparatus are discussed in the main body of the dissertation.

1.5 Thesis overview

In Chapter 2, I cover important aspects of frequency metrology, including an overview of the state of the field for stable frequency sources. An emphasis is placed on the Allan deviation and experimental evaluation of noise properties. Though the calcium system's frequency instability results are not presented here, this chapter provides necessary context and background that sets the stage for later discussions and noise analysis.

Chapter 3 covers the theoretical basis leading to Ramsey-Bordé, including important history and technological developments that impacted the course of Ramsey-Bordé research within AMO sciences. Building from Rabi and Ramsey's work, the difficulties of resolving Ramsey fringes in the optical domain are also discussed. Though the theoretical derivation of Bordé-Ramsey fringes is

left to Bordé's paper given in the appendix, practical implications for the theory are given in this chapter along with Monte-Carlo simulation results of the theoretical expression.

Chapter 4 launches into a detailed discussion of the first apparatus used in our laboratory to study Ramsey-Bordé with a single atomic beam, Ca1. Initial experimental obstacles are presented, including successes as well as some failures. Conclusions are drawn about various noise contributions to the calcium-corrected frequency instability, and results indicating room for improvement are highlighted. Many of the ancillary systems required to run Ca1 and the later Ca2 are also discussed here.

Chapter 5 builds off the short-comings elucidated from Ca1 to present an improved apparatus called Ca2. A large amount of detail is given here on the physics apparatus design as well as laboratory practices utilized to ensure its success. This system also lays out important terminology and the experimental setup upon which the following chapter's results are based.

Chapter 6 provides numerous results from the Ca2 apparatus, including measures of various noise sources and systematic effects. Sensitivity of the Ca2 apparatus to numerous perturbations are discussed, and the final instability results are put within the present-day context of the field.

Chapter 7 briefly discusses a series of proposed improvements to the Ca2 apparatus as well as additional details on other possible applications for the Ca2 or similar systems.

Chapter 2

Stable frequency references and characterization

The properties of stable frequency references fall within a specialized area of measurement science often isolated from standard physics or engineering curricula. Yet, the analysis tools provided here, such as the Allan deviation, are absolutely crucial for understanding the later presented calcium system results. Though this chapter may feel slightly disjunct from those around it, I found it necessary to include this discussion here so that the concepts it lays out can be easily referenced where appropriate in the subsequent chapters. For instance, prior understanding of noise analysis and assessment of stable frequencies provides important incite for gauging how well the calcium system was working and what future developments would provide the largest improvements to its frequency stability.

Noise in atomic clocks generally falls into two categories. The first is that of the physical apparatus, which largely pertains to the atoms themselves, and it includes any processes that cause either the amplitude or phase of the atomic signal to change. The second is that of technical noise, which includes the detection system and subsequent electronic processing, as well as the frequency correction mechanism we use. Both segments of the calcium beam system must work in tandem to allow low-noise and high-bandwidth frequency corrections. Conveniently, the formalism for noise is general enough to allow both aspects of the calcium system to be assessed similarly, and will be discussed in reasonable depth throughout this chapter.

This chapter opens with an overview of noise in atomic clocks and then delves into specific noise types, divided into their respective frequency dependencies. The main metric of frequency in-

stability called the Allan deviation is then discussed, including the various types of Allan deviations and how they differ and assessing the calcium data.

2.1 Relevant noise types

The common types of frequency and phase noise processes seen in atom-based systems will be briefly discussed, including their possible origins. These are mathematically modeled through modulating either the frequency (FM) or the phase (PM) of an otherwise ideal oscillator signal. Readily recognizing their appearance in streaming frequency measurements over time is a valuable skill for developing and maintaining an atomic clock system, thus for each noise process a sample of frequency measurements over time is also given. Filtering of the signal or changing of the averaging time can reveal the noise processes on differing timescales. Though sometimes the noise culprit is clear or the noise type obvious, often atomic systems display a range of competing noise contributions that can make identification and improvement difficult for a stable frequency reference, as is the case for the calcium system described within this dissertation.

Since many of the noise sources do not possess white noise behavior, standard statistical tools, like the standard deviation, that rely on Gaussian distributions are inadequate to quantify the performance of many clocks. To this end, clock scientists rely heavily on a more general form of statistical analysis called the Allan deviation. The Allan deviation relies on comparisons between adjacent sequential measurements of, in this case, frequency values. This method is capable of handling frequency drifts, random walk, etc, which are discussed in more detail in the following sections.

Noise can be divided into types based on the frequency dependence of the noise, i.e. the slope of the phase noise power spectral density (PSD) versus frequency plot. Throughout this section the phase noise power spectral density will be the primary metric used, though there are many other comparable methods for noise analysis. Noise that scales with a significant negative exponent (such as $1/f^2$ or f^{-4}) more strongly impacts the low-frequency end of the spectrum far more than the exponentially reduced higher frequency content. Meanwhile, white PM goes as f^0 , meaning that

this noise type impacts all frequencies equally. However, measurement quirks of spectral analysis tools like the fast Fourier transform (FFT) can cause windowing or other problems¹ resulting in degradation of the flat slope or spectral response approaching DC, i.e. 0 Hz. The impact of similarly low-frequency sources can be difficult to assess because of this measurement limitation. However, many low-frequency noise sources are slow and predictable enough to be accounted for.

What determines whether a noise source is ‘low’ or ‘high’ frequency largely depends on the bandwidth of the system in question. For our calcium system, with measurements commonly lasting only $\delta t = 10$ ms. By Nyquist sampling theorems, such a 10 ms measurement cannot distinguish frequencies higher than $1/(2\Delta t) = 50$ Hz. However, low-passing a signal close to this value can be difficult. Noise components well above this frequency tend to average out over the integration period and have smaller fluctuations, though this does not always preclude them from causing measurement difficulties.

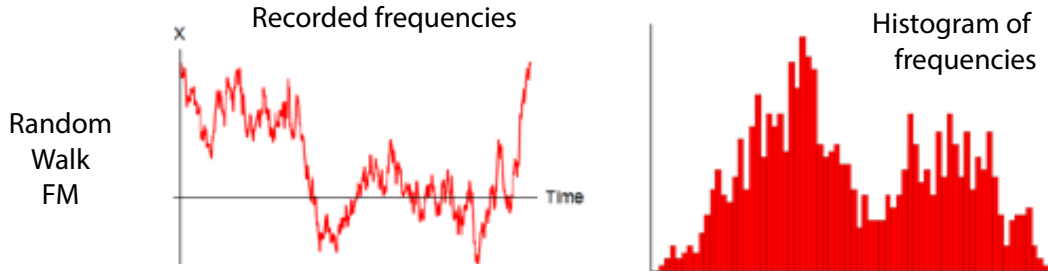
The main types of noise processes seen in passive atomic frequency standards are now briefly discussed, including common sources of the noise type and what the time-domain frequency data looks like for the noise process.

¹ This is largely a technical issue stemming from the finite time required to measure a frequency is reciprocal to the frequency being measured. Luckily, filtering or stabilization of systems at such low frequencies is common, and for the remaining noise generous averaging can still provide good measurements.

2.1.1 Random-walk FM ($1/f^4$)

The random-walk FM phase noise PSD decays as $(1/f^4)$, causing it to impact the system predominantly at lower frequencies and over longer timescales than other noise sources. Often random-walk FM corresponds to the atomic system's environmental influence. Sources of this noise can stem from mechanical vibrations, temperature fluctuations, pressure changes, or any other process that can cause the atomic frequency being referenced to randomly step up or down. It is largely absent in both time and frequency dissemination systems and telecommunication networks, but can be a difficult long-term concern for atomic clocks, cavities, and quartz oscillators. An example of random-walk FM noise is given as a frequency-time series and a histogram of the resulting frequency deviations from average are given in Figure 2.1.

Figure 2.1: Top: Simulated random walk frequency modulated data series in time. Bottom: Histogram of the measured frequency spread to demonstrate its shape. This figure is reprinted with permission of Alex Rollings[221].

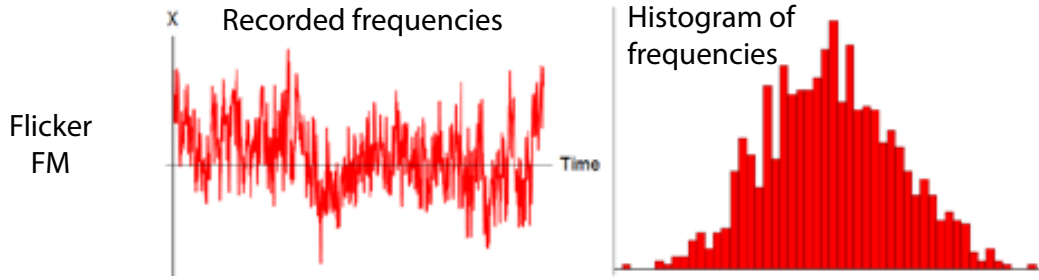


2.1.2 Flicker FM ($1/f^3$)

The flicker FM phase noise PSD decays as $(1/f^3)$ and is inherent to most electronic oscillators, including those often used for timing and frequency references in various servo systems required to operate an atomic clock. One common example is the flicker FM contribution seen in many Fabry-Perot cavities at medium timescales. In lower quality electronic oscillators, the flicker FM noise is often dwarfed by the white FM or flicker PM processes at short timescales. When dominant, flicker FM appears on both the modified and standard Allan deviation as a flat line. This relatively level portion is often called the flicker floor of the system. With flicker FM noise, additional averaging no

longer benefits the measurement stability, though it does not worsen the frequency instability. If a signal is monitored for long enough after hitting a flicker floor, its Allan deviation usually begins to turn up as random walk FM and frequency drifts begin to prevail. Often flicker FM is attributed to measurement noise. An example of flicker FM noise and its Allan deviation is given in Figure 2.1.2.

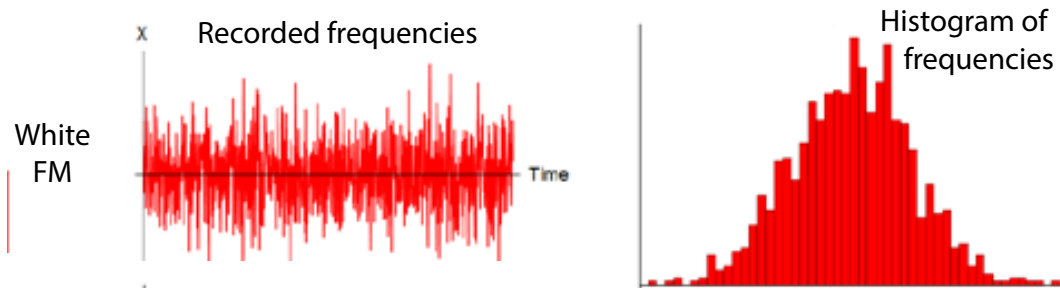
Figure 2.2: Top: Simulated flicker frequency modulated data series in time. Bottom: Histogram of the measured frequency spread to demonstrate its shape. This figure is reprinted with permission of Alex Rollings[221].



2.1.3 White FM ($1/f^2$)

The white FM phase noise PSD decays as $(1/f^2)$. White FM is quite characteristic of passive atomic stabilization systems, wherein the interrogation laser's frequency deviations relative to the atomic resonance yield random fluctuations around a nominal frequency. White FM noise is present in most time and frequency components, ranging from atomic clocks to quartz oscillators and time and frequency dissemination systems. It can be accentuated when the local oscillator linewidth is of order or larger than the narrow resonance being interrogated. An example of white FM noise and its Allan deviation is given in Figure 2.1.3.

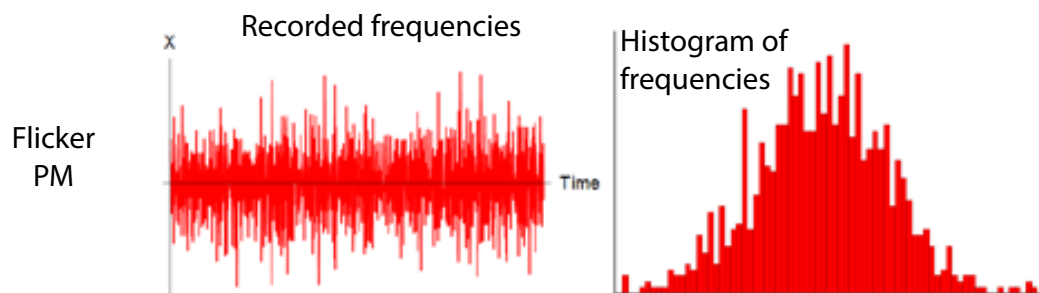
Figure 2.3: Top: Simulated white frequency modulated data series in time. Bottom: Histogram of the measured frequency spread to demonstrate its shape. This figure is reprinted with permission of Alex Rollings[221].



2.1.4 Flicker PM ($1/f$)

Flicker PM's phase noise PSD decays as $(1/f)$ and is often caused by the electronics used for the frequency servo. Flicker PM is often associated with signal amplification after the detection element and frequency multiplication, i.e. most non-linear electronic processes. Specialized electronics designs and part selections can often reduce flicker PM. It is a cascading type of noise caused by fundamental time constants required for dissipation of energy and information in processes ranging from water flow in the world's lakes down through resistors, diodes, vacuum tubes, and notoriously in MOSFETs. Conveniently, flicker PM noise is not often a limiting factor in atomic clocks, though it, along with white PM noise, can sometimes cause problems with quartz oscillators, time and frequency dissemination systems, and telecommunication networks. An example of flicker FM noise and its Allan deviation is given in Figure 2.4

Figure 2.4: Top: Simulated flicker phase modulated frequency data series in time. Bottom: Histogram of the measured frequency spread to demonstrate its shape. This figure is reprinted with permission of Alex Rollings[221].



2.1.5 White PM (f^0)

The phase noise PSD of white PM (f^0) is commonly caused by amplification stages in electronics used to process the atomic signal. Unwanted white noise on an amplified signal can sometimes be avoided by removing the electronic amplification in favor of measuring a larger atomic signal in the first place [128]. For example, in our experiment, we could increase the total atomic flux to cause a larger response on the photodiode or design specialized optics to collect a larger solid-angle of the atomic fluorescence. Of course, improving the quality of electronic processing components can also reduce the white noise they produce. Sometimes white PM can also be reduced by filtering the output frequency in a narrow range. White PM is additive throughout the electronic processing segments, and can become significant. As was the case with flicker PM noise, white PM noise is not usually a limiting factor in modern atomic clocks, though it can pose problems for quartz oscillators, time and frequency dissemination systems, and telecommunication networks. Though white PM noise can be averaged away over time, it can limit frequency stabilization on timescales of order the clock's internal cycle time (as opposed to the frequency measurement interval) and beyond. An example of white PM noise and its Allan deviation is given in Figure 2.1.5.

Figure 2.5: Top: Simulated white phase modulated frequency data series in time. Bottom: Histogram of the measured frequency spread to demonstrate its shape. This figure is reprinted with permission of Alex Rollings[221].

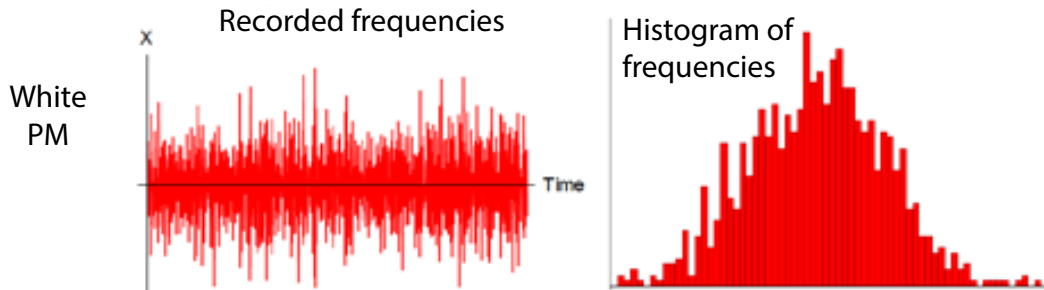
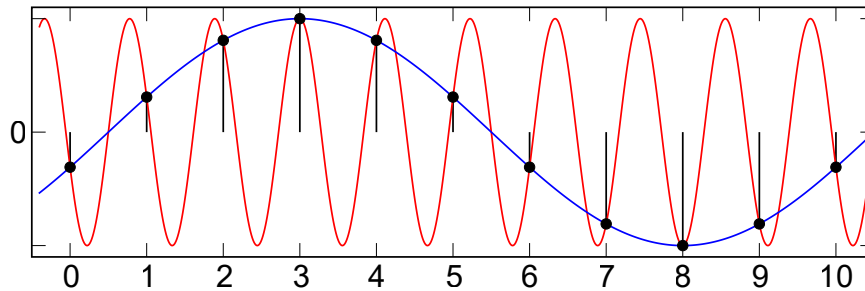


Figure 2.6: Periodic measurement of an oscillating signal at a frequency less than twice the signal's frequency is considered unsampling and results in aliasing of the oscillatory signal to a lower frequency as depicted for measurements 1 thru 10. Figure reprinted.³



2.1.6 Unwanted periodic fluctuations

Noise sources that oscillate at a relatively fixed frequency can sometimes cause strange results in an Allan deviation. Any periodic fluctuation with a frequency above $1/(2T_c)$ will be difficult to see through the clock sampling noise. Given that our calcium clock cycle times can be adjusted between about 1 to 40 ms, the system will have sampling rates of 1 kHz and 25 Hz, respectively. According to Nyquist sampling theory, these clock cycle times then under-sample frequencies more than about 500 Hz and 12.5 Hz, respectively, which leaves a lot of room for oscillating signals to discretely contaminate the atomic signal. In general, we tend to worry about noise from DC to around 10 kHz. Signals oscillating much faster than 10 kHz generally average away over the measurements and tend to be small, but they can still cause unforeseen issues through aliasing. Frequency aliasing is the process by which under-sampled oscillations are converted to lower frequencies based on the sampling rate and their initial frequency, like seen in Fig. 2.1.6.

Unwanted oscillatory signals can easily worm their way into an atomic clock's intended signal if the experiment and sampling electronics are not well designed. Pickup and ground loops are common and frustrating issues in electric signals carrying the measurement information. Pickup can be caused by proximity to other electromagnetic fields, though sometimes it can be reduced through use of ferrite cores or beads near the ends of the cables. Electrically conductive cables are basically large antennas, and the ferrite elements impede high-frequency electromagnetic waves

³ Original graph by Moxfyre - Own work, CC BY-SA 3.0, <https://commons.wikimedia.org/w/index.php?curid=6596059>

to act as a low-pass filter. They also reduce the ability of the source cable to generate unwanted electromagnetic interference that could be picked up by other circuits.

One common example stems from line noise at 60 Hz AC. This can sometimes contaminate the electronics of an experiment, causing characteristic peaks at 60 Hz and higher harmonics typically with decreasing power. Other periodic noise structures can contaminate the atomic signal, such as through ground loops, pickup, and room lighting reaching a detector. Periodic noise gets aliased down through the clock sampling cycle, sometimes also causing periodic structures in the Allan deviation as a function of the averaging time. Other times this noise appears on Allan deviations as broad lumps or single bumps diverting from the general trend. Generally speaking, abnormal bumps in an Allan deviation hint at a process with a period that is twice that of the averaging time at which the bump appears on the ADEV.

Some troublesome periodic fluctuations can be avoided through adjusting the data sampling procedure. If, for example, the laboratory pressure and temperature fluctuate regularly as part of the building's daily maintenance cycle, sampling at the same time of day may hide variations due to these environmental perturbations. For faster fluctuations, adjusting the sample time can sometimes solve the problem. If the inverse clock cycle time ($1/T_c$), is far less than the noisy oscillation, then small changes in the cycle time that do not greatly impact the atomic end can still greatly impact the measured frequency fluctuations. This case can be assessed by measuring the system instability over a range of cycle times and looking for odd structures. Of course, if the unwanted frequency contamination is fast enough compared to the cycle time, it may be simply easier to filter it.

Because the measurement (sample) time impacts the frequency aliasing process, variations of this time interval can mask frequency aliasing. This issue appeared prominently in the calcium sampling electronics, wherein a ground loop caused high frequency noise in a gated integrator, which accumulated the photodetector response for a fixed period of time. However, the timing protocols for the measurement system allowed variations in the cycle time such that noise was not initially aliased down to a single frequency, but rather a range of frequencies dependent on the

timing variation. Though at first the timing jitter was assumed to be insignificant, my reduction of the timing jitter by a factor of ≈ 100 caused the noise aliasing to become clear. The processed photodiode signal became visibly periodic, and the aliased frequency clearly changed with the integration period. However, because the high frequency noise was added within a closed circuit and integrated before output, the noise could not be filtered externally, and the circuit requires redesign and better power source isolation to improve its performance.

The general lesson here, which applies to all types of noise, is that there are various bandwidths and sampling times in the system of which you should be aware and circumspect. Solid knowledge of the characteristic frequencies utilized in the system and careful testing of all electronics - both servos and their power sources - is important to create a high functioning atomic clock.

2.1.7 Environmental sensitivity

Environmental sensitivity is one of the most deleterious sources of frequency instability that tends to dominate most every clock if given enough time or sensitivity to environmental contamination. Some environmental impacts, like temperature, tend to couple readily to most every clock at some level. If temperature does not couple either mechanically into the resonance or cause shifts from increased atomic motion, it surely at least limits the range of possible operating conditions before the materials comprising the experiment become unstable.

Background gases can be a troublesome long-term limitation for clocks of all types, and often their impact is very specific to the system in question. For instance, Micro-Electro-Mechanical Systems (MEMS) oscillators demonstrated extreme sensitivity to helium in a widespread internet mystery of 2018, when many cellular phones of employees working near a helium-leaking MRI machine mysteriously malfunctioned en masse as their MEMS devices were upset by helium seeping through their silicon structure [266]. Similarly, hydrogen gas is especially troublesome for aluminum ion clocks, as it is almost impossible to keep out, even within high-quality vacuum chambers. In this case, the hydrogen gas readily bonds with the aluminum ion, creating aluminum hydride and

disrupting the clock [56]. In the calcium system presented here, background gases are most notable for their contribution to phase noise along the interrogation laser's path in air [35]. Here, reducing the open-to-atmosphere optical path length and enclosing the path in hermetically sealed containers served to greatly reduce this impact.

Vibrations, usually caused by either acoustic coupling or physical shaking, can also severely limit an oscillator's stability. Typically, the vibrational sensitivity of a system is strongly correlated with the acceleration sensitivity, as is often characterized in novel Fabry-Perot cavity geometries. Though extreme measures can be taken in laboratory environments to isolate clocks and oscillators from vibrations and acoustics, often these efforts do not translate well into real-world environments, such as on airplanes or vehicles in remote locations without sufficient power resources. Vibrations and accelerations cause a variety of problems for atomic clocks, as these effects can couple fairly directly into the temperature or Doppler shifts of the ensemble. In ion traps, the problem of micro-motion is tangentially related to vibrations, and causes many of the same problems that vibrations can incite in other mechanical and atomic systems.

Vibrations and acceleration sensitivities can sometimes be mitigated through thoughtful design of the experiment, such as by the addition of dampening elements or natural resonance nodes in the structure. Often the design is aided through the use of finite element analysis (FEA), such as provided in software by COMSOL and others. Advanced cavity geometries have especially taken advantage of this method with great success, as fabrication methods for cavities and their common materials have advanced incredibly in the last twenty to thirty years [58]. One concern for vibrations in advanced cryogenic atomic clocks comes from the vacuum pressure maintenance systems, where even the bubbling off of helium from closed loop cryostats can pose serious concern [12]. Refrigeration systems for optics needing temperature control can cause similar problems. Cooling fans, ambient noises, doors slamming, water cooling, footsteps outside, and nearby construction can all contribute to unwanted atomic clock noise.

Of course, there is a rather infinite pool of possible environmental contamination sources. In addition to temperature, vibrations, and background gases, other sources to consider include am-

bient pressure, humidity, room lights, air currents, Earth’s rotation, gravitational fields, Bluetooth interference, wifi signals, power supply ripple, and noisy reference voltages, all of which can all cause frequency shifts of fluctuations that may or may not impede performance at the desired instability level. Fortunately, more advanced noise analysis such as the Allan deviation can help somewhat in tracking down and then mitigating many of the relevant noise sources while also characterizing the resulting calcium system frequency instability.

2.2 The Allan deviation

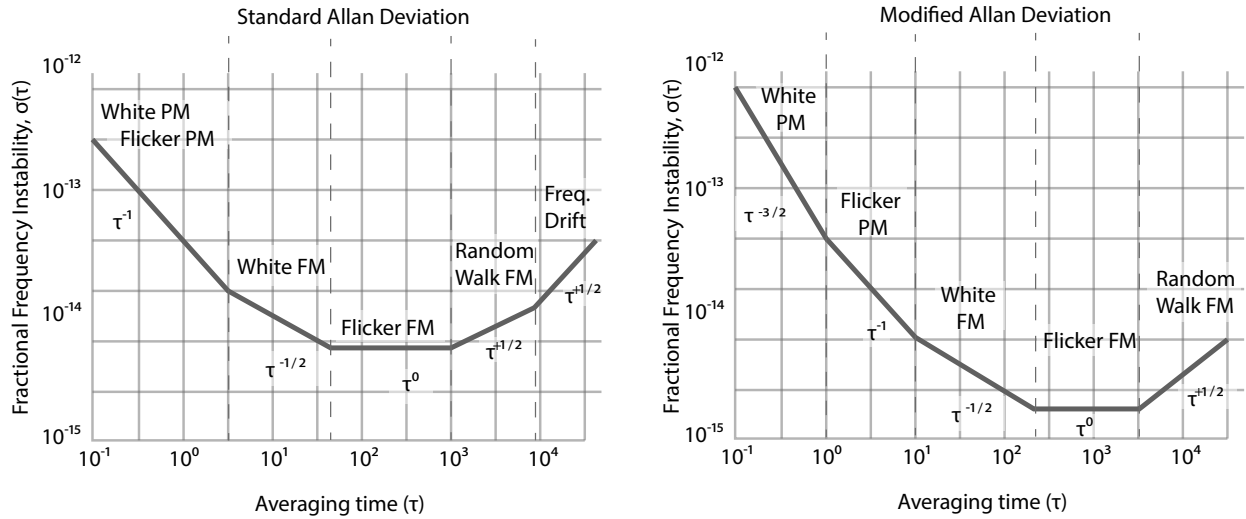
Assessing and reducing noise processes is a cornerstone of experimental atomic clock work. This task was made easier in the 1960’s by the pioneering work of David W. Allan on a two-sample variance, later known more commonly as the Allan variance. His work, in conjunction with later colleagues like David Howe, J.A. Barnes, and many others, led to the development of multiple frequency instability metrics for analysis and comparison. The Allan deviation and its offshoots now form the basis of atomic clock stability assessments and are recommended by the Institute of Electrical and Electronics Engineers (IEEE) and numerous other scientific standards agencies.

The Allan deviation was an important development for atomic clocks because typical single-sample methods of error analysis, such as the standard deviation, do not properly handle the types of atomic clock noise commonly seen, such as non-white processes including frequency modulation (FM) and phase modulation (PM). The standard deviation can become divergent when evaluating atomic clock frequency measurement data or develop false dependencies on uncorrelated variables, such as the number of frequency measurements total. Because atomic clocks often simultaneously exhibit many different noise types, including flicker, random walk, and oscillatory noises, a new method for assessing frequency stability was developed in the 1960’s called the Allan deviation. This method is quite suitable for frequency instability assessment, as it is able to handle many types of noise via a two-sample measurement that compares the frequency variations across a range of timescales.

Conveniently, the Allan deviation can differentiate between most of the dominant noise pro-

cesses seen in atomic clocks, which manifest as different slopes on the standard log-log Allan deviation plot. For the standard Allan deviation, only white and flicker phase modulation (PM) result in equivalent slopes, but a variant called the modified Allan deviation biases these two noise types differently, making them distinguishable by slope. An example Allan and modified deviation are given with the dominant noise processes labeled in Fig. 2.7.

Figure 2.7: The standard and modified Allan deviations are presented in typical log-log format, with dominant noise processes labeled for each discrete time interval. The modified Allan deviation can differentiate between white and flicker phase modulation (PM), while the two deviations agree for the frequency modulation (FM) noise processes.



Though only the standard, modified, and total Allan deviations will be covered in any sort of depth in this dissertation, there are a myriad of Allan deviation types, each with their own advantages and drawbacks. Most often seen in the general realm of stability analysis is the overlapping Allan, which is widely used in the field. Other types include the time, Hadamard, overlapping Hadamard, and thêo Allan deviations, most of which can also be extended to a "total" version, e.g. the Hadamard total Allan deviation. All types of Allan deviations adhere to certain rules: all involve differences between adjacent data groupings (i.e. are two-sample measurements) and are typically given by the deviation instead of the variance, and all converge for most clock noise processes.

Additionally, analysis of noise processes can proceed from either a frequency or a phase measurement standpoint. All information presented here will be aimed at addressing frequency data rather than phase data, but phase data methods are also well documented in a variety of resources.

2.2.1 The standard Allan deviation

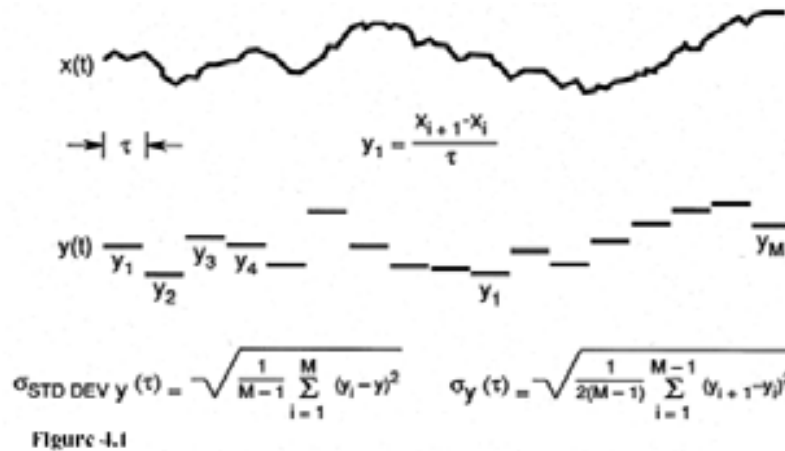
The standard Allan variance and deviation provide a metric for evaluating a frequency's stability over various timescales. Quite a few varieties of Allan types exist, and each has its own standardized procedure and well-studied properties. The Allan deviation is traditionally displayed as a log-log plot with α indicating the slope, which provides information about the dominant noise process. Because the Allan deviation plots the frequency instability, smaller values indicate less unstable, thus more stable, performance.

The Allan variance is defined as “one half of the time average of the squares of the differences between successive readings of the frequency deviation sampled over the sampling period.” Less succinctly, frequency data is collected in a time domain array $\{x_i\}$, with the data points understood to be separated by in time by the measurement interval given by T . The Allan variance divides the time domain data into M equal length segments of duration τ , where τ is usually called the sampling or averaging time. Each of the M segments is then averaged to generate a new array of these averages, $\{y_i\}$. The difference between adjacent elements of $\{\bar{x}_i(t)\}$ is then calculated. At some point before the final Allan deviation results, the frequency data needs to be normalized, though this can be done at any point during the process for the most part. The Allan deviation for a finite dataset is then given by the squared difference between adjacent averaged segments as

$$\sigma_y(\tau) = \left[\frac{1}{2(M-1)} \sum_{i=1}^{M-1} (y_{i+1} - y_i)^2 \right]^{1/2}. \quad (2.1)$$

For a more visual learner, the Allan deviation method can also be easily displayed as in Figure 2.8.

Figure 2.8: Visual representation of the Allan deviation for a time series of frequency data, $x(t)$, which is averaged over times τ and then used to calculate the frequency instability over a range of times with the equation in the bottom right. Figure reprinted with permission from the NIST Time and Frequency online reference from D.A. Howe, D.W. Allan, and J.A. Barnes [127]



Though the standard Allan deviation vastly improved frequency stability analysis starting in the 1960's, later iterations have come to replace the standard Allan deviation in most fields of research. The more used overlapping Allan deviation reduces the variability of the calculated instability values, providing more reliable analysis. The overlapping Allan does much of what its name suggests - it utilizes overlapping frequency data segments for the averaging and comparison process. Though the overlapping Allan can introduce unwanted correlations between otherwise statistically independent segments, it does overall increase the resulting instability measurement confidence. Comparisons of the variability in the overlapping versus standard Allan deviations can be seen later in Section 2.2.4.

2.2.2 The modified Allan deviation

The other main type of Allan deviation used in atomic clock systems is typically that of the modified Allan deviation, which can differentiate between white and flicker PM noise processes. Because atomic clocks and frequency stabilization systems, including cavities, can possess dominant white and flicker PM noise processes over differing timescales, the modified ADEV is often employed. Since its development in 1981, the modified ADEV has become one common form in which

to present Fabry-Perot cavity stabilization measurements. Because the modified Allan deviation possesses a τ^{-1} dependency for flicker PM over the $\tau^{-3/2}$ for white FM, frequency measurements with significant white PM noise will be seen to average down faster for the modified ADEV. Thus, the ultimate stabilities seen for sources with both of these noise types will result in a more favorable modified ADEV over the standard or overlapping ADEV. This can make comparisons of disparate frequency sources' instabilities difficult, especially when directly comparing published results from journals as in Section 2.2.5.

The modified Allan deviation [165] differs from the standard process in that it includes an additional averaging step, which essentially acts as a bandwidth filter. As the averaging time, τ , is stepped, the measurement's bandwidth is artificially adjusted. Because flicker PM falls off linearly with the frequency content while white PM is independent of it, these two noise sources appear with differing slopes on the resulting modified Allan deviation.

Mathematically, for M averaged frequency measurements segmented by averaging time $\tau = m\tau_0$, with τ_0 being the base measurement interval, the modified Allan deviation is given by either of the two following, equivalent expressions.

$$\text{Mod } \sigma_y(\tau) = \left[\frac{1}{2m^4(N-3m+1)} \sum_{j=1}^{M-3m+2} \left\{ \sum_{i=j}^{j+m-1} \left(\sum_{k=i}^{i+m-1} [y_{k+m} - y_k] \right) \right\} \right]^{1/2}. \quad (2.2)$$

$$\text{Mod } \sigma_y(\tau) = \left[\frac{1}{2} \left\langle \frac{1}{n} \left[\sum_{n=1}^n (\bar{y}_{i+2} - \bar{y}_i) \right]^2 \right\rangle \right]^{1/2} \quad (2.3)$$

The modified and standard Allan deviation should always agree for the first data point, i.e. for $\tau = \tau_0$ both deviations obtain equal fractional frequency instability values. This is not true for the total versions however.

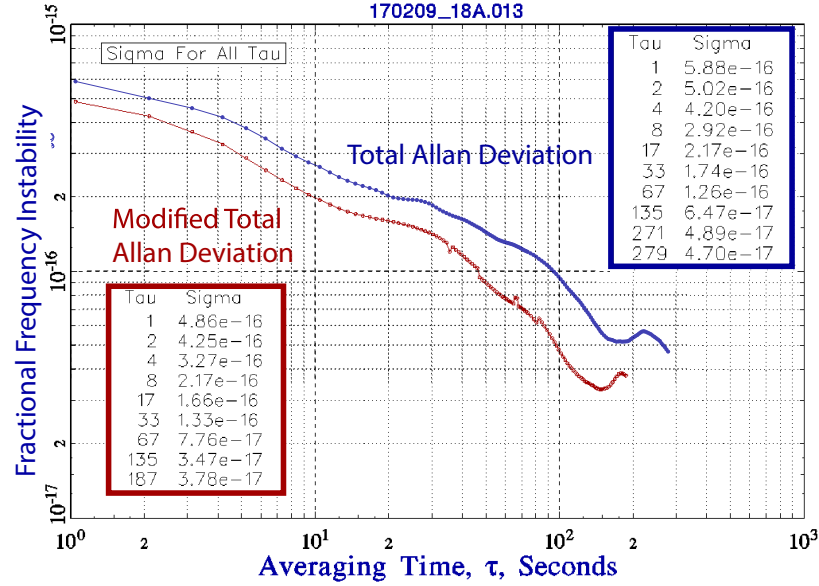
2.2.3 The total Allan deviation

The total Allan deviation method (as applied to the various types of Allan deviations) is used to increase the certainty of the calculated ADEV values over longer averaging periods (τ) than are

directly available with the standard ADEV methods. This could be done by simply duplicating the existing frequency measurement data and appending it to the end of the original array. However, this can cause a variety of issues from the discontinuity, inherent symmetry, or changes in drift. The method for calculating a total Allan deviation was standardized in the literature in the late 1990's [101, 130].

Meanwhile, the modified total Allan deviation possesses some stark contrasts to the total Allan deviation and even the modified Allan deviation. This is because of the averaging processes, which can often inappropriately bias instability data to appear better or worse than would be concluded using other Allan deviation methods[131]. The modified total Allan deviation differs from the Total Allan deviation in how the data set is extended. These quirks can cause the Allan deviation at the measurement cycle time, $\sigma_y(\tau_0)$ to differ from the standard, modified, or total standard Allan deviations. Sometimes the modified total ADEV can lead to a significantly improved data set, though it may not be considered "fair" to directly compare it to standard, overlapping, or total standard Allan deviations. An example of this in our own data is given in the following section on comparing different types of Allan deviations.

Figure 2.9: Significant differences in the processing of the total and the modified total Allan deviations results in their disparate values, even at the shortest measurement interval (in this case the initial data point). Data plotted with Stable32.



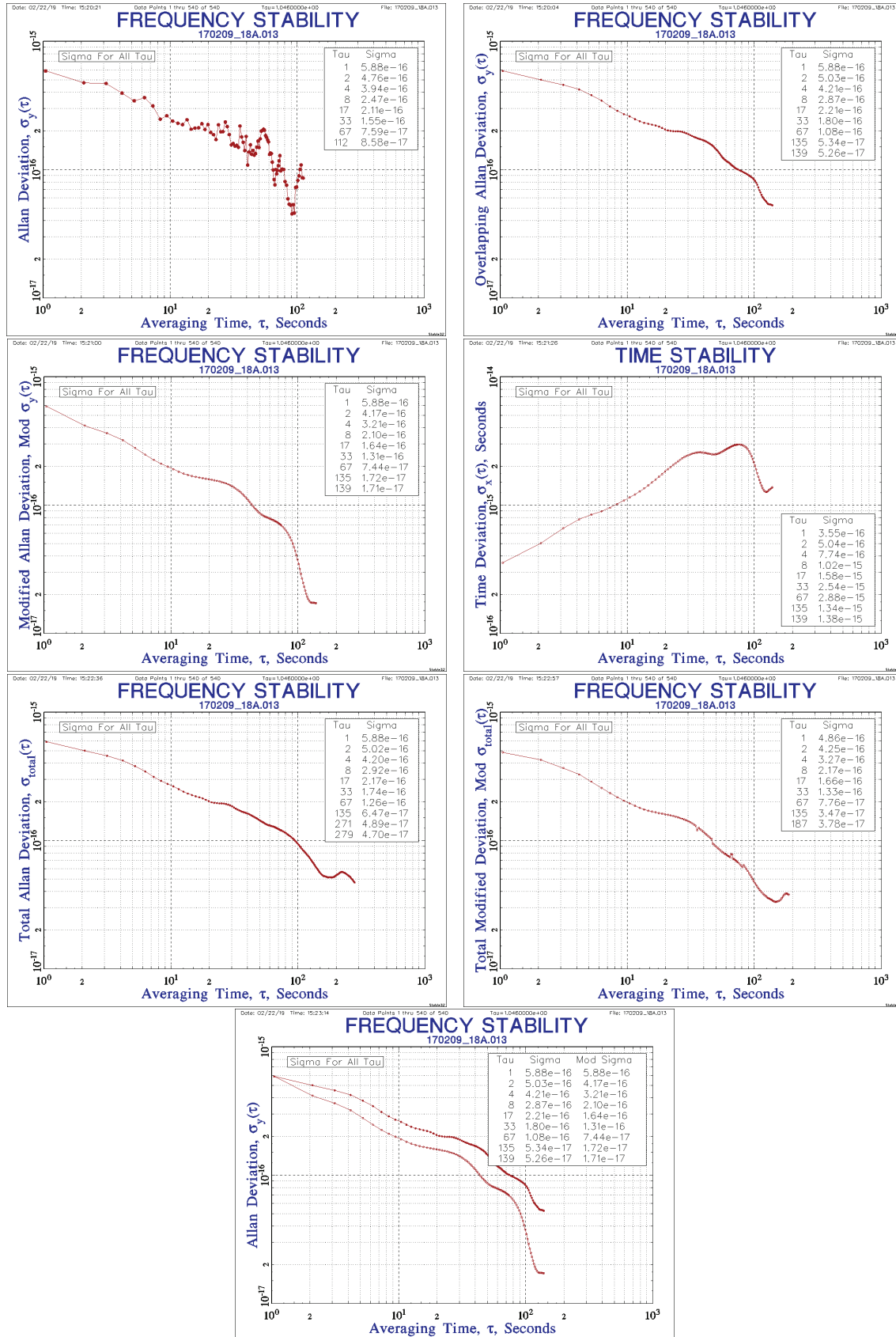
2.2.4 Comparing Allan deviation types

The Allan deviation methods presented previously are not without their drawbacks, one of which comes in the form of biasing the noise inappropriately, thus causing different types of Allan deviations to sometimes disagree in their instability results by significant amounts. However, so long as the same Allan deviation is used in comparing data, this should not be an issue when merely attempting to improve the stability of a frequency reference over the desired timescales. However, this issue does cause concern when attempting to compare vastly different frequency references relying on different types of Allan deviations. In particular, assessment of cavity instabilities tend to be depicted with modified Allan deviation plots, but atomic systems are more commonly given by the overlapping or total Allan deviation. As cavities often possess significant white PM, this often causes their modified ADEV values to appear lower than if they were analyzed with overlapping or total Allan deviations.

Because not all Allan deviations handle noise the same way, often comparisons of system instabilities must be taken with a grain of salt. The different types of dominant noise processes

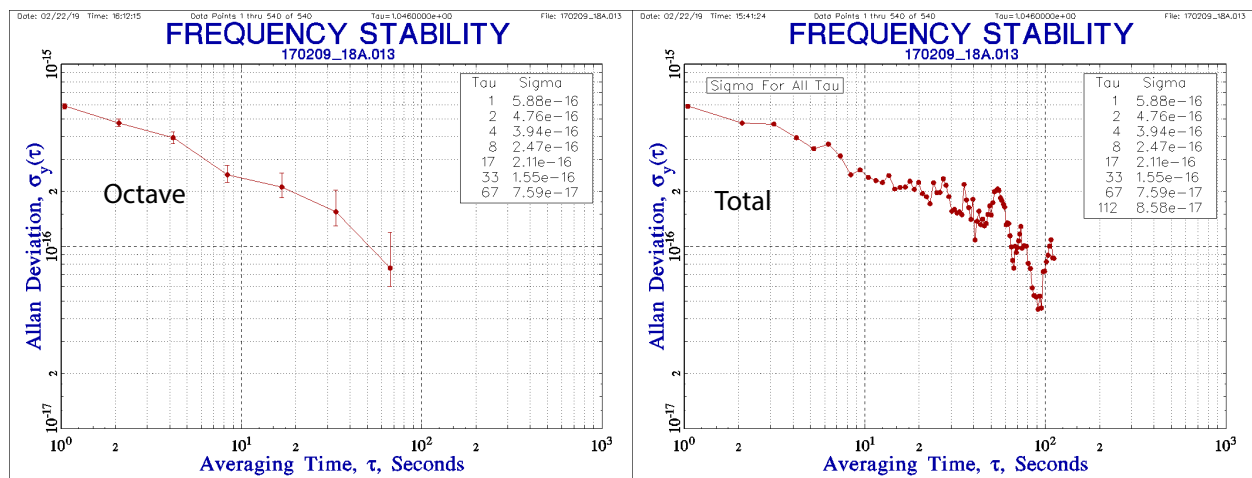
over the various averaging times can result in serious discrepancies between data sets that might appear equivalent if analyzed in a slightly different way. Choice in which Allan deviations to present in conferences, papers, or to quantify a system's performance can play a large role in the world of frequency stability, where sometimes improvement by a factor of only two may represent important and hard-won achievement by a team of researchers. To better understand the potential variations that can be seen with Allan deviations, the same 540 data points from a calcium stabilized frequency measurement are plotted for the major types of Allan deviations using the program Stable 32 in Figure 2.10.

Figure 2.10: Here, an identical data set is processed in the commonly used program Stable 32 to display the differing frequency and time stability interpretations dependent on the type of Allan deviation used. The bottom graph displays the stark contrast sometimes seen between more traditional Allan deviations (in this case the overlapping ADEV) and the modified ADEV.



Lastly, the density of data points on an Allan deviation can greatly change how it appears. Often calculating a dense ADEV and its error bars can be taxing on both the analysis program and the viewer of the resulting graph. Especially for processes without much inherent averaging, such as the standard ADEV, the total time choice can be quite chaotic looking (as in Figure 2.10.) Often both the octave and decade options are both used and displayed as a cohesive data set to provide more data points with error bars than is available with either on its own. To demonstrate how the temporal choice can impact the resulting visual data representation, the same data set from Figure 2.10 is again used, but this time displayed as a standard Allan deviation for all three temporal types in Figure 2.11.

Figure 2.11: Here, the same data is analyzed for a standard Allan deviation with the octave and total time binning choices from the instability analysis program Stable32. The choice in data density of the communicated Allan deviation chart can impact how the data is perceived, but must be weighed against the processing time it takes to generate the plots, especially for very long data sets.



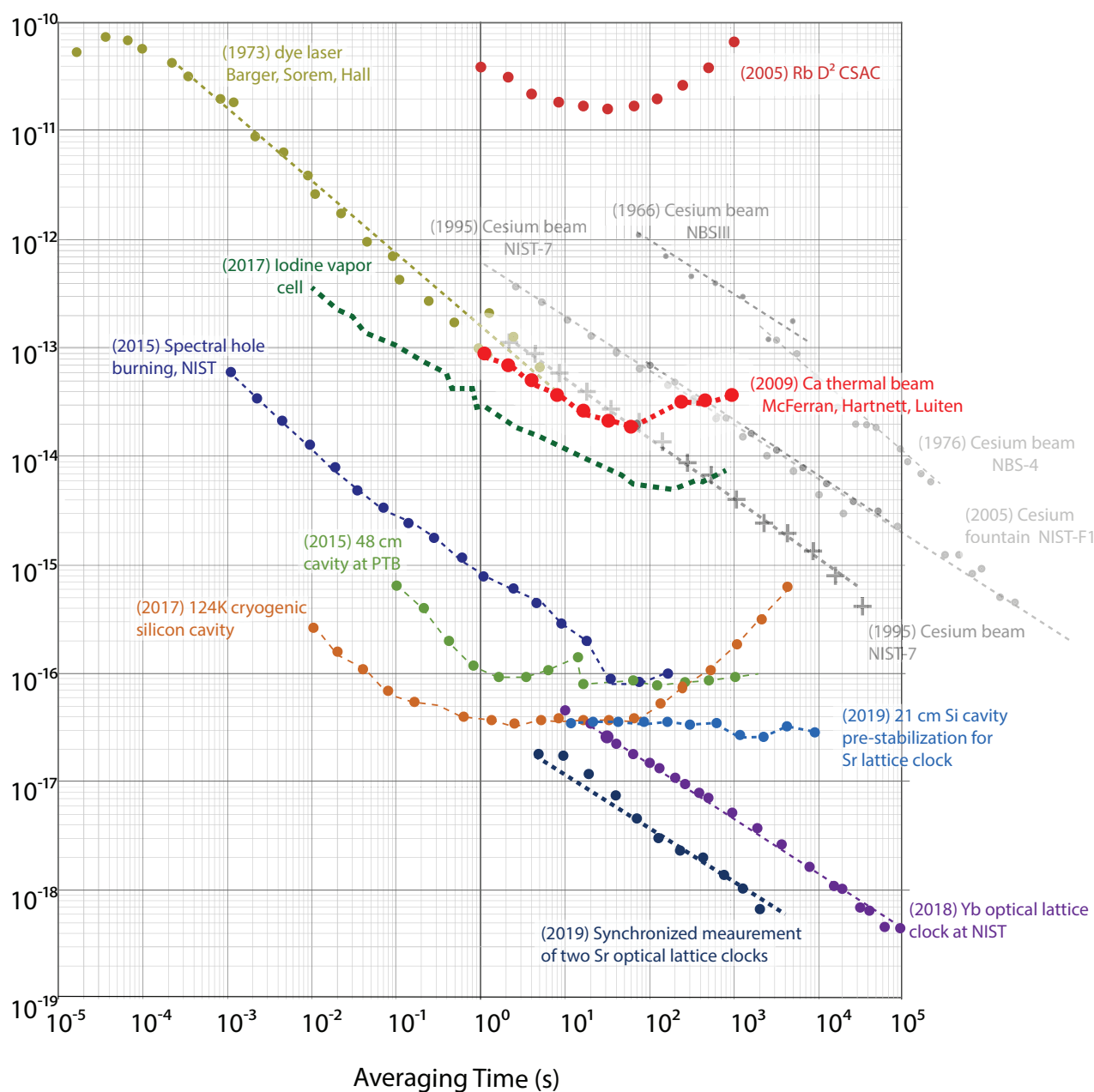
2.2.5 Collected Allan deviations of various frequency references and clocks

When researching various frequency stabilities, I found it very frustrating that there was no single chart I could find that let me see obvious comparisons of important stability measurements throughout history. Figure 2.12 below is my attempt to fill that gap, and some notes on the included Allan deviation traces. As previously mentioned, such data sets can be problematic for comparisons

- they are of varying types of Allan deviations, and some represent the single best data set ever collected from a stable frequency source, while others were intended by the authors to represent average performance exhibited by the system over much longer periods of time. The original source material should always be consulted before taking at face value any of the ADEVs presented in the summary chart to follow. They are collected to present a more complete historical overview of the frequency references that have made significant impacts or pose promising new avenues for future frequency stabilization.

Figure 2.12: Collected instability records of note from various journal publications, as cited in the following paragraphs. The Allan deviations here are not entirely compatible, as they vary in their Allan deviation type, including some that are standard, overlapping, modified, or total Allan deviations. *Fractional frequency instability in this chart should thus be taken with the understanding that the given data sets may not be perfectly comparable, and any further conclusions or comparisons of the collected results should be based upon the original publications (cited in this section).

Fractional
Frequency
Instability*



Notes on each of the Figure 2.12 instabilities presented:

- 1966 - NBS III - This system was the United States Frequency Standard at the time of measurement. It was a cesium atomic beam clock with reported performance mostly in the 10^{-13} decade between 100 seconds and nearly 10,000 seconds of averaging [256].
- 1973 - A continuous wave (CW) dye laser was stabilized to a high-finesse Fabry-Perot cavity by Barger, Sorem, and Hall [19]. It had drifts nearing 1.5 MHz/min. On short times of order 20 μ s it had residual fluctuations less 50 kHz, and after about 10 seconds could reach 100 Hz.
- 1976 NBS-4 - The data here is from the NBS-4 cesium primary frequency standard measurement [267], but the article also includes updates on the NBS-6 model also running at the time. Due to experimental apparatus design considerations, NBS-4 was more utilized for its long-term stability, thus used for maintaining the timescale, while NBS-6 (which was built from the NBS-5 base) was more prized for its contributions to frequency calibrations.
- 1995 NIST-7 - This primary frequency standard possessed an impressive accuracy for its time that falls below a fractional frequency instability of 10^{-14} with only a few days of continuous averaging [160].
- 2005 - NIST-F1 - The NIST-F1 primary frequency standard was the first laser-cooled Cs fountain standard in the United States. The improved accuracy from the cooled atoms reached below 1.0×10^{-15} [115].
- 2005 Rb D² CSAC - This system is a microfabricated physics package from the NIST ADI group (S. Knappe, P.D.D. Schwindt, V. Shah, L. Hollberg, J. Kitching, L. Liew, and J. Moreland). Though its instability was still far greater than the most advanced systems of its time, it's extremely small size gives it potential for inclusion in cellular phones, laptops, satellites, and various other real-world systems [154]. More advanced versions of the CSAC have been developed since and this technology is now being manufactured by industry.

- 2009 - Calcium thermal beam - This calcium thermal beam Ramsey-Bordé system is the closest system comparable to our own in experimental design [180]. Like ours, it utilizes a calcium thermal beam and the Ramsey-Bordé interferometry, and, for its time, achieved comparable or better performance than the existing cesium beam standards.
- 2015 spectral hole burning - Spectral hole burning is another alternative optical frequency reference receiving more attention in recent years. Two doped crystal samples undergo interleaved interrogation, wherein the interrogation laser causes saturation of a narrow spectral resonance. Once saturated, the resonance appears as a "hole" in the crystal spectrum, which can then be used to provide frequency corrections. This work was carried out at NIST by Cook, Shon and Rosenband, Till and Leibbrandt, David R [59].
- 2015 48 cm room temperature cavity at PTB - These Fabry-Perot cavity measurements represent some of the best instability numbers seen by a frequency pre-stabilization system to-date. The cavity length nears on the edge of what it capable with conventional machining techniques. The relatively long cavity length enables an extremely high quality factor, enabling one of the first cavity measurements with instability below 10^{-16} [107].
- 2017 - Iodine vapor cell - This iodine vapor cell was designed with special intent for use in space applications. It relies on a comparatively complicated beam path through the vapor relative to more standard single or double-pass geometries. It is one of the most stable vapor cell references studied thus far [200].
- 2017 - 124 K Cryogenic silicon cavity - In a collaboration between PTB and JILA/NIST, researchers have been exploring Fabry-Perot cavity development from specialized materials (in this case silicon, though sapphire is another popular option) held at low temperatures to reduce the thermal noise contribution to the instability. This measurement showed that the resulting stability was able to reach the predicted thermal-noise floor of the system design [179].

- 2018 - Yb optical lattice clock at NIST - This lattice clock has achieved the most stable frequency measurement ever taken to date [181]. However, this excellent performance required five independently variable laser systems, extreme vacuum requirements, and extensive reduction of environmental impacts through system isolation. Though this system may not translate such performance out of the laboratory, efforts are being made to develop a transportable version of the system that may one day reach similar performance levels. I also must mention that the Yb system has obtained data very similar to the Sr data also presented here. Especially for the zero-dead-time optical lattice configuration, similar instability was obtained but on shorter timescales. However, this would have overlapped with the Sr data line and made for a less interesting plot.
- 2019 - Sr optical lattice clock and cavity pre-stabilization - ArXiv pre-print results from the JILA strontium lattice clock team shows their latest instability progress with both Fabry-Perot cavity pre-stabilization and optical lattice clock frequency corrections. In this work, pre-stabilization to a specialized 21 cm silicon crystal Fabry-Perot cavity represents some of the most advanced cavity reported to date. It's performance is largely what enables the correspondingly impressive optical lattice clock stability on such timescales. The optical lattice clock ADEV is generated from frequency stabilization provided by two synchronized strontium lattice clocks, one with a 1D configuration while the other was a 3D lattice. Much like the Yb lattice clock data (2018) also in the chart, this performance requires a relatively complicated and involved setup compared to most other technologies presented.

The inclusion of the material in this section so early in the thesis highlights two aspects that are critical for the rest of this work. First, it presents the clock-specific language used to quantify atomic clock performance. Second, it emphasizes that the concept of stability is only meaningful when one includes the timescale (or averaging time). Different types of timing and frequency devices have differing performance profiles - some offer better short term performance while others are built for long-term. This context is critical for understanding the the performance

of the calcium system presented here.

Chapter 3

Ramsey-Bordé spectroscopy

This chapter begins with the general historic background to provide motivation for the following theoretical chapters. Both the theory and history follow a similar story, from early probes of quantum mechanics with single-zone Rabi spectroscopy, leading to Ramsey's method of separated oscillatory fields (two-zone), and then later developments that led to multi-zone laser-atom interactions under the general term of optical Ramsey methods, which includes Ramsey-Bordé. These predecessors of the Bordé method provide great insight into challenges and advantages that face Bordé spectroscopy while giving a solid background in how laser-atom interactions have developed, including reinterpretation of the earlier 1984 proposal from Bordé, Salomon, Avrillier, Van Lerberghe, Brant, Bassi, and Scoles [41] as a matter-wave interferometer [38].

The spectroscopic tools employed in this dissertation are more complicated than what a graduate student might learn in typical quantum mechanics and even spectroscopy specific classes. Particularly, some of the common assumptions and approximations that apply for microwave or lower frequency fields no longer apply in the optical, and some typically ignored effects - like atomic recoil and Doppler effects, are crucial to the understanding of Ramsey-Bordé interferometry. This chapter will establish both a historical and theoretical context for the Ramsey-Bordé technique. Both follow the same general sequence of events - beginning with tests of microwave two-level atomic systems in early quantum mechanics through their eventual application to the optical domain. This path was heavily influenced by the technological limitations of the time, which were already introduced in the opening history section.

In particular, the history and theoretical basis will be presented with our ultimate experimental goals in mind. Ramsey-Bordé spectroscopy was chosen here for its ability make high signal-to-noise measurements of thermal atomic ensembles. The resulting spectroscopic features can possess linewidths below even the transition's natural linewidth. Because such a broad velocity atomic distribution can be used, the number of atoms contributing to each measurement ($\approx 10^9$ per 10 ms) is many orders of magnitude larger than what is obtainable in most other types of atomic sources for clocks, including optical lattices, single ions, vapor cells, or magneto-optical traps. Thermal beam experiments with Bordé-Ramsey fringes stand in contrast to the more generally followed research flow from the 90's to today in which laboratories tended toward cooled and trapped ensembles or narrow velocity selection. Such ensembles of atoms moving relatively uniformly reduced inhomogeneous motional shifts and broadening mechanisms, albeit with smaller numbers of atoms.

Ramsey-Bordé, as implemented here, benefits extraordinarily from its high atom number, as this, along with electron-shelving detection, allows signal photons to be extracted from nearly every atom of our ensemble. By being so efficient with signal collection and utilizing a kHz-level resonances, each measurement can be performed in a relatively short period of time ($\ll 1$ s). Throughout the chapter, key historic and theoretical developments will be related back to these desired properties of the thermal beam system used in the calcium interferometer. Though presented here to show their specific progress toward the final Ramsey-Bordé system we present. Of course, many of the systems we discuss are important and effective in their own right, and, though these will not necessarily be discussed here, the interested reader is encouraged to explore them through the given references and citations.

Furthermore, it does this with exceptional signal-to-noise.

Furthermore, this chapter makes clear how interwoven the physical theories are to the choice in ensemble and apparatus geometries. Thus, while a general understanding of the physics behind Ramsey-Bordé is useful, for the theory to be useful at these precision levels, it must be specifically tailored to an intended experimental configuration. So, while this chapter provides more generally

applicable physical theories to motivate how our Bordé apparatus works, subsequent chapters will include more details as they specifically apply to our apparatus.

3.1 Historical foundations of Ramsey-Bordé Spectroscopy

The pre-history of Ramsey-Bordé spectroscopy begins around the turn of the quantum revolution, when scientists first began to realize the wave-particle duality of matter. Some of the earliest and simplest experiments utilized a type of spectroscopy relying on a single atom-field interaction, often called Rabi spectroscopy after Isidor Isaac Rabi¹ (1898 - 1988). Rabi's contributions to science won him the 1944 Nobel prize in physics and now numerous awards bear his name.

The wave of single-zone atom-field interaction spectroscopy that followed in the wake of Rabi's seminal research was very promising. One important result from single-zone spectroscopy was the discovery of Rabi flopping, wherein a two-level system's wave function is seen to oscillate between the levels over time in the presence of a coupling field. These oscillations could be well-controlled, thus were useful for preparation and measurement of atomic states. Rabi flopping is still an important tool today, as it can be used for determining ensemble coherence and measuring external fields precisely [133, 148]. In essence, the early exploration of Rabi flopping led scientists to a method for reliably controlling or testing the state of atoms, molecules, and ions.

Though Rabi spectroscopy was an essential and important development in atomic physics, it has some key drawbacks. In addition to Doppler effects from the thermal atomic velocity distribution (which were especially severe before atomic cooling and trapping began in the 1980's), Rabi spectroscopy also suffers from time-of-flight broadening. In short, the energy-time uncertainty principle limits how well the atomic resonance can be probed when the atom-field interaction lasts a finite amount of time. Thus, it can be difficult to resolve narrow transitions with a single inter-

¹ Rabi refused to accept female students, discouraged hiring of female professors, and stood by the view that women were incapable of being worthwhile researchers [259]. Even into the 1980's, Rabi maintained these views in an interview, stating that that women are "temperamentally unsuited to science" and that, as compared to men in science, "It's simply different. It makes it difficult for them (women) to stay with the thing. I'm afraid theres no use quarreling with it, that's the way it is. **Women may go into science, and they will do well enough, but they will never do great science.**" [95] Unfortunately, Rabi's views were shared by much of the physics 'old guard' during his heyday, and even his statements in the 1980's were not considered controversial.

rogation field, even with a rather large apparatus. To be successful, electromagnetic fields had to be created that were quite large, which was not always feasible. Lastly, decoherence mechanisms usually find a way to creep into real world atomic systems, causing Rabi flopping to resemble a standard damped harmonic oscillator, which can severely impact the coherent Rabi flopping of the interrogated ensemble.

Many of these problems inherent to single-zone spectroscopy were overcome with the advent of two-zone experiments, pioneered by Rabi's student, Norman Ramsey. Now often referred to as Ramsey's method of 'separated oscillatory fields,' this technique no longer required large area electromagnetic interactions, and it furthermore allowed access to narrower spectroscopic resonances than possible with single-zone experiments. When Ramsey's method of separated oscillatory fields was first unveiled in 1949, microwave fields were still the most convenient electromagnetic field choice for exciting atomic transitions. Lasers weren't even invented until the 1960's, and for some time their utility was limited by the scant choice in wavelengths with narrow tunability and limited control. Also of note, atomic and molecular sources suitable for spectroscopy during this time were largely limited to thermal beams or vapors with substantial velocity distributions, thereby incurring large Doppler effects. However, even after developing laser stabilization techniques, applying the same two-zone framework on narrowband optical resonances was extremely problematic. New methods had to be developed to overcome dephasing of the ensemble-averaged wavefunction, which was far more severe at optical frequencies.

In the 1970's, a variety of theoretical physicists began to explore methods for rephasing the atom ensemble via additional laser-interactions and specialized two-photon transitions. Some of the important early theoretical papers began pushing toward a practical implementation of Ramsey fringes using frequencies much greater than those in the microwave domain [77, 14, 76, 113]. These theories first came to fruition with initial experimental demonstrations on sodium, neon, methane, SF_6 , and calcium in the second half of the decade. These earlier optical Ramsey fringe experiments were largely based on either three standing-wave laser interactions with single-photon transitions [13, 24, 39] or else two standing-wave laser-atom interactions relying on two-photon transitions

[249, 224, 159, 158].

Many experimental configurations (geometries) of lasers were suggested to perform multi-zone spectroscopy on thermal beams of atoms or on vapor cell ensembles. However, almost all configurations presented experimentalists with new challenges. First, generation of numerous spatially separated phase-coherent laser interactions generally requires long optical path lengths wherein the beam is reflected and folded back upon itself many times. Such a laser geometry is difficult to achieve without introducing optical aberrations or unwanted phase noise. Second, the laser beam alignment required for traveling-wave geometries utilized for optical Ramsey fringes needs extremely parallel beam paths, usually within a few micro-radians for resolving low-contrast fringes. Obtaining such parallelism between spatially disparate laser beams is experimentally challenging, even when utilizing the classic cat's eye lenses. Third, limitations of the laser linewidths available in the 1970's made resolution of relatively narrow optical Ramsey fringes difficult. In fact, cavity stabilization expertise from Jan Hall and later Jim Bergquist was central to the first demonstrations of optical Ramsey spectroscopy.² These earlier efforts provided the breakthrough necessary for he and Dick Barger, T.C. English, and D.J. Glaze to publish the first experimentally observed calcium optical Ramsey fringes, which occurred at NIST in 1979 [18], after years of attempts.

The optical-Ramsey fringe geometry used by the NIST calcium group in the late 1970's was not quite that of Ramsey-Bordé, but it was rather similar in its approach. As will become evident following sections, multiple approaches to optical Ramsey fringes have been demonstrated with success, and they can vary greatly depending on the experimental goal. Though these experiments may appear different, they all boil down to the same crucial elements - multiple coherent atom-field interactions separated in time and/or space to isolate a spectroscopic signal. These same elements are now ubiquitous throughout AMO physics for unprecedented control of both the quantum and macroscopic properties atoms and molecules. Ramsey and optical-Ramsey processes were part of a paradigm shift to segmented multi-step atomic state preparation, manipulation, and readout.

² The first two demonstrations of optical Ramsey fringes relied on optical domain single-photon transitions in methane and neon [25].

In this vein, additional processes were later developed, including cooling and trapping methods to first generate comparatively uniform ensembles. With the ability to generate very uniform or trapped ensembles, improved optical frequency standards focused on obtaining highly accurate measurements became popular and spatially-separated beam experiments fell out of favor. The number of thermal beam publications in this field has continually decreased over the last thirty years, with only a handful of papers being published in the last decade. However, thermal beams interrogated by spatially separated laser fields (such as utilized in this dissertation) remain powerful spectroscopic tools that have often been overlooked.

3.2 Theoretical basis

Ramsey-Bordé interferometry theory is rooted in fundamental quantum mechanics. This method requires four laser-atom interactions, which can be sequentially applied to the atomic state, including a dark-time evolution between atomic interactions. Before considering the full four zone Ramsey-Bordé method, we look at laser-atom interactions in a coupled two-level system. To eventually give a satisfying theoretical basis for Ramsey-Bordé interferometry, many of its building blocks will be discussed, including Rabi spectroscopy, Ramsey’s method of separated oscillatory fields, and examples of how Ramsey’s method is applied to the optical domain. Various tools for interpreting the quantum mechanical processes will be used where applicable, including the Bloch sphere picture. The ultimate solutions to the four laser-atom interactions of Ramsey-Bordé are quite complicated, and, thus, while not explicitly derived here, they are explained through the Bloch sphere. Additionally, the relevant portion of the 1984 paper by Bordé *et al* is included in the appendix of this dissertation, which does include a full derivation of the Bordé fringes. The paper’s derivation builds off the formalism established within this chapter.

3.2.1 Coupled two-level systems and Rabi flopping

Two-level systems are fundamental to a variety of quantum mechanical phenomena. They form the basis of theoretical discussion underlying Ramsey-Bordé interferometry and much of the

preceding work. Though the Ramsey-Bordé process technically involves many of possible atomic states ($2^4 = 16$ final states), the theoretical predictions of Ramsey-Bordé fringes can be carried out while only ever considering a single two-level atomic system at any given part of the calculation. As such, the basic two-state system will be briefly discussed, exploring how an oscillatory coupling between the two energy states impacts the quantum system, leading to a phenomenon known as Rabi flopping.

With Rabi flopping, an atom with two states, ground ($|g\rangle$) and excited ($|e\rangle$), develops a periodic probability dependence in time to be found in either state when in the presence of the field. In fact, this process can be quite well controlled, enabling precision measurements of fundamental constants and radiation frequencies. This phenomenon relies on a single field zone, and the atoms, ions, or molecules being used only interact with the field a once for a duration t .

Rabi spectroscopy is typically accomplished by applying an external radiation field, $\vec{E}$, that couples the otherwise isolated atomic energy levels. Coupling to an oscillating field introduces some slight changes to the earlier coupled two-state system model. In this case, the perturbation is dominated by the electric field given by $\mathbf{E} = \mathbf{E}_0 e^{i(\mathbf{k}\cdot\mathbf{x}-\omega t)}$ where $\mathbf{k}$ is the field's wavevector and $\mathbf{x}$ the position vector at the location being considered. The field's frequency remains somewhat near the resonance of the transition to and from the ground and excited states such that $\omega \approx \omega_0 = E_1 - E_2$. The Hamiltonian for this two-level system coupled by an oscillating electric field is then given by $\hat{V}' = e\mathbf{E}_0 \cdot \mathbf{x} \cos(\omega t)$.

The time-dependent atomic state of the total matrix $\hat{H} = \hat{H}_0 + \hat{V}'$ can be given by a generic normalized quantum state $|\psi(t)\rangle = c_1(t)e^{-iE_1t/\hbar}|\psi_1\rangle + c_2(t)e^{-iE_2t/\hbar}|\psi_2\rangle$. Then applying the Schrödinger equation to this generic state and taking the inner product with the either state yields two coupled differential equations given by

$$i\hbar\dot{c}_1(t) = c_1(t)\langle\psi_1|\hat{V}'|\psi_1\rangle + c_2(t)\langle\psi_1|\hat{V}'|\psi_2\rangle e^{-i\omega_0t} \quad (3.1)$$

$$i\hbar\dot{c}_2(t) = c_1(t)\langle\psi_1|\hat{V}'|\psi_2\rangle e^{+i\omega_0t} + c_2(t)\langle\psi_2|\hat{V}'|\psi_2\rangle, \quad (3.2)$$

and the energy difference between the energy levels is given by $E_1 - E_2 = \hbar\omega_0$. In the dipole

approximation limit assumed here, only non-diagonal elements of $\hat{V}'$ yield non-zero elements by parity). These non-zero elements are often rewritten in terms of the Rabi frequency, Ω_0 which characterizes the oscillation rate between the two states such that

$$\Omega_0 = \frac{e}{\hbar} \mathbf{E}_0 \cdot \langle \psi_1 | \mathbf{x} | \psi_2 \rangle \quad (3.3)$$

and the coupled equations then, with the cosine expanded into exponentials, yield the following:

$$\dot{c}_1 = -\frac{i\Omega_0}{2} c_2(t) (e^{+i(\omega-\omega_0)t} + e^{i(\omega+\omega_0)t}) \quad (3.4)$$

$$\dot{c}_2 = -\frac{i\Omega_0}{2} c_1(t) (e^{-i(\omega-\omega_0)t} + e^{i(\omega+\omega_0)t}). \quad (3.5)$$

At this point in the derivation, the commonly utilized rotating-wave approximation can greatly simplify the calculation by removing the $\omega + \omega_0$ terms. These terms are far larger than the difference terms, and thus oscillate very fast. In fact, these terms oscillate so quickly that they have very little impact, and, for the purposes here, is quite applicable. The coupled differential equations are now reduced to $\dot{c}_1(t) = -\frac{i\Omega_0}{2} e^{+i\Delta t} c_2(t)$ and $\dot{c}_2(t) = -\frac{i\Omega_0}{2} e^{-i\Delta t} c_1(t)$ where Δ is now the detuning of the field from the transition resonance, given by $\Delta = \omega - \omega_0$. The simplified differential equations can now be solved. For a system initially in the ground state $|\psi_1\rangle$, the coefficients can be found such that the probability of the atom being in the excited state at some later time t is given by

$$P(1 \rightarrow 2) = |c_2(t)|^2 = \frac{\Omega_0^2}{\sqrt{\Omega_0^2 + \Delta^2}} \sin^2 \left(\frac{\sqrt{\Omega_0^2 + \Delta^2}}{2} t \right), \quad (3.6)$$

which is often instead expressed in terms of the generalized Rabi frequency, $\Omega_G = \sqrt{\Omega_0^2 + \Delta^2}$. Thus, away from the transition resonance, atoms no longer switch between states at the Rabi frequency, but rather at the generalized Rabi frequency. Clearly, the generalized Rabi frequency must be less than the bare Rabi frequency for $\Delta \neq 0$, thus, further resonance, the atomic cycling between ground and excited states occurs at a faster rate and with smaller amplitude. Additionally, away from resonance the atom can never be perfectly transitioned to the excited state from the ground state, i.e. the probability amplitude for this case is less than unity.

While it may be feasible to provide perfectly resonant light to a single quantum absorber or even a very uniform ensemble of them, this cannot be realistically accomplished for an entire

thermal beam of atoms. This is because the resonance frequency of the atomic transition is dependent on a wide range of factors. The largest of these experienced by the thermal Ramsey-Bordé ensemble utilized in this dissertation is the first- and second-order Doppler shift. The large velocity distribution from the thermal calcium beam thus presents a challenging ensemble to probe with a single atom-field interaction without only selecting out a narrow portion of the population.

Additional restrictions apply for Rabi spectroscopy to be useful. The interrogated atoms must not spontaneously decay while being driven, or else the Rabi flopping loses coherence, i.e. the ensemble no longer ‘flops’ together. Thus, atomic energy levels chosen for these types of experiments often display reasonably long lifetimes. However, the atomic lifetime can easily dwarf the amount of time an atom remains within the electromagnetic field. Thermal atoms are still usually moving hundreds of m/s, and thus don’t have very long before either leaving the interrogation field, colliding with another atom or background gas particle, or hitting the end of the apparatus. For example, a 1 Hz linewidth atomic transition takes of order 1 s to resolve by the energy-time uncertainty principle. During this time, a thermal atom with a speed of only 100 m/s would require a 100 meter long apparatus to carry out such an experiment.³ . It is clear that these two experimental challenges - choice in atomic transition and suitable apparatus to probe it - were not trivial, especially with early thermal ensembles, which were the only available option for many decades.

Lastly, it is useful to consider how Rabi flopping might be useful for the determination of the driving field frequency. If all other pertinent experimental conditions are held fixed, then the state probability correlates well with the field frequency. To provide frequency discrimination, it is most effective then to probe the Rabi flopping on the side of the spectral feature, where the state probability versus frequency detuning function’s slope is maximal. This slope, when sampled in a reasonably narrow frequency range, provides an effectively linear response to the driving field frequency. Thus, monitoring changes of atomic populations after the Rabi flopping process provides

³ Some atomic collisions with physical walls can be rendered inconsequential with the use of paraffin wax or teflon. Teflon coatings in hydrogen maser chambers were found to increase the coherence times significantly over uncoated surfaces and even paraffin [23] This enables smaller apparatuses to be constructed in some instances, such as with hydrogen masers.

a relatively simple and straight-forward method for determining how much the driving field has shifted in frequency between measurements.

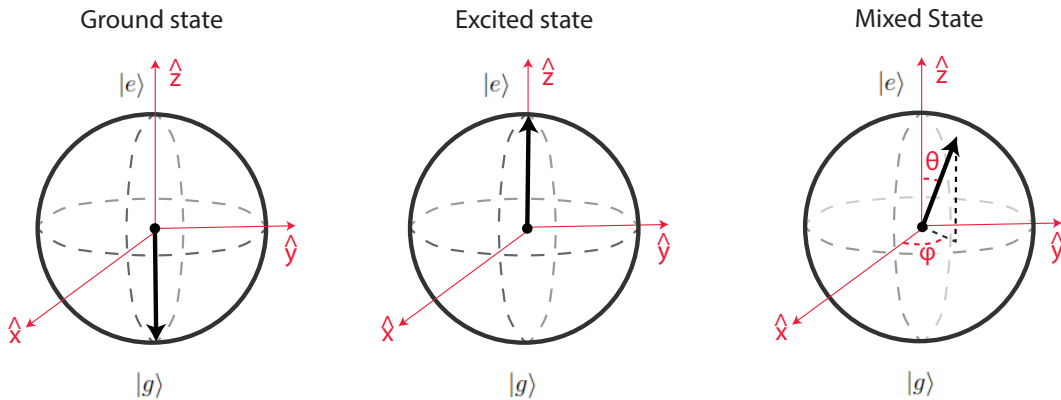
As the discussion of two-level atomic states in the presence of electromagnetic fields continues to develop beyond Rabi flopping, both the equations and the processes become more complex. Thus, it is prudent to now introduce the concept of the Bloch sphere, which is an invaluable tool for discussing and visualizing two-state system evolution.

3.2.2 The Bloch sphere and pulse area

The two-state system transfers easily to the Bloch sphere: a unit sphere helpful for tracking both the phase and population amplitude of a two-level atom through visual representation. The Bloch sphere formalism is especially useful for later understanding the more complicated Ramsey-Bordé process and demonstrating visually how the fringes are preserved despite large Doppler shifts. For the sake of this chapter, the Bloch sphere will exist in the rotating frame of the oscillating field that drives transitions. For a two-state system decomposed as in Equation 3.7, where $|g\rangle$ and $|e\rangle$ are the ground and excited states, respectively, various states on the Bloch sphere are depicted in Figure 3.1. An arbitrary superposition of ground $|g\rangle$ and excited $|e\rangle$ states of a an atom can be parameterized by polar and azimuthal angles on the Bloch sphere given by

$$|\Psi\rangle = \cos\left(\frac{\theta}{2}\right) |e\rangle + e^{i\phi} \sin\left(\frac{\theta}{2}\right) |g\rangle. \quad (3.7)$$

Figure 3.1: The standard Bloch sphere with examples of both pure states and one superposition state with a mixing angle θ .



The atomic phase is tracked by the angle ϕ while the mixing angle, θ , indicates the relative population between $|e\rangle$ and $|g\rangle$. Both the θ and ϕ angles are indicated in the mixed state of Figure 3.1.

Resonant atom-field interactions yield rotations of the atom's state vector on the Bloch sphere about the $\hat{x}$ axis. For off resonant excitation (coupling), the axis of rotation lies in the $\hat{x}$ - $\hat{z}$ plan such that an initially ground state atom cannot reach a pure excited state. The tilted axis can be thought of as the vector representation of the generalized Rabi frequency, $\vec{\Omega}_G$. Without the driving field (e.g. the dark time between field interactions), the atom's state evolution on the Bloch sphere is akin to rotations around the $\hat{z}$ axis at a rate proportional to the detuning. Thus, atomic states on the Bloch sphere remain in their superposition without coupling, i.e. atoms in mixed states trace out horizontal circles on the Bloch sphere in the absence of state coupling. The phase angle ϕ effectively tracks the relative phase between the two considered eigenstates. Of course, the atomic states can be decomposed into any valid basis not represented by the poles of the Bloch sphere, but this is akin to a simple rotation of the original coordinate axes.

As part of the Bloch sphere picture, pulse area plays an important role. The pulse area is derived from eqn. 3.6, given here in terms of the generalized Rabi frequency, Ω_G as

$$P(1 \rightarrow 2) = \frac{\Omega_0}{\Omega_G} \sin^2 \left(\frac{\Omega_G t}{2} \right), \quad (3.8)$$

where $\Omega_G t$ is the pulse area. For the case of resonant excitation, where the generalized and bare Rabi frequencies are equal, it is clear that, for a pulse area of π , the atom is fully transferred from the ground to excited state. Meanwhile, a pulse area of $\pi/2$ creates a superposition such that the atom is equally likely to be found in either state. Often these terms are expressed as simply a π -pulse and $\pi/2$ -pulse, respectively. Thus, a resonant $n\pi/2$ -pulse corresponds to a $n\pi/2$ rotation of the atom's Bloch state vector about the $\hat{x}$ axis. Any deviations from resonance tilt the rotation axis, causing, for example, a π -pulse to longer transfer the atom perfectly from the ground to excited state.

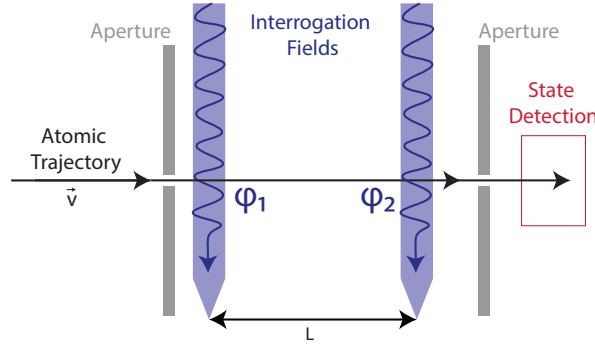
The Bloch sphere formalism allows for a simple and visual explanation of how the atomic

state evolves when undergoing more intensive processes beyond the single atom-field interaction. The following sections will largely rely on this formalism in their explanations, though citations will be given where appropriate to direct the reader to thorough mathematical derivations as well.

3.2.3 Ramsey fringes

Ramsey proposed a method of ‘separated oscillatory fields’ that revolutionized atomic physics. He realized that the single large interaction region could instead be replaced by two short interactions at the beginning and ends of the original extended interaction (Fig. 3.2). The previously problematic large area electromagnetic fields were supplanted by two reasonably small interaction zones but the total interaction time was able to be extended where beneficial.

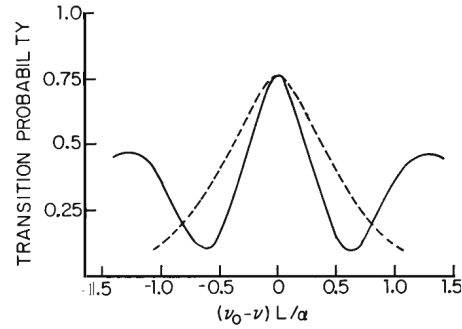
Figure 3.2: Ramsey’s method of separated oscillatory fields as utilized in an atomic beam experiment. (a) Atoms with velocity v pass through two electromagnetic fields separated by some distance L and then have their quantum state read. Apertures are often used to ensure that the $\vec{k} \cdot \vec{v}$ is small so most atoms are resonant with the laser beam after time-of-flight broadening. The field widths is exaggerated here, with typical widths being far less than the distance between interactions though much greater than the wavelength of radiation.



Importantly, the resulting Ramsey spectroscopic linewidth is narrower than the same feature resolved over the same interaction with Rabi spectroscopy [212] (see Fig. 3.3). Furthermore, first-order Doppler shifts can be greatly reduced through the use of counter-propagating fields, similar to saturated absorption spectroscopy. Additionally, in both methods, the energy-time uncertainty causes broadening inversely proportional to the entire interaction time, which, for dual-zone Ramsey experiments, includes the dark time between interactions. It is far easier for an experimentalist to create two small but spatially disparate fields than a single large uniform field. Thus, the two

Ramsey interactions could be spaced significantly further apart than single field could be made wide. Wider Ramsey spacing results in less transit-time broadening, and thus smaller spectral features can be resolved.

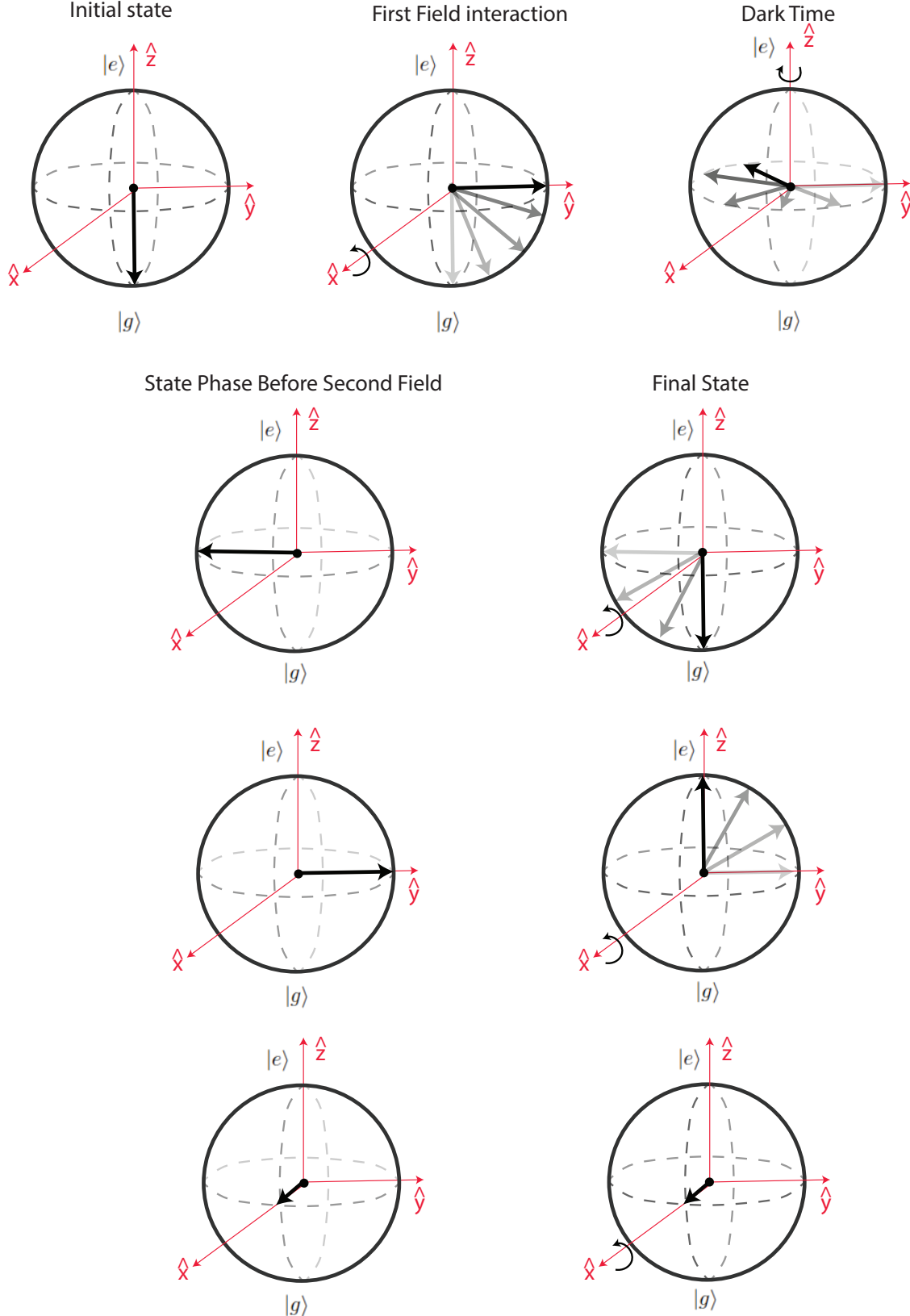
Figure 3.3: Rabi (dashed) and Ramsey (solid) lineshapes for a thermal velocity distribution with average velocity α for a distance L between atom-field interactions as a function of laser frequency ν relative to the resonance ν_0 [212]. Figure reprinted with permission from Ramsey, Norman F., Science, 248.4963: 1612-1619 (1990). Copyright 1990 by the American Physical Society.



A possible implementation Ramsey's method for a frequency reference is now briefly given. First, the external oscillatory fields are brought near the atomic transition resonance such that an atomic response is seen. The fields are then tuned to possess a frequency and amplitude such that atoms being measured experience an overall $\pi/2$ -pulse. The atom then enters the field-free central zone, wherein its Bloch state vector precesses in a plane parallel to the $\hat{x} - \hat{y}$ plane about the $\hat{z}$ -axis at a rate dependent on the detuning. A phase is accumulated during this time, and how much the phase has evolved then impacts how the atom reacts to the second field, which also has a $\pi/2$ pulse area with a fixed phase relationship to the initial pulse. Depending on the accumulated phase, ΔT , the second interaction can transfer the atom back to the ground state, up to the excited state, or anywhere in between. This is depicted in Figure 3.4.

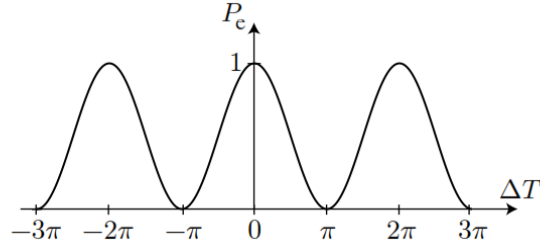
For the previous experiment, the probability of being in the initial state after the second interaction is a sinusoidal function of the field detuning, and these oscillations are called the Ramsey fringes. Simple fluorescence measurements can then be used to read off the Ramsey fringe amplitude at a chosen driving field frequency. Any change in the driving frequency will cause the generalized Rabi vector to tilt off the equator of the Bloch sphere, and thus the population transfer

Figure 3.4: These figures display the evolution of an atomic state on the Bloch sphere through two laser interactions as in Ramsey spectroscopy. Three different amounts of dark time are displayed along with their differing final state outcomes. Depending on the earlier superposition state precession, the final state can either be a return to the ground state, reach a pure excited state, or remain in some superposition state.



between states will be incomplete, causing a drop in the final state fluorescence. Of course, this process breaks down if the laser frequency deviates between measurements by more than the fringe linewidth.

Figure 3.5: Ramsey fringes from the method of separated oscillatory fields with a dark time ΔT given. The final state probability is a function of the field detuning, Δ and the dark time between interactions, T , during which the ground and excited state developed a relative phase. Figure reprinted with permission from Daniel Steck: Atom and Quantum Optics Lecture Notes [240].



Were the atoms in an Ramsey separated-field experiment to have vastly different longitudinal speeds (as could be expected for a simple thermal ensemble), the atoms would then experience a variety of pulse areas, thus a smear of rotations would result across the population. The second atom-field interaction would not be able to bring the entire population back to a single state, but rather to a smear of states around the average mono-velocity final result. This results in a loss of fringe contrast, i.e. the peak-to-peak amplitude difference of the Ramsey fringes would decrease or disappear entirely, although a strong central fringe usually remains for typical thermal distributions.

Ultimately, the Ramsey method is extremely powerful for RF and microwave interrogation fields and is now thoroughly incorporated in the world of spectroscopy including most modern versions of the cesium standard. However, Ramsey spectroscopy still possesses some drawbacks that are worth considering. As the Ramsey fringes are based upon the relative phase of the electromagnetic field at the interaction coordinates, changes in the field phase can cause shifts, drifts, and noise that degrade the atomic signal. Delivery of phase-coherent fields can be tricky depending on how many atom-field interactions are desired. Inhomogeneous broadening of the atomic transition can lead to a smear of free-precession rates in the dark time between interactions,

causing dephasing of the Bloch vectors and thus a range of final probabilities. Though some dephasing has to be expected for thermal ensembles, velocity selection or experimental design of atomic sources with narrower velocity distributions (such as a supersonic jet) can be used to increase the Ramsey fringe coherence.

3.3 Optical Ramsey fringes

The two-zone Ramsey setup does not translate smoothly to the optical domain because of the significantly smaller wavelengths involved, which impact the results in numerous ways. Firstly, it is difficult to imagine the sheer magnitude of the change in length. From the perspective of atoms, the world is a rather big place ⁴. Yet, compared to the length scales over which these tiny calcium atoms travel in a thermal beam, their motion can be large compared to optical wavelengths while still being very small relative to microwaves or RF.

The major obstacles of translating from the microwave to the optical domain for Ramsey spectroscopy lies in the significantly shorter wavelength, i.e higher energy photons. The Doppler shift for a 10 GHz microwave transition versus a 456 THz transition (the one we utilize in ⁴⁰Ca) is smaller by a factor of 45,600. Clearly, the motional shifts and collimation of the atomic beam are far more significant in the optical case. Furthermore, the length scale over which we care about atomic motion is much smaller for optical than for microwave scales. For instance, were a 600 m/s calcium atom to experience a 657 nm photon recoil (as in our Ramsey-Bordé transition) versus a 10 GHz microwave photon, the recoil-induced displacement 20 cm downstream is 5 μm and 1 Ångström, respectively. For the microwave fields, this displacement is a few millionths of the 3.26 cm wavelength, but, for the optical, this displacement is closer to seven full wavelengths. These displacements map directly onto the relative phase experienced by the atoms between the two fields, so variations in the recoil magnitude and direction across the ensemble then result in

⁴ With a radius of just 197 pm, the calcium atomic radius is 3335 times smaller than the size of the laser field with which it interacts at 657 nm. For an average 1.7 meter tall human, this is similar to standing near the tallest building on earth - the Burj Khalifa tower in Dubai at 828 meters, plus another 5.8 additional towers stacked on top of that. To the atom, even microwave radiation is huge. For a 10 GHz (3.23 cm wavelength) field, it's comparing a typical human height to the distance that light can travel in 0.93 seconds or nearly three quarters of the way to the moon.

dephasing for the two-zone Ramsey case. Small variations in the experienced transverse momentum kick of the atom for the microwave case thus barely affect the final phase of the Ramsey fringes, while, for the optical fields, such variations can easily cause enormous dephasing.

Furthermore, technological challenges inherent to using lasers for Ramsey-type spectroscopy can be significant. The laser profile, wavefront, and phase are easily distorted by optical elements and air currents. Such issues tend to worsen as the radiation's wavelength is shortened, hence the higher difficulty in the optical. Even within the optical regime, lasers with wavelengths in the blue and ultra-violet tend to require extra care when utilized in phase-sensitive processes. Just consider the well-known Rayleigh scattering expression, which says that the intensity I of light with wavelength λ scattered from particles (like air molecules) that are much smaller than the wavelength goes as $I \propto (\frac{1}{\lambda^4})(1 + \cos \theta)$ where θ is the scattering angle relative to the initial wave-vector. In the case that little scattering occurs (such as over the small optical path lengths in a laboratory), more significant than the loss in power caused by large angle scattering is the phase shift that results from scattering at small angles, which is far more probable. This leads to phase and pointing noise in the laser beam. In general, phase sensitive laser processes tend to benefit from boxes, enclosures, and short optical path lengths. In my dissertation work, the number of times that simply putting another box around the experiment solved a frustrating technical noise issue is perhaps a bit embarrassing. Additionally, the traveling wave optical Ramsey method used here requires a long optical path length of order 1 m to create the desired laser geometry. Even when mostly enclosed within vacuum, Gaussian optics and the desire for very narrow laser waists make maintaining a uniform beam throughout the optical path extremely challenging. These optical phenomena inherent to laser beams result in non-uniform and slightly disparate fields at each atom-field interaction zone. These deformities can easily cause changes in the perceived laser phase or pulse area, causing additional dephasing of the ensemble.

Thus, despite relying on the basic building blocks of 'separated oscillatory fields' pioneered previously by Ramsey, optical Ramsey experiments require additional experimental care and complexity than their lower frequency counterparts. This is especially true for thermal ensembles with

various Doppler shift mechanisms, which are generally overcome in one of three ways. First, using slowed, trapped, or velocity-selected ensembles can reduce the variations in experimental conditions experienced by the atoms, reducing the dephasing. Of these, the most straight-forward is to use cooled and trapped ensembles that are held in place. Such an ensemble can easily be interrogated by a pulsed laser sequence acting at one location, thus the field can be nearly identical from pulse-to-pulse and not suffer from laser propagation issues. However, these techniques generally reduce the number of interrogated atoms by many orders of magnitude. As signal-to-noise is a priority for fast atomic corrections as desired from our Ramsey-Bordé experiment, continued use of a high-flux thermal beam is paramount. Another way to modify Ramsey spectroscopy for the optical is by utilizing additional laser-atom interactions to rephase the ensemble. Rephasing is an exceptional powerful technique, which allows very efficient interrogation of a thermal beam that would otherwise experience severe dephasing. In thermal beam cases, rephasing is accomplished by the inclusion of more atom-laser interactions, which apply a mirror image of the initial two laser-atom interactions. However, the rephasing process was not trivial to discover or implement, and the development of rephasing methods is left to the next subsection.

3.3.1 Rephasing optical Ramsey fringes

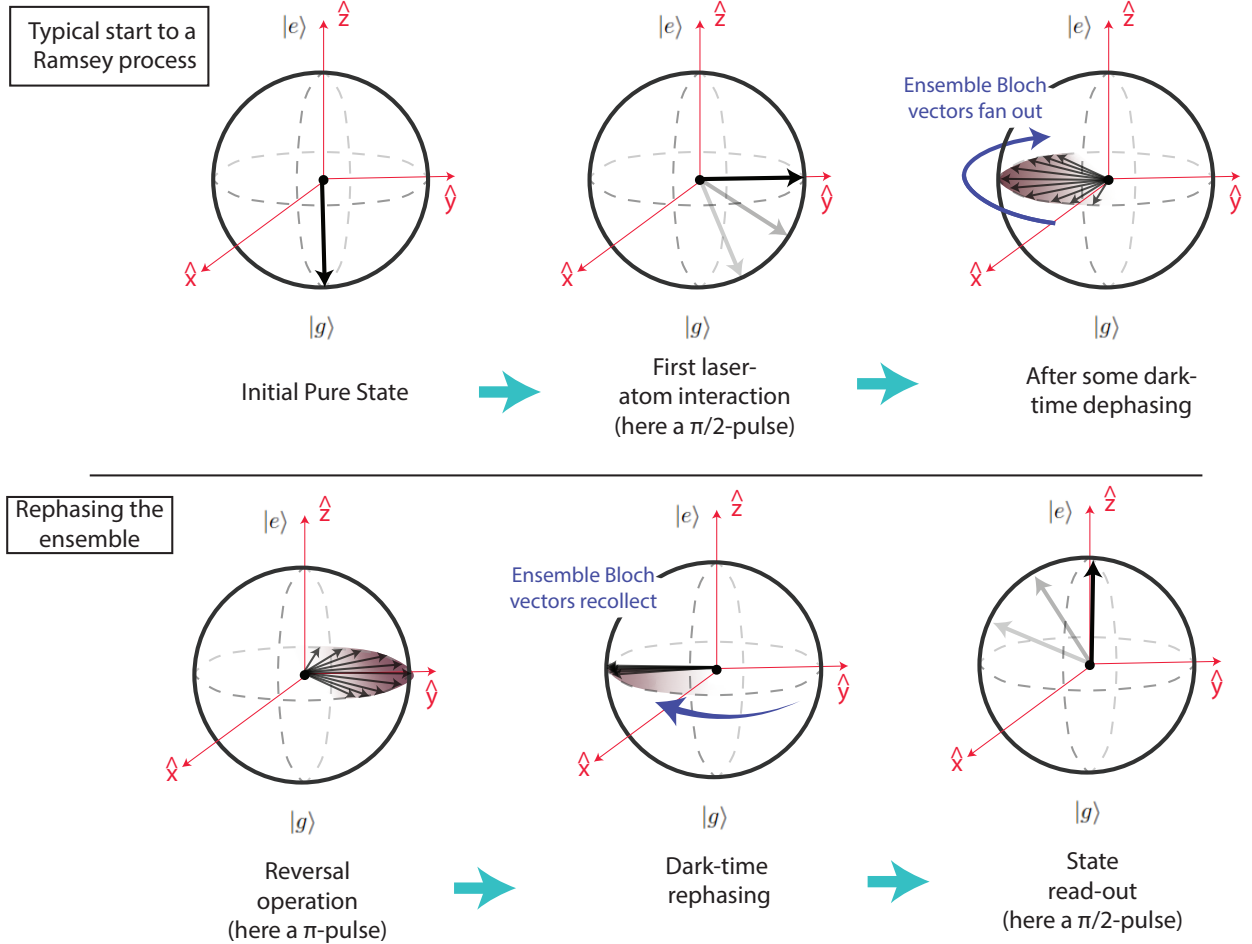
Though many scientists have contributed to the success of laser-based Ramsey experimental techniques, some of the more well-known names include Blaklanov, Dubetsy, Bergquist, Lee, Hall, Salou, Cohen-Tannoudji, Bordé, Hansch, and Chebotayev. These and many other scientists developed techniques that supply essential rephasing to an atomic beam ensemble via additional laser interactions. A similar rephasing mechanism is employed in the photon-echo technique, which is common in ultra-fast sciences. Regardless of the terminology, a rephasing process was found that allowed the Ramsey fringes to again be resolved even in the presence large inhomogeneous Doppler effects.

Photon echoes are now used in many veins of scientific research, each with their own jargon and notation. One of the most prominent areas of photon echo study involves fast pulsed experi-

ments that can control the phase, frequency, and/or timing of their atom-field interactions, which are usually performed on atoms that are effectively stationary over the time periods of the interaction. Often the presence of a wave-mixing energy level (WMEL) or a double-Feynman diagram is a quick cue that the physics or chemistry under study utilizes photon echoes. They are also used in four-wave mixing, such as in biological imaging via coherent anti-Stokes Raman Scattering (CARS).

I find the rephasing process, which is absolutely central to the concept of Ramsey-Bordé spectroscopy, to be most easily thought of on the Bloch sphere, as depicted in Fig. 3.6, wherein the inhomogeneous dark-time phase propagation is allowed to progress, but then “unwound” through a subsequent interaction with identical duration. This process allows a once varying phase shift across the ensemble to be reduced to a uniform phase shift, allowing constructive interference of the Ramsey fringes near the ensemble’s interference resonances at $\pm\nu_{recoil}$.

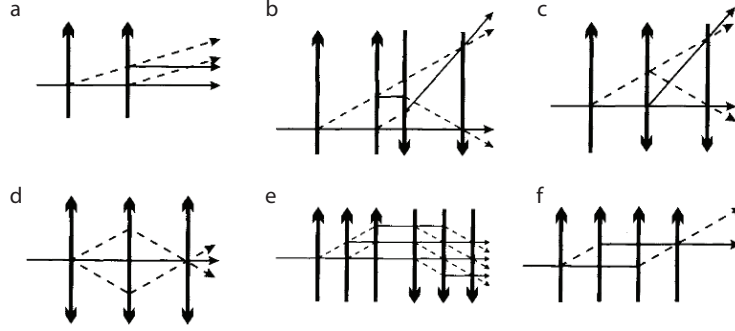
Figure 3.6: An example of dephasing caused by differing dark-time precession rates for the superposition state of a Ramsey process. The faster Bloch vectors pull ahead from the slower, causing a smear of phase values across the ensemble. This dephasing can largely be undone by providing a secondary rephasing process. In this case, the π -pulse reflects the Bloch vectors, and now the fast precessing Bloch vectors that had pulled ahead of the ensemble average are near the back of the pack. If the two dark-precession periods are the same, then, at the very end of the rephasing process, the ensemble Bloch vectors have returned to being in phase. Then readout can be performed as per usual.



The most commonly seen optical Ramsey fringe methods typically involve only three interaction regions, but many are comparable to the Ramsey-Bordé method used here. Examples of optical-Ramsey configurations are shown in Figure 3.7, relying on the momentum state diagrams to depict the atomic states, as is more formally introduced in the following chapter. Not all of these interferometers are the same, and some do not possess sensitivity to the interrogation laser

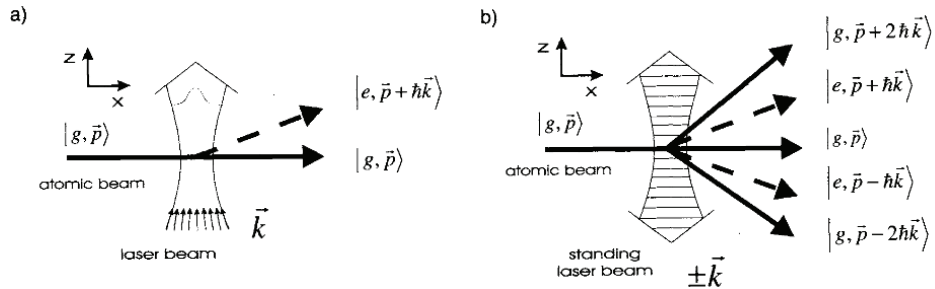
frequency [26].

Figure 3.7: Reprinted with permission⁶ A wide variety of experimental geometries exist that can successfully generate optical Ramsey fringes with separated light fields. Examples *a* to *f* depict just a few of the possible laser and atom trajectories, with laser beams indicated by thick lines. Thin dashed(solid) lines indicate excited(ground) state atoms. More information available in [26].



Though both standing- and traveling-wave geometries have been successfully used for optical Ramsey fringes, perhaps the most direct way in showing the superior signal-to-noise capabilities of the traveling-wave version is through simply viewing the possible final atomic states that can occur after an interaction with each type, as seen in Fig. 3.8. It is evident that the standing wave geometry splits the atomic wave function into far more states than with the traveling wave, which ultimately causes a smaller portion of the atom population to enter the desired states for self-interference.

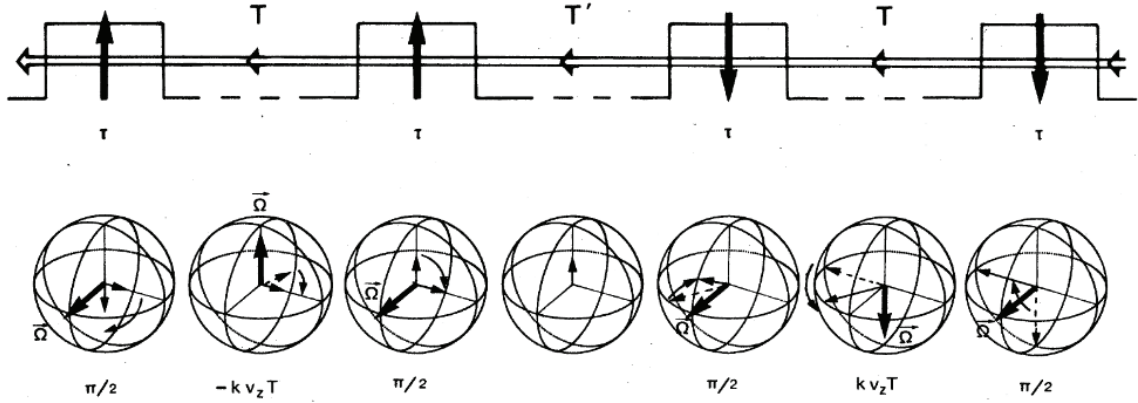
Figure 3.8: Atomic momentum states after both a) traveling wave and b) standing wave atom-field interactions. (a) Atoms undergoing traveling wave interactions split the initial $|g, \vec{p}\rangle$ atomic state far more efficiently than the standing wave version of (b), in which large portions of the population wind up in higher-order momentum states that cannot yield the desired interference fringes. This figure is borrowed from [26].



3.4 Ramsey-Bordé spectroscopy

As to our specific Ramsey-Bordé process, the four laser-atom interactions are structured as in Figure 3.9 from the 1984 paper [41], along with visuals of the Bloch sphere evolution of the atomic state. From the perspective of an atom undergoing Bordé spectroscopy, it experiences four laser pulses of duration τ with time T between the first and last pair of pulses and T' between the middle pair of pulses. In our experiment, T' is reduced as much as possible by spacing the middle two laser paths as closely as possible without degrading the fringe signal. The first pair and last pair of pulses have opposing wave-vectors. For the resonant Bloch sphere formalism of Fig. 3.9, the process goes right to left as follows. The first $\pi/2$ pulse rotates the atom's state vector from the ground state into the equatorial plane as superposition of $|e\rangle$ and $|g\rangle$. During the dark time T , the atom's state evolves in phase through an angle $\phi_1 = (kv_z T)$ while remaining in the equatorial plane. For this illustration, the state vector winds up at some arbitrary angle with respect to the rotation axis, causing the state vector to fall short of reaching the pure excited state. The middle Bloch sphere begins anew in a pure excited state, as if the atom had been perfectly interrogated at first. From here, the third $\pi/2$ pulse rotates the atom back into the equatorial plane, and then during the following dark time the atoms precesses by an angle $\phi_2 = -kv_z T = -\phi_1$. In this illustration, the dark time precession brought the state's phase to position such that the final $\pi/2$ pulse returns the atom to the ground state. It is the equal but opposite phase evolution in the dark times, combined with the symmetric $\pi/2$ rotations that cause inhomogeneously shifted atomic resonances to still maintain some coherence.

Figure 3.9: Figure reprinted⁸ from the 1984 Bordé *et al* paper [41]. Laser-atom interactions occur for a time τ , the time between outer interactions is T , and T' is the time between middle interactions. The atom passes through four laser fields whose wave vectors are indicated by arrows in the top portion. Below is the Bloch sphere representation of the Bordé process, with the magnitude of the rotation below each sphere and the vector drawn upon the sphere. Initial states are given by dashed arrows and axes of rotation are indicated by bold arrows. Curved arrows show the direction of rotation



Regardless of the method used to describe the physical Ramsey-Bordé system⁹, all methods arrive at equivalent expressions for the Bordé fringe spectra. The most interesting part of the spectrum simplifies greatly near the interference resonances at $\pm\nu_{recoil} = \pm \frac{\hbar k^2}{2M} \approx 23$ kHz, such that the excited state probability, P_e is given by

$$P_e(\nu) \propto \cos(2T(\nu - \nu_0 \pm \nu_{recoil}) + \Phi), \quad (3.9)$$

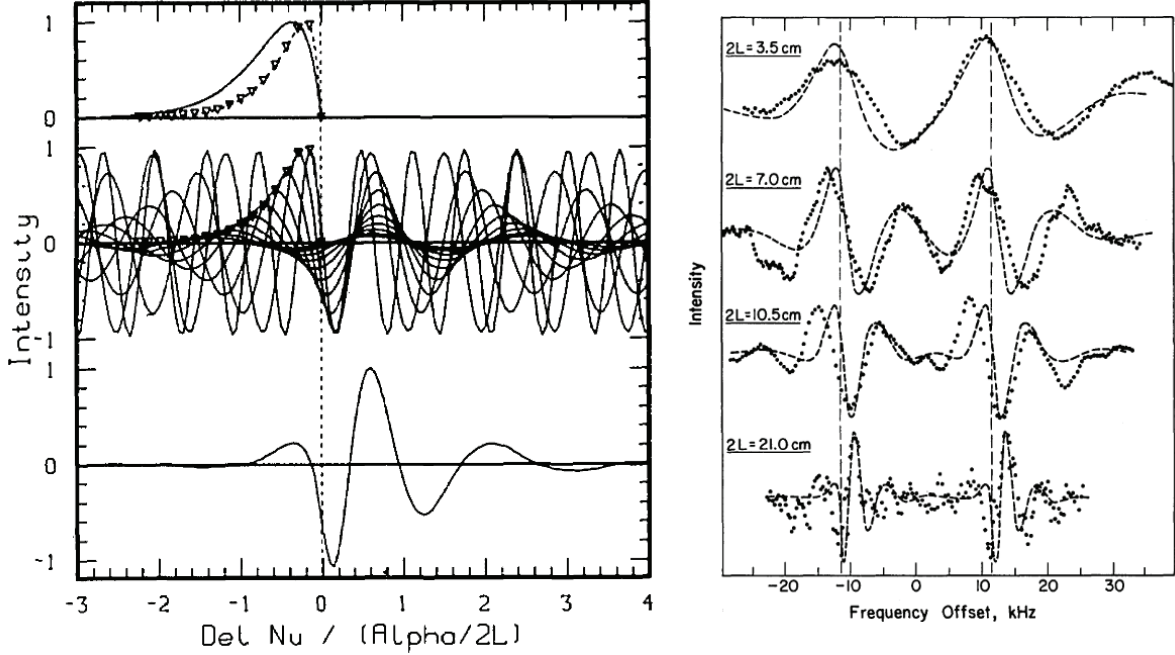
where ν is the interrogation laser frequency, ν_0 is the transition resonance, and the accumulated phase experienced by an individual atom Φ . For the Bordé geometry followed here, the phase contribution is given by $\Phi = (\phi_2 - \phi_1) + (\phi_4 - \phi_3)$, where the subscript of ϕ_i is given by $i = 1, 2, 3, 4$ is the sequential order in which the atom experiences the laser beams. Of course, this formula applies individually to each atom based on its velocity and trajectory through the Bordé spectroscopy lasers,

⁹ Like most good problems in physics, there are a great variety of formalisms that can explain the phenomena. Initially Bordé and colleagues utilized the standard quantum mechanical treatment of a two-level system exposed to successive coherent laser interactions. Later, Bordé instead framed the experiment via a matter-wave formalism wherein the interferometers appear in momentum space and atomic recoil is used to track state changes. Particularly enlightening is a chapter from Uwe Sterr, Klaus Sengstock, Wolfgang Ertmer, Fritz Riehle, and Jürgen Helmcke found in the text *Atom Interferometry*, edited by Paul Berman [26]. Though no longer in print, this text is one of the most informative sources I have found for understanding the Ramsey-Bordé formalism, and it explains the process through multiple lenses - wave mechanics, atom interferometry, and scattering theory - while highlighting various attributes more obvious in different formalisms.

Thus, to yield the ensemble result seen in the laboratory, Eqn. 3.9 must be integrated over the distribution of Ramsey dark times (T) and second-order Doppler shifted resonance frequencies (ν_0). The residual first-order Doppler shifts are usually included in the phase term, but can alternately be included in ν_0 .

A great deal of work has been done to characterize second-order Doppler shifts over thermal ensembles, and a 1981 paper by Dick Barger [17] showed some of the earliest measured second-order Doppler shifts for a thermal calcium beam. For theoretical comparison, he numerically integrated the expression for the Ramsey fringes including various envelopes for spontaneous decay, the Maxwell-Boltzmann velocity distribution, the recoil shift, and time-of-flight broadening to produce some rather beautiful spectra reprinted in Fig. 3.10. Despite these efforts, the most accurate atomic systems are still those whose motion can be highly suppressed, causing the shifts to be inherently small and calculable by statistical mechanics.

Figure 3.10: This figure reprinted from Barger's 1981 solo publication in Optics Letters¹¹ was the result of numerical integration over the thermal atomic beam velocity distribution to display the theoretical prediction for a calcium optical Ramsey fringe experiment. The first plot shows the distribution of second-order Doppler shifts, the middle curves shows a collection of calculated individual fringes, and the bottom shows the ensemble integrated result. All plots are shown as a function of the laser detuning from the recoil frequency 'Del Nu' normalized by one over the the average total time an atom spent in the interferometer, ' $\text{Alpha}/2L$ '. The right-hand plots show the calculated Ramsey fringes versus the experimentally obtained fringes. Notice the degradation in signal-to-noise for larger spacings.



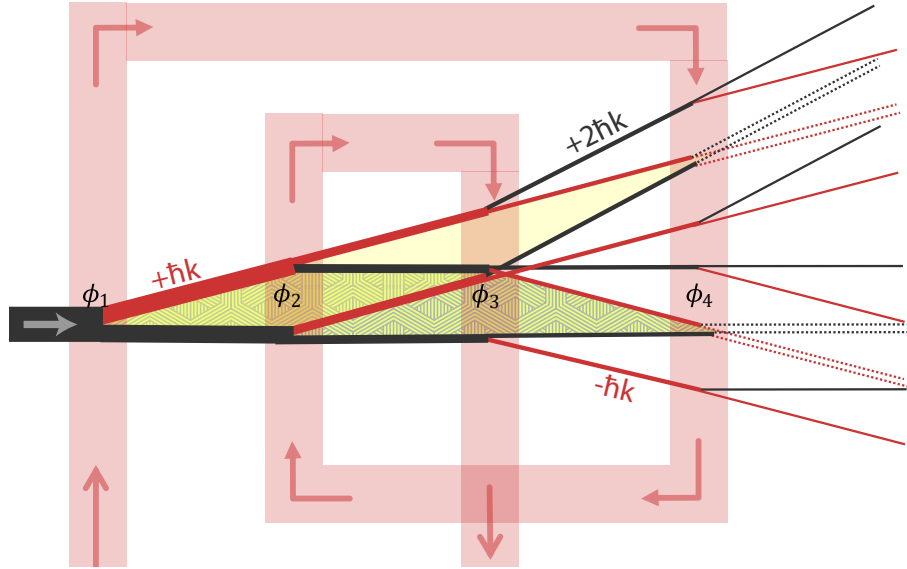
3.4.1 Atom interferometry

Though the Bloch sphere method to present the optical Ramsey fringes in Fig. 3.9 is helpful, I find visualizing the Bordé process from a matter-wave perspective to be extremely useful. In this view, the state of the atom is decomposed into discrete momentum states in units of $\hbar k$, which correlate to whether or not the atom is in an excited or ground state. Of all the momentum states that could result at the end of the Bordé sequence,¹² most do not experience matter-wave interference. In fact, by viewing the momentum-state diagram of Fig. 3.11, it is easy to simply count the states that do not interfere, as they do not take one of the two closed paths with

¹² This assumes that we are only inciting first order interactions wherein only $\pm \hbar k$ can be exchanged at each laser interaction.

indistinguishable initial and final atomic states.

Figure 3.11: This diagram most succinctly summarizes the Bordé process central to this dissertation's work. The angle of the state line indicates the momentum transfer that has occurred in units of $\hbar k$, where k is the magnitude of the laser's wave-vector along the atom's velocity component. Black lines indicate ground states while the red are for excited states. The phase of the laser perceived by this atom is given by ϕ_i where i represents the sequential order of the laser beams from left to right. The two closed quantum trajectories that display matter-wave interference are given by the filled trapazoids. The relevant final interfering states are given by dashed lines. The laser is demonstrated in a folded traveling wave configuration similar to the one used in our apparatus.



Momentum-state type of diagrams (see Fig. 3.11) were first considered in the context of Bordé spectroscopy in 1989, in a publication where Bordé related many years of previous work with atoms and molecules interacting with multiple laser fields to matter-wave interference [38]. Until this, such atomic work had not been as closely connected to matter-wave interferometry, which was already a well-established research field. Once the connection between the two formalisms was made clear, a wave of atom-interferometry experiments were proposed and carried out via established matter-wave methods. Examples of these include precision measurements of external fields and atomic properties, inertial sensing and measurement applications (gyroscopes, accelerometers, Sagnac effect), measurements of exotic quantum mechanical predictions like the Aharonov-Bohm and Aharonov-Casher phase shifts, and tests of cavity quantum electrodynamics

and quantum optics [263, 55, 4, 270, 185, 112, 216]. The large atomic mass compared to previously utilized matter-wave particles (usually electrons, neutrons, or very light atoms like hydrogen [64, 251, 218, 177, 213]) also opened the possibilities for tests of gravity and how it might interact with quantum mechanical systems [185, 149].

Relating our atom interferometry process to standard optical interferometry, we see how the lasers act like mirrors, beam-splitters, and beam-combiners. The atomic momentum states are localized by our experimental procedure and possess definite phase relationships, so it is appropriate and convenient to describe the atomic wavefunctions in position space not just as plane waves, but rather as localized wave-packets. The first laser-atom interaction splits the wave-packet, then the middle two act as a mirror to set off a phase-evolution process that counteracts the one of the first half of the interferometer. Finally, the last laser interaction recombines the wavepackets. The middle mirror in this case instigates the rephasing process previously described, which is optimal as the interferometer is symmetric. The two interferometers generated by the Ramsey-Bordé laser geometry are given by closed paths. For each individual closed path final state, the atom could have taken either route to its destination, and, in the wonderfully quantum nature of the world, that means it *did* take both paths. The phase difference accumulated along these two paths determines the phase of the resulting interference fringes. For thermal beams, the atomic beam width is typically much greater than the physical separation caused by the laser interactions, and thus the interfering states cannot be selected by their final location.

While a rich atom interferometry formalism has been developed for Ramsey-Bordé (see [26] among others), further detail is beyond the scope of this dissertation. It should also be noted that the interferometry formalism provides additional tools for predicting the atomic beam dimensions that can be maximally addressed for a given laser beam profile.

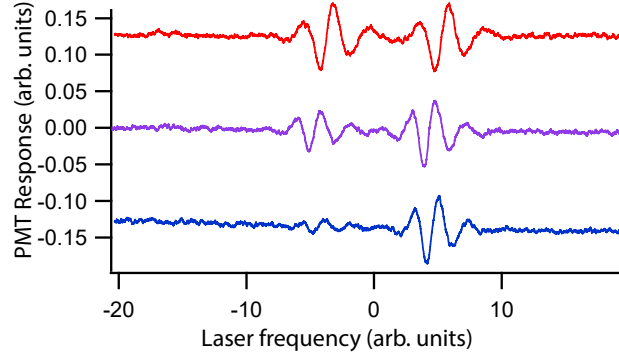
With the basics of Ramsey-Bordé well developed, the specifics of how the calcium beam's velocity distribution affects the interference fringes can now be explored.

3.4.2 Fringes: recoil components and velocity distribution impact

Given our calcium thermal beam ensemble, Ramsey-Bordé interference fringes are only resolvable by the interrogation laser within two narrow regions of frequency space of order 10 kHz. Very coherent interference fringes from uniformly moving ensembles can exhibit coherent interference fringes over a large range of frequency space spanning hundreds of kHz. However, only one cycle of the fringes are needed to provide frequency discrimination. Thus, so long as the thermal averaging is kept limited enough to resolve a cycle or two of Bordé fringes, the method can be used to provide frequency stabilization.

Of course, there are two recoil components in a Bordé interferometer, as depicted in Figure 3.11. However, it is possible to destroy either of these interference fringe components by disrupting the coherence of their process. Often this is done in the central region between the second and third laser interactions. Though this central distance is often made as small as possible, it is only here that the two interferometers are in pure states again, albeit in spatially separated wavepackets. Thus, the interferometer can be ‘selected’ here by disrupting either the ground or excited states. This trick has been demonstrated before with application of an additional laser connected to the ground state on the 423 nm calcium cycling transition is able to disperse the ground state atoms in this zone, destroying the higher frequency fringe. A similar process was attempted with our calcium apparatus, and the results from it shown in Fig. 3.12. Here, the 431 nm laser (also use for electron shelving detection) was used to destroy the lower frequency fringe component’s coherence while leaving the higher frequency one unchanged.

Figure 3.12: Three frequency sweeps showing the reduction of the lower-frequency fringe component as its coherence is degraded by additional application of a blue laser between the second and third laser interactions. The top trace (red) shows the fringes in their normal state, which the middle (purple) has the blue laser tuned near the excited state transition, while the bottom (blue) trace shows the maximum effect. The fringe could not be destroyed completely due to the unavoidable angle between the blue laser and the atomic beam, causing a portion of the beam to be shifted out of the blue laser's resonance.



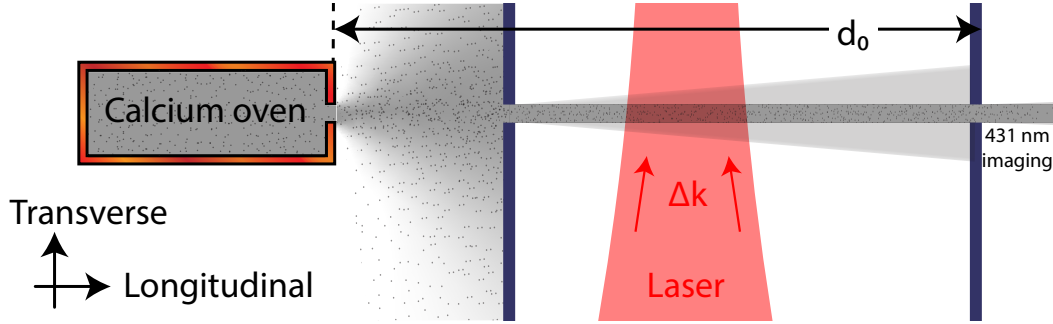
We have never noticed any significant differences in the properties of the higher versus lower frequency recoil components. In theory, with some finite distance between the second and third laser interactions, the interferometer in the excited state here would experience some spontaneous decay while the other interferometer path would not. However, we keep the distance between the central interactions so small that this impact is negligible. In general, we locked to the lower-frequency recoil component interference fringe, but it was largely out of habit.

The physical modeling of our Bordé interferometer can largely be constrained to a plane rather than a three-dimensional space, leaving two primary velocity components of interest. The first is defined by the average velocity vector of the atomic beam and is called the longitudinal velocity. The second axis is referred to as the transverse direction, and is orthogonal to the longitudinal and vertical directions, ideally lying along the average $\vec{k}$ -vector. We use both atom and laser beams that are elongated, but little impact is seen on the fringe signal other than signal-to-noise improvements. This is because the important dephasing terms that destructively interfere away from $\pm\delta\nu_{recoil}$ concern a term akin to $\vec{k} \cdot \vec{v}_{atom}$, which is minimized in the plane defined by the longitudinal and transverse axes through our choice of aperture geometry and laser alignment.

Despite the large magnitude difference between the transverse and longitudinal velocity com-

ponents, both contribute important dephasing mechanisms to the calcium interferometer and thus impact the obtainable interference fringe properties. The velocity distributions have been calculated numerous times elsewhere [211, 146, 6]. The pertinent dimensions and experimental alignment of interest for discussion of the atomic beam velocity distribution are given in Fig. 3.13.

Figure 3.13: The relevant details related to the following discussion on the atomic beam are given. The velocity vector is decomposed into the orthogonal longitudinal and transverse components. A distance d_0 lies between the first aperture (the oven nozzle) and the far fine aperture, which, limit the transverse velocity distribution, while the oven temperature corresponds to the longitudinal velocity distribution. Only one laser-atom interaction is depicted between the two fine apertures rather than the four that are used for Ramsey-Bordé. Not to scale.



The velocity distributions discussed here are based upon some basic assumptions. First, the apertures are purely open spaces, i.e. we are not employing microtubules, capillaries, or gratings in the apertures that might otherwise alter the velocity distribution. Second, the atomic beam's longitudinal speeds are roughly approximated by a standard Maxwell Boltzmann distribution. Third, the angular probability distribution from a point source oven is readily approximated by the cosine of the deflection angle. . For the very limited range of transverse velocities allowed by our apertures, the transverse velocity distribution is a relatively flat function.

Longitudinal velocity distribution

The distribution of atomic velocity components along the average propagation direction of the atomic beam is what is meant here by the term ‘longitudinal velocity.’ It is rather easy and informative to consider atoms only varying in their longitudinal velocities. Two atoms differing in their longitudinal speeds obviously undergo different Bordé interferometer processes because

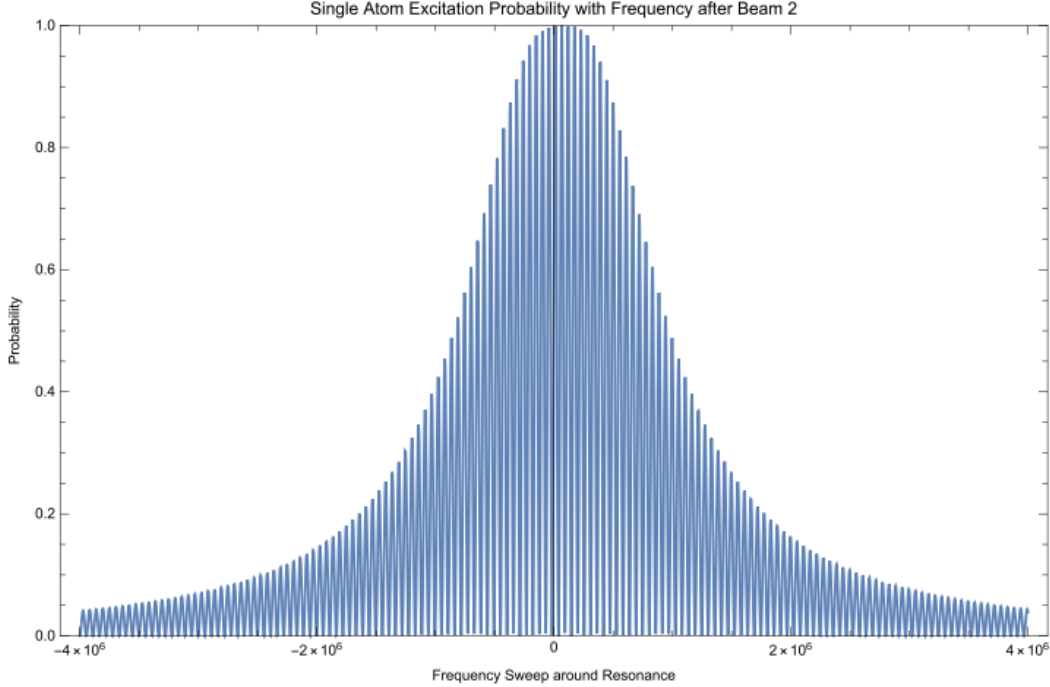
they experience differing transit times through the interferometer as well as different laser-atom interaction times. Additionally, these two atoms have differing second-order Doppler shifts (time dilation), so their 657 nm transition is shifted relative to each other.

The excitation probability after each sequential Bordé laser interaction for a single atom possessing zero transverse velocity component is fully given in Appendix B along with plots of the excitation probability versus 657 nm laser detuning. After the first Bordé interaction, the atom's excited state probability as a function of the laser frequency is roughly that of the standard Gaussian lineshape¹³, with a linewidth determined by the inhomogeneous time-of-flight broadening of the interaction (all transverse velocity components are zero or negligibly small here). The peak is frequency-shifted by the photon recoil¹⁴ and time dilation (second-order Doppler). After the second Bordé laser-atom interaction, the excitation probability displays clear velocity gratings for the ground and excited state probabilities as a function of 657 nm Bordé interrogation laser detuning. This velocity grating is seen in the Monte-Carlo simulation of Appendix B and is reprinted here in Fig. 3.14.

¹³ Spectral lineshapes dominated by homogeneous effects are usually well-approximated by Lorentzian lineshapes. Inhomogeneous broadening usually causes Gaussian lineshapes. When both mechanisms are present, the result is a convolution of the two lineshapes, called a Voigt profile. Since the feature linewidth is determined by the inhomogeneous time-of-flight broadening, a Gaussian is largely what one would expect to find.

¹⁴ Of course, recoil would impart a finite transverse velocity, but for purposes here it is assumed small enough to ignore.

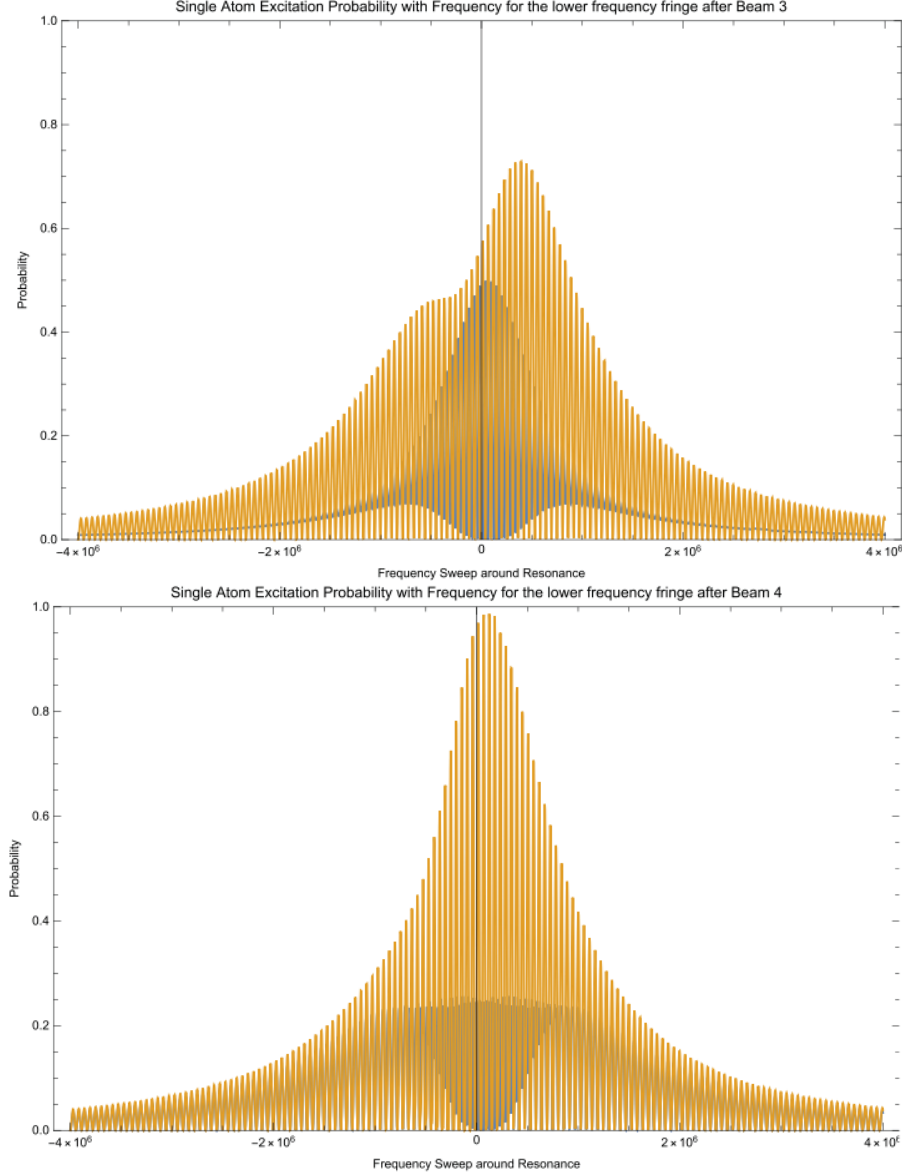
Figure 3.14: The clear probability grating imposed upon the spectroscopic lineshape after the second atom-laser interaction of the Bordé-Ramsey sequence. The period of the oscillatory grating's frequency dependence is determined by longitudinal velocity of the atom being interrogated. The initial transverse velocity here is assumed zero.



Though the first two laser interactions can be approximated in a simple two-level basis (See Ch. 3.2.1) for both of the recoil components of the interferometer, but the subsequent third and fourth interactions can be greatly simplified by breaking the atomic states for each closed interferometer into two disparate two-level systems, as demonstrated by Bordé *et al* in [39]. The third laser interaction is where the two closed interferometers of the Ramsey-Bordé system diverge, with differing velocity gratings for the two interferometer recoil components. By making the physical distance between the first pair of laser interactions equal to that the distance between the last pair of laser beams, the differing dark-time precession phases accumulated by most every atom can then be 'unwound' (see Ch. 3.3.1) so that coherent interference fringes can be recovered. Thus, the fourth interaction primarily serves to close both interferometers at the proper time such that the two halves of the interferometer are balanced and there is minimal dephasing across the ensemble. Thus, the fourth laser interaction allows the atomic state to be read off at some later arbitrary time and place along the atomic beam.

Plots of the excited state probability after the third and fourth laser interactions are given in Fig. 3.15. While it can be difficult to see the two interferometer components in these plots, there are some interesting features worth mentioning. Firstly, both probabilities after the 3rd beam have clearly moved to a higher frequency than the bare resonance demarcated at zero on the horizontal axis. The orange trace in particular shows clear skewing toward higher frequencies. Such Doppler profile traits were routinely seen when optimizing the Bordé fringes in the lab. For some time there had been concern that the rather odd Doppler profiles resulting when the fringes had the most contrast indicated technical problems with the 657 nm interrogation laser alignment or profile. However, after the 3rd and 4th laser-atom interactions, all simulations showed similar skewing toward higher frequencies, and I was able to conclude that such spectroscopic signals actually indicated very good alignment of the 657 nm laser. Knowing this, alignment of new laser geometries was made easier as was searching for the fringes along the Doppler pedestal, as they did not typically appear perfectly centered within the profile.

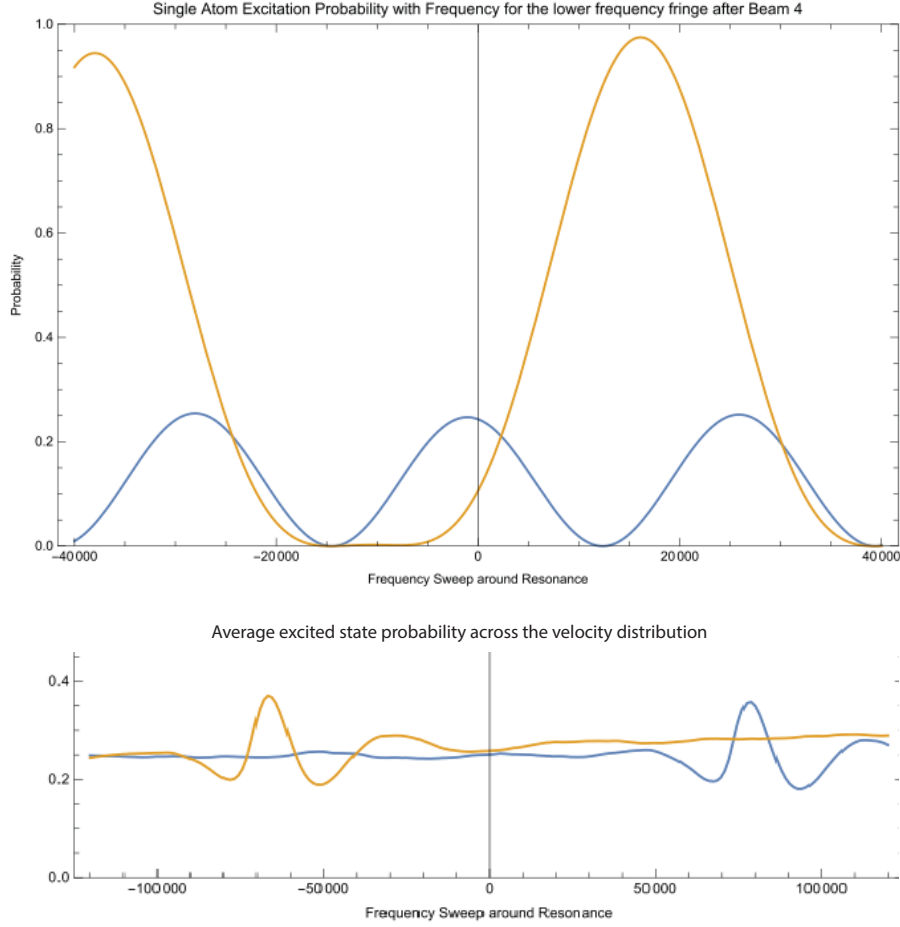
Figure 3.15: The excited state probability of a single atom moving at the most probable longitudinal velocity with an initially zero transverse velocity are given. Clearly, for a mono-velocity distribution, many iterations of the Bordé fringes would be visible. See text.



Even when the interferometer lineshapes are averaged and weighted by the longitudinal velocity distribution, Ramsey fringes remain visible near the $\pm\delta_{recoil}$ resonances, albeit shifted at the kHz level by time-dilation shifts. In fact, the fringes are not well predicted by the final mono-velocity result, as seen in Fig. 3.16. It is only when averaging over the ensemble that the Monte-Carlo predicted fringes begin to resemble what we see in the laboratory. Both the monovelocity and

the ensemble averaged probabilities are given in the figure, showing the stark differences in their excited state probabilities.

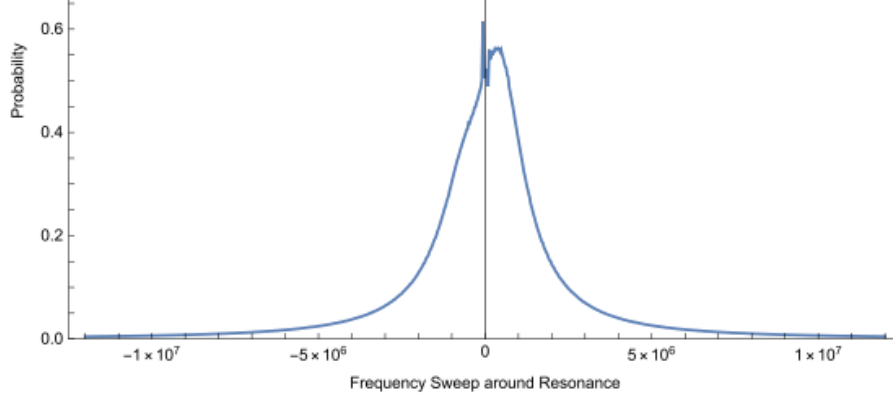
Figure 3.16: The excited state probability of a single atom moving at the most probable longitudinal velocity with an initially zero transverse velocity are given. Clearly, for a mono-velocity distribution, many iterations of the Bordé fringes would be visible. See text.



When summing over the entire ensemble, the fringes can then be clearly found not at the Doppler peak, but rather on the side of the Doppler, as seen in Fig. 3.17.

Of course, other physical mechanisms not included in this model also impacted the Doppler profiles seen in the laboratory. Notably, we often found the Doppler to display a rather flat and broad peak compared to what would be expected from models. This likely originates from the laser configuration, which acts somewhat like saturated absorption spectroscopy, albeit with a spatial displacement. While we did not typically see a single large lamb dip, the large angular distribution

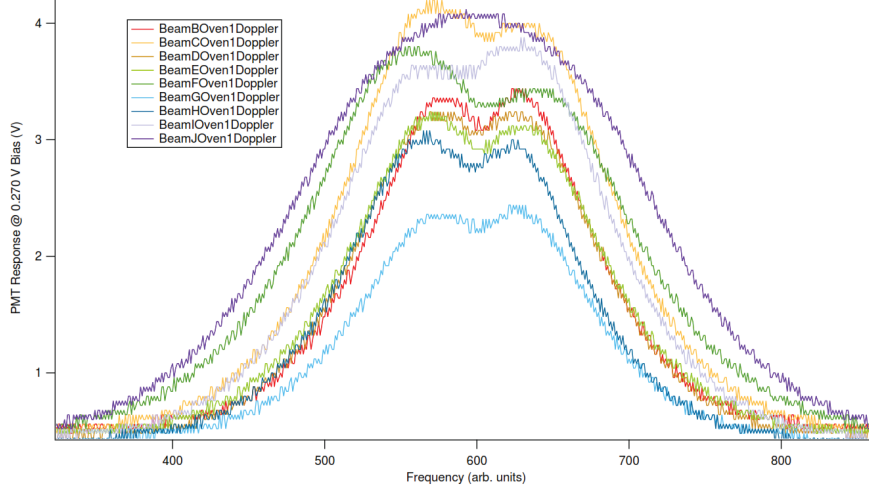
Figure 3.17: When averaging over the velocity distribution, the Bordé fringes can be clearly seen to be off-center from the Doppler profile. Though such results were commonly seen in the laboratory when the fringe contrast was optimized, it wasn't until these Monte-Carlo simulations also displayed the same asymmetry that we finally stopped trying to erroneously 'fix' the skewed Doppler profiles. Simulation results here suffered from low resolution in the plot, but the fringe location can be clearly seen as being near the large spike just sky of the horizontal axis zero.



of the laser wave-vector could have easily caused a smear of lamb dips to result, causing a rather large flat region near the Doppler peak. Some of these abnormal looking Doppler spectra can be seen in Fig. 3.18, which was low-resolution experimental data pulled off the Ca1 12-bit digitization measurement for a variety of fringe-contrast-optimized interrogation laser profiles and collimations.

The velocity distribution's impact can be easily addressed through the pulse-area method previously discussed (Sec. 3.2.2) and yields a simple result. For a sufficiently powerful laser, a single laser interaction yields an average $\pi/2$ -pulse (50% excitation) for the longitudinal velocity distribution as seen in Fig. 3.19. We eventually arrived at 657 nm laser intensities near 2500 W/m², as measured before entering the Ca2 vacuum chamber, which is coincidentally near the 50% excitation level for this simple model. Laser intensities beyond the excited state probability maximum were not obtainable with our apparatus, so, unsurprisingly, we saw no point at which increasing the 657 nm power caused a decrease in the single laser-atom interaction signal strength. However, this model utilized the basic Rabi flopped expression derived in Eqn. 3.6 back on p. 60, with only the time parameter being altered, and thus does not account for Doppler shifts,

Figure 3.18: Given here are a variety of Doppler profiles resulting from an assortment of 657 nm interrogation laser beam profiles and collimations. All Dopplers were recorded after the fringe contrast had been optimized via alignment for the chosen input laser properties. All Dopplers given here corresponded to similar fringe contrasts within a factor of 2.

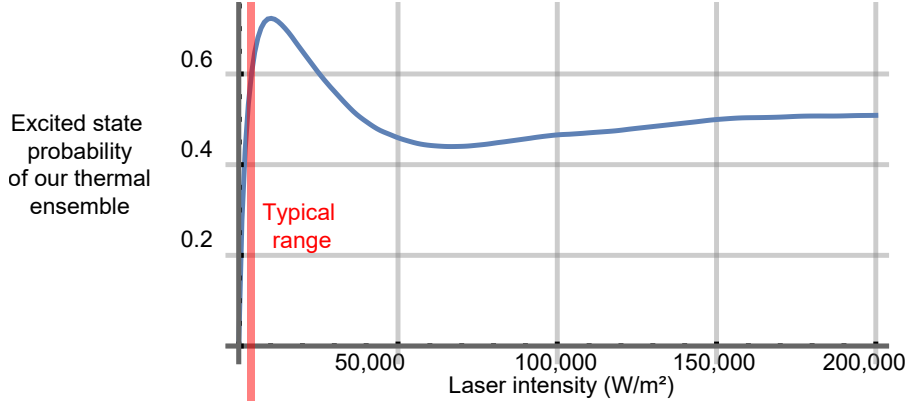


time-of-flight broadening or the laser's intensity profile.

The varying interaction times also result in a spread of time-of-flight broadening with a thermal distribution, with the average broadening of our calcium ensemble falling at hundreds of khz. Ultimately, this means that the slower moving atoms will experience the laser field for longer times, giving them the sharpest sensitivity to being Doppler-shifted out of resonance from the laser. Thus, faster moving atoms can participate in the interferometer with a greater variation in their transverse velocity distributions (Sec. 3.4.2.1). The excited-state probability from faster atoms within the atomic beam should therefore be weighted more heavily than the slower moving portion of the ensemble population as the atomic beam width is increased.¹⁵ The variation of transit times within the Ramsey-Bordé traveling wave system results in a wide variety of fringe frequencies (linewidths) and pulse-area-impacted amplitudes, as can be seen in Fig. 3.20. These fringe shapes were calculated using the formalism established in Dick Barger's 1981 paper [17] with variables

¹⁵ This effect is somewhat counterbalanced. First by the electron-shelving detection scheme, which collects more photons from slow moving atoms that remain in the cycling zone for longer periods of time. Second by the reduced pulse-area faster atoms experience. Though the reduced pulse area is offset somewhat by the large laser powers and wave-vector distributions we use, which negate some of the Doppler-shifts and low pulse-areas that otherwise might be experienced by faster moving atoms.

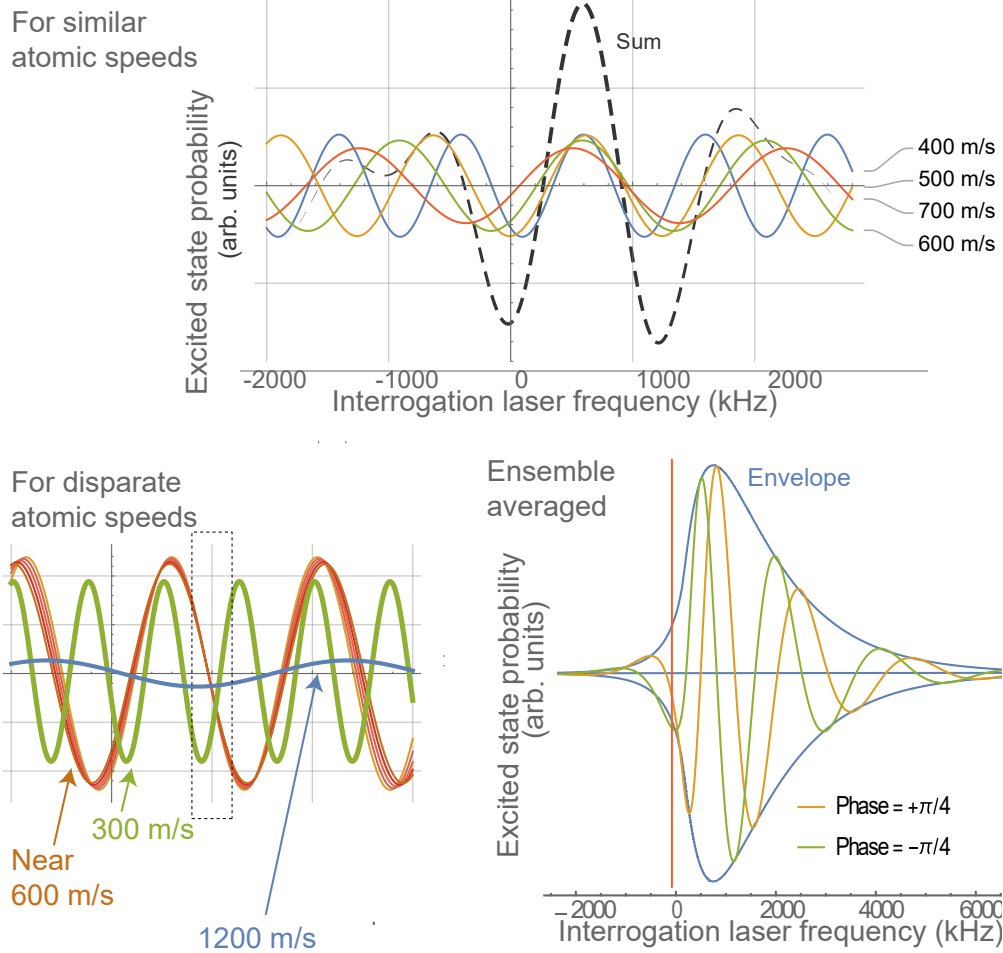
Figure 3.19: As the laser power is increased for a fixed beam area, the atomic response can be predicted using Monte Carlo modeling with 10,000 atoms, weighted by the probability distributions behind their transverse and longitudinal velocities. This is the excitation probability given intensity for a calcium atom passing through a uniform radiation field with dimensions mimicking our experimental system. Fringes were optimized in the 10-35 mW range for beam sizes discussed in the text, corresponding to values around the peak of this plot.



specific to our apparatus. The traces are for a range of velocities from 500 – 800 m/s such that all of the components roughly overlap constructively.

Yet even for a broad thermal population, Ramsey-Bordé yields phase-coherent interference fringes in the region near the average ~ 1.8 kHz time-dilation shift recoil resonance. All calculations involving the ensemble must be averaged over the Maxwell-Boltzmann weighted statistics for our given oven temperature and calcium atomic mass. However, were the longitudinal speed distribution to be more of a flat function, the fringes would begin to disappear. This is clear when considering Fig. 3.20, where, for a group of speeds around the mean, their interference fringes generally interfere constructively. However, for speeds a factor of two or more from the mean speed, the fringes no longer do so. Thus, we actually don't want to image every atom in our atomic beam - just those with speeds near the middle of our calcium Maxwell-Boltzmann distribution. Thus, the interrogation laser properties are tuned to give the most favorable pulse-areas for the atoms near the peak of the longitudinal distribution.

Figure 3.20: Top: Bordé interference fringes modeled after [17] are shown for an interrogation laser frequency sweep about the recoil-shifted resonance at zero. Bottom: The unidirectionality of the second-order Doppler shift causes asymmetries in the fringe envelope when averaged over the ensemble and weighted by the speed probability. Due to the inherent velocity selection from using short cycle times and atomic beam collimation apertures,¹⁷ we see more subtle asymmetry in our fringes.



For cooled and trapped atomic systems, the interaction time can be made nearly identical for the entire ensemble, which can yield many, many iterations of Ramsey fringes. However, for the thermal ensemble used here, less than two cycles of fringe remain with sufficient signal-to-noise for the level of stabilization achieved here. This is no loss, as a half cycle of very well resolved fringe is all that is really necessary for the locking method to work. A few cycles ensure that the fringe waveform is relatively symmetric and possesses good contrast.

Overall, this work suggests that even hotter ovens, thus with denser and faster moving atomic

beams, can still display coherent optical Ramsey fringes despite the widened spread of longitudinal atomic speeds. We found that it would take a far greater longitudinal velocity spread to wash out the fringes through time-dilation shifts, beyond what is accessible with heated Knudsen cell style ovens relying on vapor pressure. However, within the oven temperature ranges available to us, we did generally find that the system's quantum projection noise does decrease favorably as the ovens are further heated and more atomic flux exits, even though the longitudinal dephasing is worsened and the fringe linewidth broadens.

Lastly, the form of the Maxwell-Boltzmann distribution utilized here is given by

$$P(v_l) = \left(\frac{2}{\pi}\right)^{1/2} \left(\frac{m}{k_B T}\right)^{3/2} v_l^2 \exp\left[-\frac{mv_l^2}{2k_B T}\right] \quad (3.10)$$

with a most probable (mode) speed of $\tilde{v}_l = \sqrt{\frac{2k_B T}{m}}$ and the average (mean) speed being $\bar{v}_l = (2/\sqrt{\pi})\tilde{v}_l$, where m is the particle's mass and T is the average temperature of the ensemble. However, many derivations of the longitudinal thermal distribution, including Ramsey's work in the infamous *Molecular Beams* text [211], instead weight the exponential function by a factor of v_l^3 along with an altered normalization factor. Though the v^2 distribution form of Eqn. 3.10 was initially used out of ignorance, its properties seemed to better replicate the observed spectroscopic linewidths of our atomic system, and thus it is used throughout the bulk of this dissertation. A possible explanation for this is that our electron-shelving detection scheme preferentially weights slower moving atoms over faster ones in a linear fashion, such that a $1/v$ dependence arises in the detection. Thus, the weighted distribution we actually image is closer to the v^2 form of the exponential Maxwell-Boltzmann.

When modeling the atomic beam with a Monte Carlo simulation, an ensemble of atoms is initially selected with some initial speed given by Eqn. 3.10, which serves as the longitudinal velocity distribution. Though not entirely correct, the atom's transverse velocity is then selected using a relatively broad normal distribution with cutoff values chosen to account for the apertures.

3.4.2.1 Transverse velocity distribution

The transverse velocity distribution causes a far less obvious impact on the beam ensemble than the longitudinal variation previously described. The atomic velocity components along the laser wave-vector $\vec{k}$ are responsible for the residual Doppler shifts experienced by the interferometer and can ultimately degrade the experimentally realizable signal-to-noise of the ground state population. For a calcium atom moving ≈ 650 m/s, our 1 mm wide apertures almost 24 cm apart limit its maximum transverse velocity to a few meters per second. For our 657 nm wavelength, this imparts a maximum Doppler shift of about 3.8 MHz, which makes sense given the FWHM of the atomic beam's Doppler profile falls around 4 MHz, as seen in most Doppler profiles presented here such as back in Fig. ???. Clearly, the Doppler shift is about ten times greater than the time-of-flight broadening created from our short laser-atom interactions, and some atoms will thus not be resonant with our laser beam. However, nonzero wave vector distribution of our interrogation laser can compensate for this somewhat. Then, because the Bordé process is essentially first-order Doppler free, a wider atomic beam allows for an increase in signal-to-noise without degrading the spectroscopic interference fringes.

Consider for a moment a calcium beam very similar to that used in our experiment. The beam is defined by the oven nozzle and the far aperture, both of width r and separated by a distance d . For $r \ll d$, the maximum transverse velocity, v_z , for an atom with speed of v is given by $v_z \approx v \frac{r}{d}$. In our system, $d \approx 30$ cm while $r \approx 1$ mm, and a reasonable atomic speed is 600 m/s, giving a transverse speed of ± 2 m/s. This yields a first-order Doppler shift of $\approx \pm 3$ MHz. Time-of-flight broadening due to laser waists of 100 μm (this is an underestimate) only account for ≈ 0.5 MHz of broadening (overestimate). The main mechanism for addressing the majority of our atomic beam thus lies in the slightly uncollimated laser beam, for which I found a significantly larger atomic response than with a well-collimated beam. The spread in wave-vectors from our laser allow it to address a large spread of transverse velocities resonantly. In essence, the laser beam has some $\vec{k}$ -vector component such that $\vec{v}_{atom} \cdot \vec{k} \approx 0$. However, using an uncollimated beam like this then

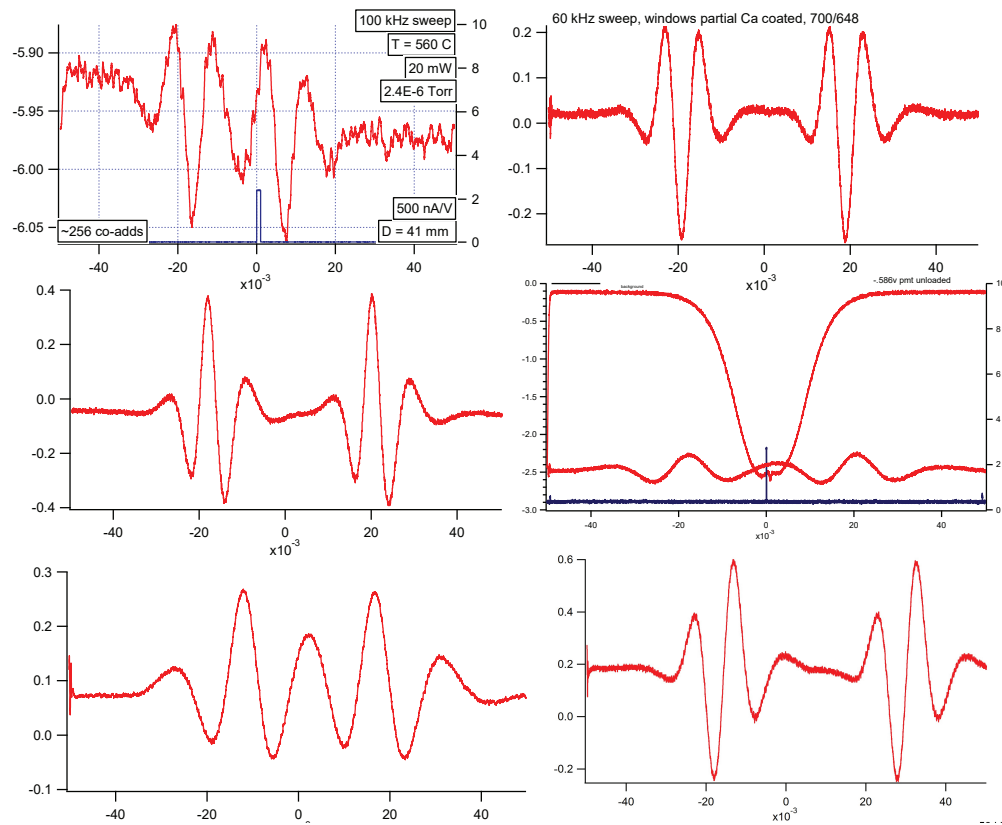
also requires more total laser power to ensure that adequate pulse areas are obtained given that only a portion of the total laser beam is resonant with the given atom.

If the transverse velocity distribution were to be further widened beyond what is resonantly addressable by the interrogation laser, then the system would begin to generate a large population of atoms that are not likely to contribute to the fringe signal but may emit photons in the detection zone anyway. In other words, the Doppler pedestal upon which the fringe signal sits would grow faster than the fringe amplitude. In theory, this is not an issue, but it does create some practical challenges for the experimentalist, such as an increase in detector shot noise. Due to the electronic conversion and amplification noise that occurs after the atomic system, increasing the contrast is a more directly beneficial improvement to the system's potential stability than simply increasing the total signal amplitude. Further increasing the atomic beam width well beyond this would eventually see a leveling out of the fringe contrast value, and atoms Doppler-shifted far from resonance would largely contribute to broadening of the Doppler pedestal rather than increasing its amplitude.

Total result

Assuming all of the atoms in the beam are resonant with our interrogation laser, then the atomic beam yields a 25% maximum contrast fringe signal lasting a few cycles sitting atop a Doppler broadened peak. For the Ramsey-Bordé system presented here, we often measure fringe contrasts around $\approx 13\%$, with a maximum measured value of 19%. Over my years on this calcium experiment, I have looked at many thousands of different Ramsey-Bordé fringes, and I would like to take Figure 3.21 to give a general picture of how varied the Ramsey-Bordé fringes appear over a frequency sweep for a variety of our system parameters.

Figure 3.21: A collection of Bordé fringes from our apparatus showing the many variations in linewidth, signal-to-noise, and phase. The phase would often slowly shift as the system reached thermal equilibrium after changes to the calcium oven temperature set points.



With the theoretical basis of Ramsey-Bordé on a thermal beam now well established, it is finally time to discuss how the system was implemented and the results obtained.

Chapter 4

Exploratory assessments of Ramsey-Bordé

This chapter covers a variety of experiments performed with an initial calcium thermal beam frequency stabilization system dubbed Ca1.¹ Ca1 was largely designed with flexibility of the Ramsey-Bordé laser geometry in mind. While our laboratory collaboration was developing the Ca1 system, I was primarily invested in optimization of the physics package, which includes the lasers, vacuums, and atomic beam systems. These systems are covered in detail first. Then I discuss the ancillary control and processing electronics systems utilized to lock our laser to the atomic signal. While some of the ancillary electronic systems/servos did differ between Ca1 and the later system iteration, Ca2, the basic principles underlying these systems were consistent. Thus, the Ca2 control systems are also largely explained at the end of this chapter but updated ancillary system specs are briefly discussed in the following chapter.

The intent of my Ca1 physics package analysis given here goes beyond merely reporting which experimental values (field strengths, Ramsey beam spacing, etc) worked best for our frequency stabilization system. This section also conveys how the Bordé process can elegantly resolve a powerful spectroscopic signal from the chaos of a thermal beam ensemble, complete with discussion of relevant thermal beam velocity distributions, flux levels, oven temperatures, etc. Yet even without a solid understanding of atom interferometry or the specific physical mechanisms that govern the Ramsey-Bordé fringes we utilize, this chapter should at least make clear how such a system responds to nearly every measurable variable in the laboratory. Where an effect could not

¹ Pronounced /sē/ā/wæn/.

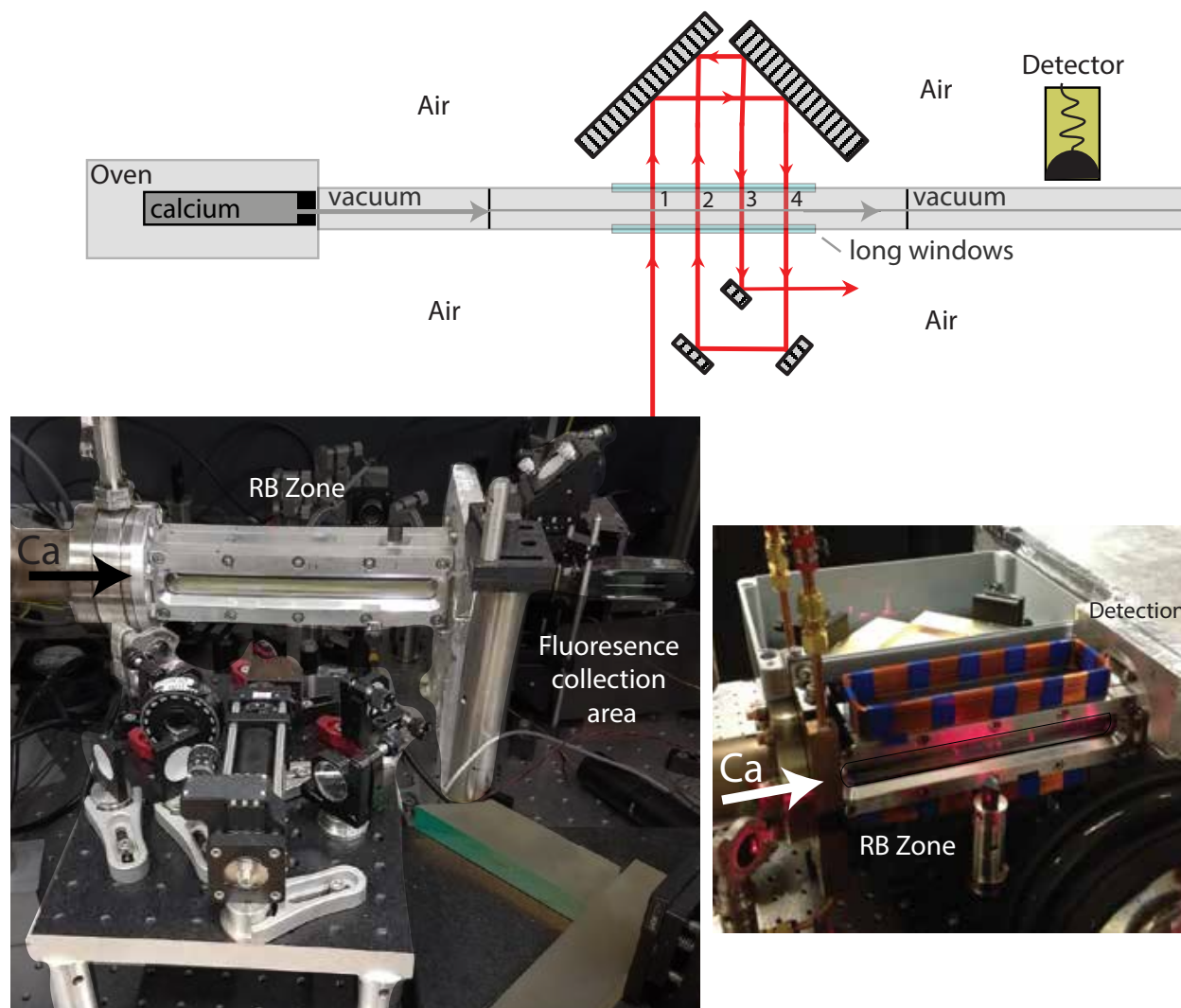
be measured with enough accuracy to infer its noise contribution to frequency stabilization, limiting cases were then implemented to at least draw some conclusions about the noise source.

Part of why experimental physics work was so crucial to the development of this calcium interferometer systems stems from how interconnected many of the relevant experimental variables are through their impact on the calcium atoms. As noted in the previous chapter, some of the resulting Doppler lineshapes or asymmetries were originally thought to indicate problems, but successful modeling of the experiment revealed that what appeared abnormal should have actually been expected. As modeling cannot possibly encompass all of the technical details of an experiment, laboratory results were sometimes unexpected but provided a more reliable understanding of the fringes. Higher order effects can be significant and insidious noise sources can find unexpected methods of coupling to the Ramsey-Bordé interference signal. This interconnectedness makes it difficult to determine the ‘optimal’ experimental configuration through theoretical calculations alone, and, indeed, I found some rather counterintuitive results yielded the largest fringe signal sizes.

4.1 The Ca1 system

The preliminary apparatus, Ca1, was designed for Ramsey-Bordé interrogation of a single atomic beam enclosed within vacuum. This beam was generated with ovens that heated a molybdenum crucible full of ^{40}Ca to $500 - 610^\circ\text{C}$. The ovens could not reach temperatures above this range. A rectangular hole in the crucible nozzle allows calcium to effuse into the main vacuum chamber body, where the beam is collimated using two fine apertures with slits of approximately 1 mm by 1 cm separated by ≈ 40 cm (though the oven nozzles themselves also act as a fine aperture). Long windows were placed on the vacuum chamber walls on either side of the calcium beam such that the 657 nm Bordé laser configuration could be easily controlled and manipulated from outside the chamber. At the end of the long windowed chamber was an imaging cell, where the calcium beam excited-state population was read off. An unfortunately long distance of 15 cm was required between the last Bordé laser zone and the imaging cell. A picture of this apparatus can be seen in Fig. 4.1.

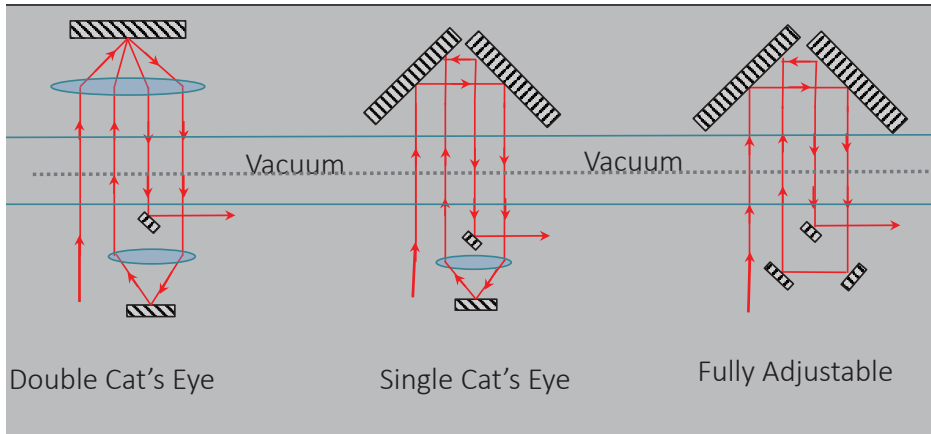
Figure 4.1: Early version of the calcium apparatus called Ca1. Ca1 utilized a single atomic beam, generated by an effusive oven usually held between 500 to 600 °C. The atomic beam passes through two apertures, one before and one after the Ramsey-Bordé (RB) zone. In the RB zone, a traveling wave laser is directed to intersect the atomic beam at four locations. After the RB zone is the detection region, where fluorescence is collected from either the 657 nm spontaneous decay or using the 431 nm blue cycling transition. This system allowed for great flexibility of the laser beam configuration from the long windows.



As to the remaining experimental basics of Ca1, the magnetic fields were initially applied to the Ca1 chamber with simple permanent magnets laid atop the vacuum chamber. Later solenoid coils were also tested. Vacuum levels typically reached the 10^{-7} Torr level (1.3×10^{-5} Pa). Numerous types of Ramsey-Bordé laser path creation were evaluated, including a (1) full double cat's eye retro-

reflector², (2) half cat's-eye and half large mirror retro reflector, (3) Large double retro-reflector, and (4) individually mounted mirrors aligned as retro-reflectors. Some of these are depicted in Fig. 4.2. None of the configurations demonstrated a clear advantage, though the alignment was less time-consuming with the cat's eyes. Compensation plates were also incorporated to the single cat's eye to correct for beam misalignment caused by passing the laser near the edge of the large lens.

Figure 4.2: A variety of methods were used to generate the two parallel pairs of anti-parallel laser beams needed for Ramsey-Bordé. A traditional double cat's eye was initially used. Degrees of freedom were eventually added until arriving at a fully adjustable laser configuration.



Optimization of the fringe signal for frequency stabilization, i.e. the goal of this experiment, actually relies on the interplay of two systems. The first is the atomic ensemble excitation, i.e. the quantum-mechanical possibilities afforded by the ensemble being interrogated. This is the world of optimizing quantum projection noise of the calcium atoms and making sure that the measurable atomic response has the best signal-to-noise possible. The second is that of the measurement itself, which converts the atomic response into a frequency correction. These two system components must work together for the system goals to succeed. For instance, a detector's quantum efficiency may require tens of photons per emitter to generate at least one electron per emitter. Thus, the for quantum projection noise of the system to be realized, the experiment had better be designed so that each signal atom sends many tens of photons at the detector. In this way, sometimes the

² An excellent resource with guidance on aligning cat's eyes for Bordé-Ramsey spectroscopy is given in [186], which is an excellent and very informative paper in its own right.

challenges of this engineering problem can be solved through once again taking a look at the hard physics of the situation.

I attempted to investigate most every experimental variable at my disposal to fully understand the intricacies of our atomic signal. The goal of these investigations was not necessarily to accurately measure the exact value of our system variables, but rather to explore the nearby parameter space for each variable, searching for clues as to how to improve the system stability. To organize this section, variables are roughly partitioned into subsystems, such as the red laser or electronic processing.

4.2 Interrogation (657 nm) laser for interferometry

There are roughly two orthogonal parameter spaces for improving the interrogation laser performance. One deals with the laser's spatial properties including collimation and beam profile at the atom-laser interaction zones. The other realm deals with the laser frequency properties, including mode content, linewidth, and nominal frequency. Both realms include the laser's phase at the four laser-atom interactions, which can be easily distorted by physical elements or fluctuations in the laser frequency. For Ramsey-Bordé spectroscopy, it is the phase of the laser interaction experienced by the atoms that determines the phase of the interference fringes, and so nearly every aspect of the interrogation laser must be considered to optimize the Ramsey-Bordé fringes. In addition to reducing the noise of these individual laser parameters to negligible levels, the most optimal values of these variables within feasible operating conditions had to be found.

Given that the Ramsey-Bordé apparatus consists primarily of the 657 nm laser system and the atomic beam, it was far easier to change the laser properties to accommodate our atomic beam than the other way around. Changes to the thermal atomic ensemble are far more cumbersome to implement given the large wait periods between breaks in vacuum that our apparatus required. Thus, the main degrees of freedom for improving the atomic signal lay in adjusting the interrogation laser. These properties will now be discussed individually and their optimization parameters given

with respect to our apparatus.

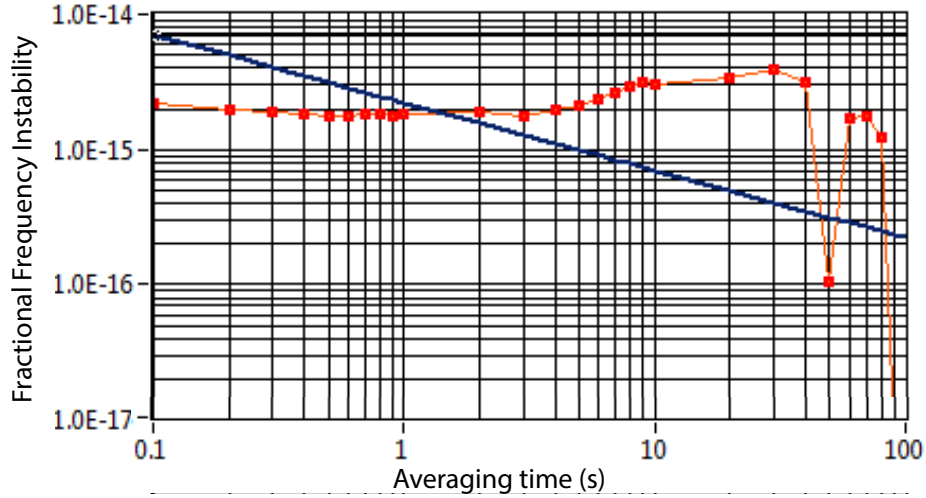
4.2.1 657 nm laser setup and pre-stabilization

During Ca1's use, the 657 nm Ramsey-Bordé interrogation laser began as an extended-cavity diode laser (ECDL) that was roughly tuned to the 1S_0 - 3P_1 transition via the ECDL grating angle and diode temperature/current. Fine tuning at the MHz level and below was then accomplished with acousto-optic modulation.

After ample optical isolation, a small portion of the ECDL light was sent to the cavity for PDH frequency stabilization, while the majority was then passed to a slave diode for amplification and passed through further optical isolation.

The cavity frequency stabilization was provided by the same ultra-low expansion glass (ULE) Fabry-Perot cavity for pre-stabilization throughout my time in the laboratory. The cavity had been around for quite some time prior to my arrival, and had previously been utilized in the calcium MOT experiments under Chris Oates at NIST. It was routinely able to reach frequency instabilities (measured via the comb laboratory against the superior Yb optical lattice clock primary cavity) given by the Allan deviation in Fig. 4.3. Typical drift levels are of order 1 Hz/s even with temperature stabilization. Optimized performance usually fell between $(2-3) \times 10^{-15}$ at 1 s. With linear drift removal, the cavity-stabilized 657 nm light could sometimes stay near the low 10^{-15} range for minutes, but without drift removal usually took off for the 10^{-14} 's rather steadily. Cavity PDH corrections were fed back to the initial diode, such that all light, including what reached the atoms, benefited from the cavity frequency stabilization.

Figure 4.3: Linear drift removed Allan deviation for the 657 nm cavity-stabilized laser frequency when running at optimal levels.



The properties of the pre-stabilized 657 nm laser did cause a decent amount of concern. Because this laser system is absolutely essential to resolve narrowband Bordé fringes, it was important to ensure that the 657 nm laser system was on its best behavior. At times the laser system suffered from a lack of robustness and would require re-alignment of laser paths or adjustment of temperature servos. In retrospect, creation of a far more reliable and less environmentally sensitive laser system would have saved many months or work spent maintaining, re-aligning, and trouble shooting.

The important electro-optic elements of note in the 657 nm laser system prior to arriving at the calcium apparatus include two AOM's, referred to throughout this dissertation as AOM1 and AOM2. AOM2 actually comes first in the optical path, and it is responsible for the calcium frequency corrections to the 1S_0 - 3P_1 ($m=0$) resonance. A small portion of laser light after AOM2 is sampled and sent to the frequency comb laboratory for measurement along a phase-noise canceled fiber optic path. The majority of the light is passed along and reaches AOM1 shortly before the fiber leading the atomic apparatus. AOM1 provides the frequency modulation used in our locking procedure and also actuates for phase noise cancellation along the the following fiber path.

4.2.2 Optical path of the Ramsey-Bordé laser

Our early attempts to resolve Ramsey-Bordé fringes relied on a counter-facing pair of cat's eye lenses, the traditional method of achieving the Bordé and similar geometries [186]. We also experimented with some of the established methods for correcting cat's lens aberrations via wedged optics [137]. The different methods we tested with success to create a single traveling wave optical retro-reflector are depicted in the earlier Figure 4.2.

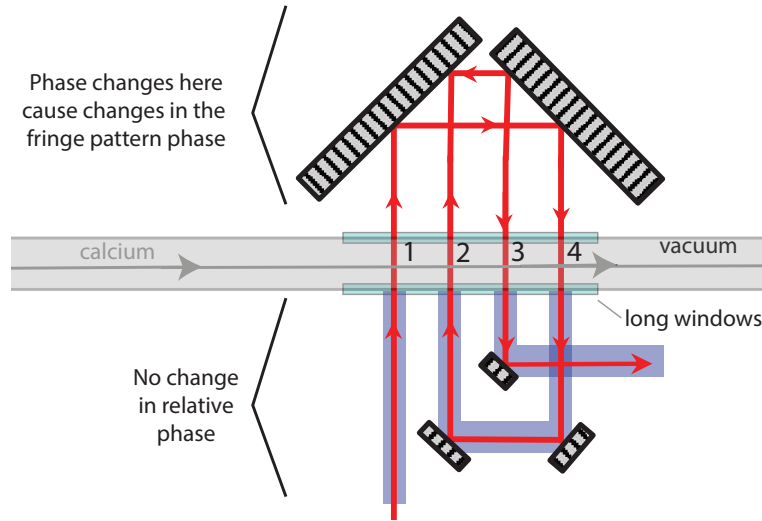
No significant differences were noted in the the Bordé fringes between the double cat's eye, single cat's eye, and full adjustable configurations, however the cat's eye simplified the alignment process. By testing these various optical path geometries, we were able to conclude that the basic retroreflection system would work satisfactorily, which made moving a fully constrained laser reflector into the vacuum chamber a reasonable next step so as to overcome the difficulties caused by fluctuations in the optical path length in air discussed next.

4.2.3 Air fluctuations along the optical path

Air currents generally contribute to noise in our calcium system through a combination of wavefront distortion, phase noise, and pointing noise. Unfortunately, quantifying these individual effects can be extremely difficult, though much work has been devoted to studying laser propagation through atmosphere and turbulent environments such as for LIDAR and defense operations [261, 47]. The pointing noise contributions are sufficiently small that, over the length scales in the lab, they cannot be measured using a high-resolution CCD, a task further complication by the large physical extents of our beam. While all effects of atmospheric turbulence are difficult to measure, it is fairly simple to discern whether or not the effect is a limiting one. For instance, extending the path length in air by 20% should roughly introduce 20% more phase noise, and pointing noise and wavefront deformities would also worsen. An obvious solution is to just shorten the path length, but there are minimum length requirements for the necessary optical elements. Of course, enclosures around the laser path can lessen the air currents and insulate the laser from some of these issues.

Ultimately, we found that air currents were a potential limiting issue with our first calcium apparatus, Ca1. In particular, the airborne optical paths after the first laser-atom interaction were found to be very problematic. Prior to the first interferometer interaction, slow changes of the laser phase relative to the sampling time do not cause much issue, as a global phase change is common to each atom-laser interaction, and thus no phase difference³ is seen in the fringes (see Eqn. 3.9). However, phase fluctuations occurring after the first laser-atom interaction can contribute directly to phase noise of our fringes. For our beam geometry, not all phase fluctuations along the laser path contribute to changes in the Bordé fringe pattern (see Fig. 4.4). However, our single traveling-wave beam geometry also requires a long path length that was difficult to shorten much more without reducing the Ramsey time, thereby compromising our fringe pattern linewidth.

Figure 4.4: The geometry of our Ramsey-Bordé laser (red) as it intersects an older version of the apparatus with vacuum chamber and calcium beam marked in gray. Slow phase fluctuations occurring in the lower half of the figure, highlighted in blue, do not contribute to an overall change in the Bordé fringe pattern, which has a phase given by $\phi_2 - \phi_1 + \phi_4 - \phi_3$. This is only valid for phase fluctuations over timescales much longer than our cycle time.



In addition to deformities from air particles, the 657 nm laser must enter and exit the vacuum chamber multiple times by passing through an elongated glass window, which can distort the laser

³ Of course, air fluctuations causing phase changes on timescales faster than the atomic transit time through the interferometer can still cause noise, but, for the sake of this introductory argument, I am ignoring this.

path and introduce phase variations with temperature, pressure, or other factors as the glass material changes. Calcium from the atomic beam would deposit over time, causing additional power loss with each pass. Furthermore, these windows were found to bow during pumping, and stress-induced birefringence potentially contributed pressure-dependent fluctuations in the optical path.

We were able to conclude that air fluctuations were likely the dominant noise source in the frequency stabilization system through a process of elimination. One advantage of abandoning the double cat's-eye lenses was that mirror optics on the phase-sensitive side of the vacuum chamber (See Fig. 4.4) could be more easily enclosed within a chamber. The total system performance was measured with and without a lid on the optics chamber, and the presence of the enclosure was found to correlate with an overall decrease in the measured frequency instability. A larger enclosure was then made around the entire 657 nm red laser path existing after a phase-stabilization sampling pickoff reference point. However, no significant improvements in the system instability were noted from this change. The chamber and larger enclosure were also flooded with helium (to benefit from its lower index of refraction) and tested, but the effectiveness of retaining a dominantly helium environment within the enclosure was doubtful and no changes to the system instability were noted. Additionally, intentional disturbances of air near the 657 nm laser beam were noted to be capable of extreme deformation or loss of the fringe features as viewed on an oscilloscope in real time.

Unfortunately, the literature seems to be a bit empty in regards to the phase, pointing, and wavefront distortions caused at this noise level and over such mundane laboratory conditions. Generally, if such problems are through to be a significant issue, then the beam path is moved to a vacuum environment, which is the exact course of action taken when designing the next generation apparatus, Ca2.

4.2.4 Angular deviations

Though the fully adjustable system of Fig. 4.2 (p. 99) was, by far, the most challenging laser geometry to initially align, it did let us fully characterize the angular sensitivity of the laser alignment. The laser steering mirror's directly before the first calcium beam intersection was carefully tilted while the locked calcium frequency was measured. The angular sensitivity was measured for both tilts in the vertical and horizontal direction. The laser profile during this test was taller than the atomic beam, allowing for some test of angular tolerances before the laser would begin to clip the atomic beam. Ultimately, we found a vertical angular sensitivity of 5.5 Hz per μrad and a horizontal sensitivity of 0.5 Hz per μrad . This difference in sensitivity makes sense given that the retro-reflector optics were designed to yield parallel laser paths regardless of the horizontal input angle, while the vertical beam could instead simply walk off further over the optical path length. These angular sensitivities were later utilized in determining the machining tolerances to which we wanted for the fully constrained, in-vacuum ULE retro reflector⁴.

4.2.5 Laser profile and Gaussian properties

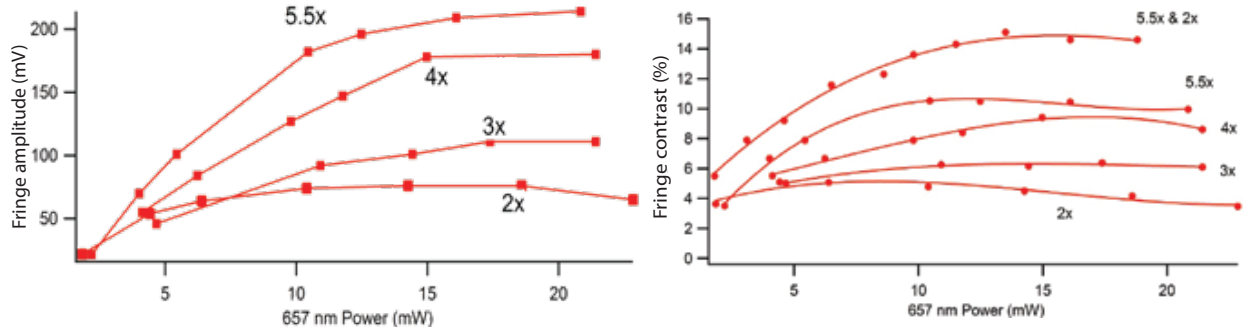
There is no generally accepted treatment of Bordé-Ramsey interferometry that accounts for the Gaussian laser profile. Additionally, the laser beam profiles over long distances, after experiencing multiple types of optical aberrations, are difficult to characterize. Because lasers exiting optical fibers are essentially Gaussian beams, the interrogation laser's properties along the optical path can be parameterized by its waist and wavelength. Of course, we also torture our poor laser beam profiles and stretch them into very narrow elliptical beams. To keep path lengths through air short, beam shaping is usually done with fairly short focal length lenses, which are quite sensitive to physical alignment and can quickly cause debilitating aberrations in the far field.

⁴ Somewhat intriguingly, we evaluated the retro-reflecting mirror setup, affixed in place, so as to see how parallel the beam and its retro-reflection were to each other. However, they were found to be far from parallel. They were found to be off in the vertical angle by more than a degree and in the horizontal by much more than the measured tolerance but less than a full degree. These deviations could have been caused by misalignment during transport. They also may have been important for compensating for angular deviations caused by other optical elements in the beam path. The large vertical deviation from parallel could have easily been caused by mounting the mirrors to the optical bench at a slight angle relative to the atomic beam and thus allowed us to compensate for it slightly.

So, while Gaussian approximations are still the best theoretical models for the two dimensions of the laser profile, they do not necessarily describe exactly what we wind up seeing in the laboratory.

As to the ideal laser waist at the atom interaction zones, we generally found that the narrower we could make our interrogation laser, i.e. the shorter the laser-atom interaction, the better our fringe contrast became. A chart of relative fringe contrasts for a sampling of ellipticity (height to width) ratios is given for various laser powers in Fig. 4.2.5. The leveling out of these curves shows the atomic response entering the saturation regime for the atomic and laser beam parameters we used at the time on Ca1. It is also clear that the atomic response had largely flat-lined by around 15 mW for these collimated laser beams.

Figure 4.5: Bordé fringe saturation properties for a range of laser profiles. The beam ellipticity ratios are listed next to their respective traces and realized with anamorphic prism pairs. Both clearly show advantages of using narrower waisted interrogation laser profiles. Top: Bordé fringe peak-peak signal measured on a photomultiplier tube for a variety of anamorphic prism pairs. Bottom: Later measurement with an additional 5.5x then 2x (11x total). This graph parameterizes the fringes by their size relative to the Doppler.



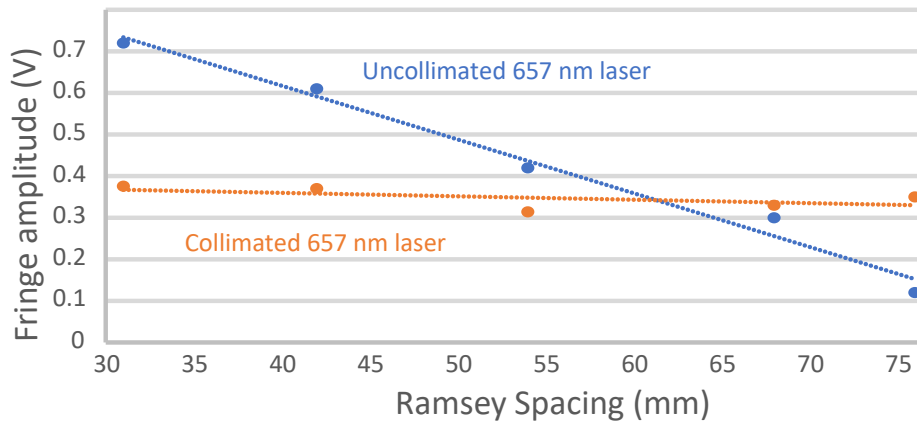
It is expected that as the laser waist is made smaller, that the atoms of our thermal beam will experience greater time-of-flight broadening. Through this mechanism, a larger transverse velocity distribution can be successfully interrogated. Of course there exists a limit to how small the laser beam width can be at the laser-atom interaction before the Bordé-Ramsey method breaks down⁵. However, for laser profile dimensions much larger than the laser wavelength, Bragg scattering

⁵ In this limit, the atom-laser interactions are better described by Kapitza-Dirac scattering process [147, 96], more commonly seen with stand-wave laser interactions instead of the traveling waves used here. With that type of scattering description, the atom can receive multiple units of $\hbar k$. Typically such conditions are obtained by relying on very short pulses, which are better understood with the energy-time uncertainty relation.

[176, 91] describes the interactions well. Bragg scattering of atoms and molecules can be readily achieved with either a traveling or standing wave configuration and has been seen in optical lattices [28] and BECs [241]. In this limit, only first-order scattering is seen where the change in momentum is restricted to $\hbar\vec{k}$, though the direction of $\vec{k}$ can vary for a realistic Gaussian laser source.

Considering the spread of wave-vectors in Gaussian beams enlightened a strange phenomena we routinely saw when adjusting the interrogation laser's collimation. I consistently found that the fringe contrast and peak-peak amplitude was maximized not when the laser appeared to be optimally collimated, but rather when slightly either converging or diverging. This was particularly obvious at smaller Ramsey spacings, where the uncollimated laser produced much larger fringes than the collimated option, as seen in Figure 4.6. The drop in fringe amplitude versus Ramsey spacing for the uncollimated laser is perhaps explained by the increasing inhomogeneities between the four laser-atom interaction zones when the laser traveled a much longer distance out of vacuum to obtain these spacings.

Figure 4.6: Measured variation in the fringe peak-to-peak PMT detector response with Ramsey spacing. Oddly, when the 657 nm interrogation laser delivered uncollimated light to the atoms, the fringe amplitude was able to far exceed that reached with well-collimated light. See text for more.



The reasons for the loss of fringe signal with a very well collimated beam most likely derive from the corresponding narrowness of available wavevectors. Given the reasoning laid out previously in Ch. 3.4.2, the FWHM of our atomic beam's Doppler profile of a few MHz far exceeds what should be accessible with only time-of-flight broadening (a few hundred of kHz). To address the entire

atomic beam, the 657 nm laser must be slightly uncollimated to provide a range of wave-vector orientations that account for transverse velocity Doppler shifts of some atoms. More discussion of this is provided in [242].

The Ramsey spacing chosen for the Ca2 apparatus was larger than what was even tested in Fig. 4.6, which led us to believe that a very well collimated laser system would be desired. However, when the optical path was entirely enclosed within vacuum, uncollimated 657 nm laser beams still produced the largest amplitude and highest contrast fringes as is discussed in the following chapter.

4.2.6 Power level

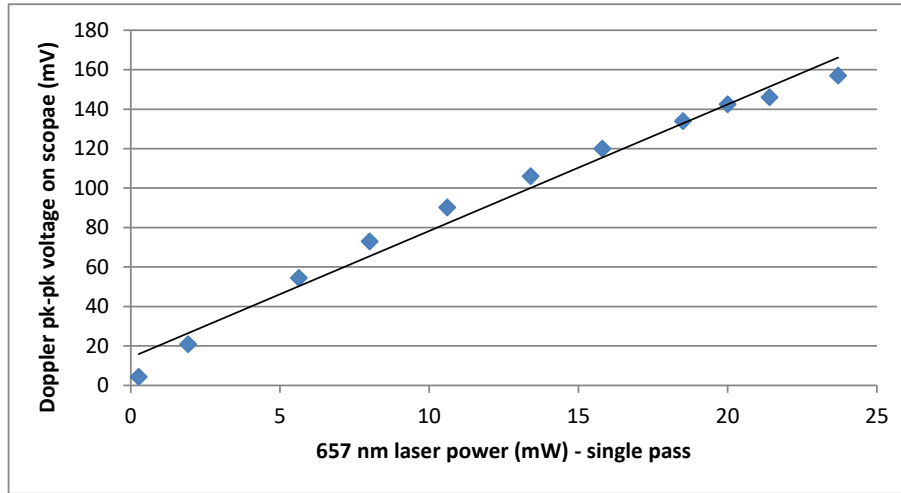
Both lasers employed - the red interrogation laser at 657 nm and the blue electron shelving laser at 431 nm, require intensity stabilization to avoid adding noise to our interferometer signal. For both, running them with power levels near saturation for the given laser beam profile area can greatly reduce the impact of amplitude noise on the fringe signal, as the ensemble sensitivity to power fluctuations drops. Of course, the laser power also couples in the pulse-area experienced by our atoms, which complicates matters slightly. The ensemble response to a fixed laser beam size was evaluated for a range of laser powers.

The 657 nm system (for an approximately 1 mm by 1 cm beam profile) saturated reasonably near 15 mW for the lower oven temperatures of our test apparatus, but, the higher oven temperatures often did not saturate until ≥ 20 mW. When available, we preferred to run at 25 mW, as this produced the largest fringes and still limited the calcium system response to intensity noise from the interrogation laser regardless of the oven temperature. Unfortunately, providing significantly more than 25 mW to the calcium system was very difficult. However, this was not too concerning because (1) our stability was not limited by red laser intensity noise and (2) the fringe amplitude and contrast largely plateaued or began to decrease slightly beyond this power range.

Additionally, a saturation curve is only valid for a given interrogation laser profile and oven temperature. As the oven temperature is increased, the atomic speed distribution shifts higher, causing the ensemble overall to experience less time in the laser fields, reducing the pulse area. A

plot of saturation behavior for fairly large 657 nm laser profiles (≈ 1 cm round beams) is given in Fig. 4.7.

Figure 4.7: Measured saturation behavior for an early choice in 657 nm laser profile. The gentle leveling off the atomic response can be seen at higher powers, but depicts that for the accessible range of laser power ≤ 25 mW, the atom ensemble did not exhibit clear saturation.

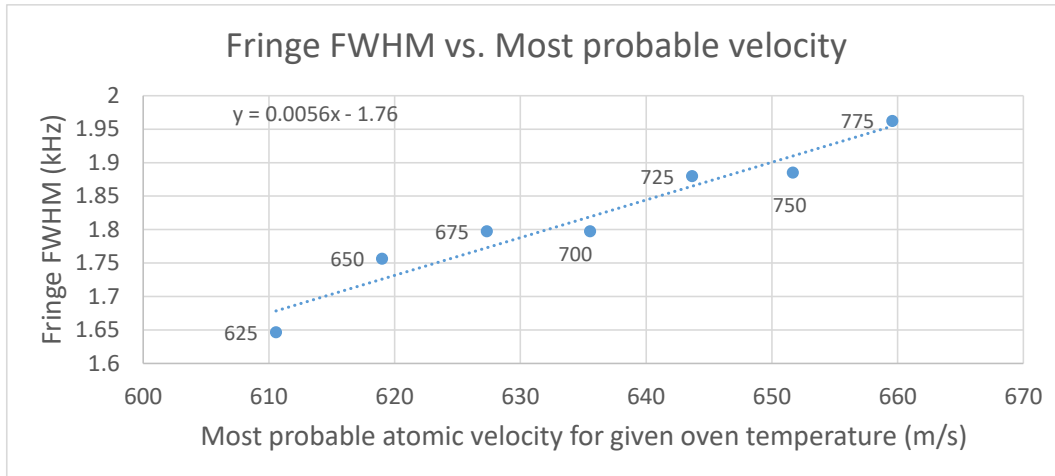


4.2.7 Spacing of laser-atom interactions

One of the most basic design aspects of Ramsey-Bordé spectroscopy is how long to make the dark time between interactions. In the extreme, the dark time can be made long enough to even achieve interference fringes with linewidths less than the natural linewidth of the transition being utilized[192]. However, as the dark time is made longer and the fringes linewidth decreases, the signal-to-noise begins to worsen due to spontaneous decay. Furthermore, the choice in oven temperature comes into play here, as faster atoms require more laser power to achieve the same pulse area but also experience shorter Ramsey times, thus the spacing can also be influenced by the power limitations of the interrogation laser in addition to the oven temperature. Thus, for optimum frequency discrimination, it was not immediately clear what the Ramsey dark time should be. Given all of the variables that couple into the optimum Ramsey-Bordé dark-zone length, some experimental observations were needed to form a better understanding of this multi-parameter space.

Of importance to our determination of the Ramsey spacing was various data we had collected when also exploring the impact of the interrogation laser's collimation, as was shown in Fig. 4.6. Though broader beam spacing created narrower fringes as is shown in Fig. 4.8, the loss in fringe contrast due to spontaneous decay and worsening laser imperfections over its length drove us to opt for a well-collimated laser beam with a Ramsey beam spacing of 9 cm, which, for typical oven temperatures (550-650 °C), yield fringes with FWHM measurements between 1.5 and 2 kHz. The data presented here is from the Ca2 apparatus, which could reach much higher oven temperatures than in Ca1.

Figure 4.8: The linear relationship between the fringe signal's linewidth and the most probable longitudinal velocity is displayed. Oven temperatures are given next to each data point in °C. This data was taken from Ca2, but the trend is relevant here.



The change in fringe linewidth with oven temperature was far less drastic than the change in flux with oven temperature, leading to overall improvements in the fringe signal-to-noise as the oven temperature was increased. This, combined with the fringe amplitude versus Ramsey spacing data of the previous Fig. 4.6, led us to believe that a future apparatus could benefit from Ramsey spacings beyond 7 cm to accommodate higher oven temperatures (thus faster atoms) while still producing fringes similar to those already characterized.

4.3 431 nm electron-shelving laser

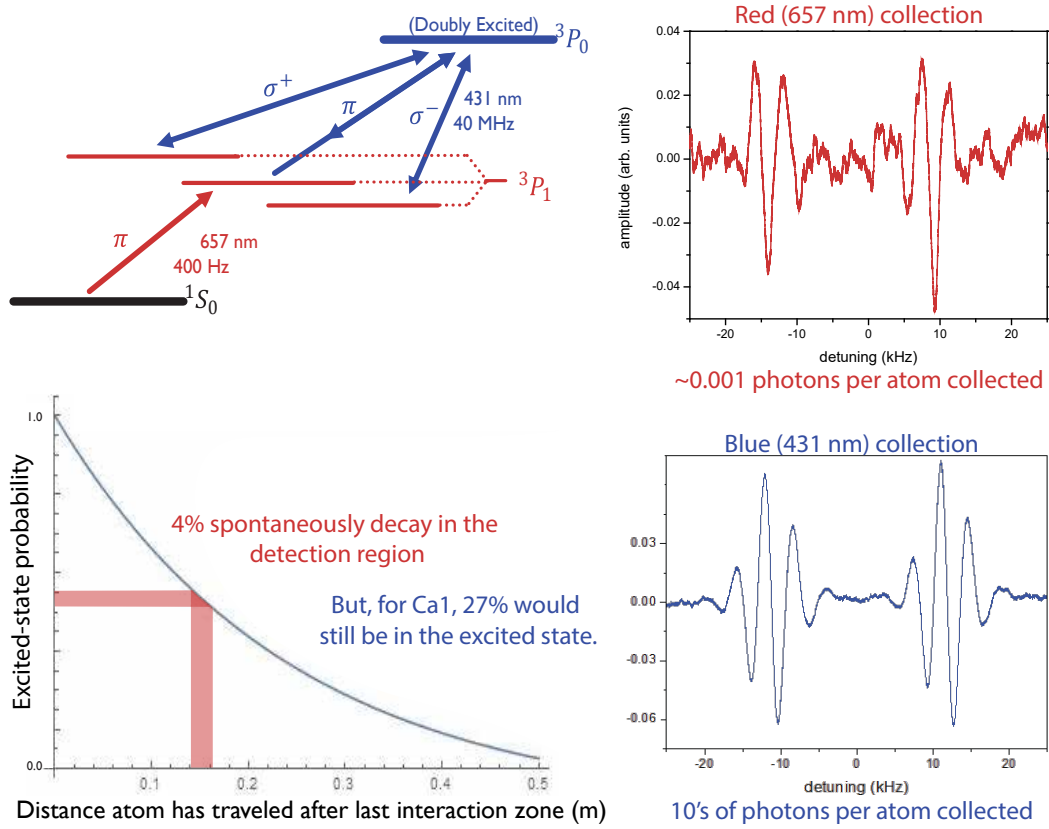
One of my first projects on the calcium system was to implement a frequency doubling cavity for the 862 nm IR laser the lab had purchased. This laser was intended to be tuned near the 431 nm $(3p^6 4s 4p) \ ^3P_1 - (3p^6 4p^2)$ doubly-excited 3P_0 transition [248], which connects from the excited state population of the Bordé interferometer. Electron-shelving for readout of ensembles in long-lived excited states is a routinely used procedure for calcium Ramsey-Bordé. However, the chosen cycling transition is normally from the ground state population on the $^1S_0 - ^1P_1$ transition at 423 nm [271, 145, 234], and it has been used quite successfully.

So why use the doubly excited shelving transition instead of the ground state transition? Both are much broader than the 657 nm clock transition (375 Hz), with linewidths of 34 and 31 MHz, respectively. Both are in the blue end of the spectrum and require similar techniques to generate the laser light, as outlined in the following subsection. They are primarily distinguished by final Bordé atomic state they couple to: excited or ground. In theory these population are roughly equal if the measurement can be made very shortly after the last Bordé laser-atom interaction and the Ramsey spacing is small, i.e. no spontaneous emission is allowed. However, this is not practical for a thermal beam experiment with the primary motivation of frequency stability. The thermal velocity distribution means some atoms move slowly enough to possess some non-zero probability of spontaneous decay from the interferometer, and we want a decently long interferometer that optimizes the frequency discrimination process for our calcium beam (Sec. 4.2.7, p. 110). Furthermore, any atoms in the ensemble non-resonant with our laser that are not appreciably excited by the interferometer remain in the ground state. Thus the ground state population has a larger fraction of atoms that are not correlated to the Bordé fringes. For this reason, a measurement of the excited $(4s 4p) \ ^3P_1$ population should possess more favorable signal-to-noise properties than the 1S_0 ground state, assuming all other factors are equal.

4.3.1 Red spontaneous decay versus blue electron shelving detection

Though spontaneous decay is an ever present issue as atoms traverse the interferometer, spontaneous decay after the atom's last interaction with the lasers can be used for measuring the excited state population. For illustrative purposes, it is easy to consider the spontaneous decay probability assuming the atoms were excited by the final laser-atom interaction. After this last interaction, there was some ≈ 15 cm before the atoms reached the ≈ 4 cm long detection region of the Ca1 apparatus. When collecting ensemble fluorescence, exponential decay predicts that about 4% of the excited atoms would decay where our detector might collect one of these spontaneously emitted photons. If instead a second transition at 431 nm was utilized, roughly 10,000 times more photons could be collected per atom on average, as depicted in Fig. 4.9. This electron-shelving enables access to the $\approx 27\%$ of the original excited state population. Furthermore, each excited state atom can be made to rapidly emit many hundreds of photons in this region, ensuring that every excited state atom in the region contributes at least one electron to the locking signal. More on difference between red and blue collection is given in Fig. 4.9.

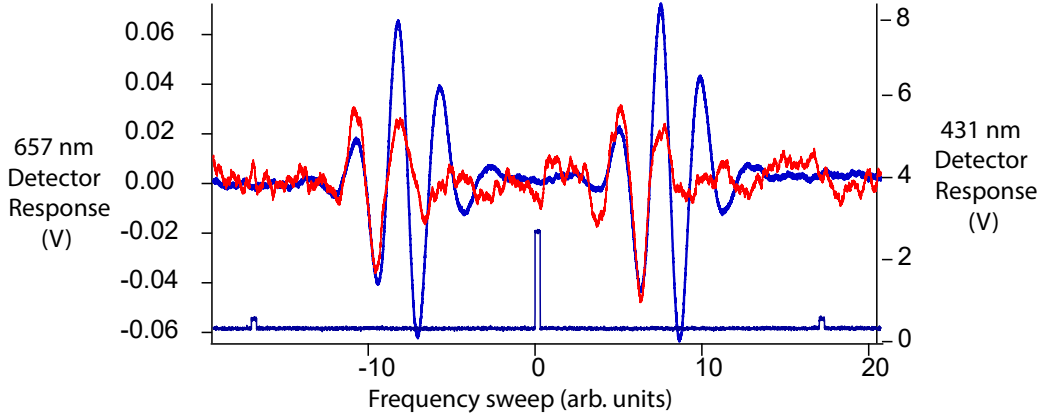
Figure 4.9: Comparison of the red (657 nm) and blue (431 nm) fluorescence collection methods tested on the previous Ca1 apparatus. The blue electron shelving technique obviously improves the measurement signal-to-noise, in this case by a factor of many thousands.



For a more direct comparison of the red versus blue photon collection methods of imaging the Bordé fringes, consider Fig. 4.10. There are a few subtle but interesting aspects of this figure beyond the obvious stated in the caption. First is that the fringe profiles appear very similar for the lower-frequency (left-hand) portion of the fringe envelope. This is somewhat expected, as the fringe packet asymmetrically peaks toward the lower frequency end of the fringe envelope. It is around this peak that the fringes interfere most constructively, and thus remains most visible for both imaging schemes. However, the higher frequency (right-hand) portion of the fringes visually differs between the blue and red imaging. The blue imaging result yields a slightly narrower linewidth than the red, perhaps because electron-shelving results in a heavier weighting toward slower moving atoms, which can emit many more photons than their faster moving ensemble cohorts. Furthermore, slowly

moving atoms are more likely to spontaneously decay before reaching the detection region of Ca1, and thus are perhaps under-represented in the resulting fringes.

Figure 4.10: Interference fringes resolved from collection of red 657 nm spontaneous decay (top) and blue 431 nm electron-shelving fluorescence collection (bottom). Both are displayed for two averages and depicted here primarily to demonstrate the stark difference in signal to noise. Also of note is the relative alignment of the two signals in frequency space. See text for further discussion.



4.3.2 Optimizing 431 nm fluorescence cycling for Ca2

Though the advantages of the 431 nm fluorescence cycling are obvious, some additional experimental tweaks were required to ensure this method worked. Most obvious is the loss of atoms from a pure 3P_1 ($m=0$) – 3P_0 doubly excited state cycling transition, as the doubly-excited atom can decay freely into any of the lower $m=\pm 1,0$ triplet states. However, a combination of applied magnetic field and polarization-rotated 431 nm light are able to roughly balance the excitation probabilities of the three triplet states for the duration of our measurement, and no dark states noticeably formed. The blue laser polarization is rotated with a half-wave plate just prior to entering the vacuum chamber so as to provide the mix of polarizations to the atomic beam that elicits the largest response. A brief test was done to ensure that the signal size was not limited by inefficient cycling or partial dark state creation. The blue light prior to the detection region was passed through an EOM and polarization modulated. The rapidly mixed polarization from the EOM should suppress dark state creation, but it also severely limited the transmitted power

at 431 nm. The modulated polarization light's resultant fringe signal was compared to a static linear polarization held at an angle to the magnetic field axis such that the atomic fluorescence collection signal was maximized, with each signal compared for equal 431 nm input power levels. No substantial difference was seen between the two cases, but the greatly reduced signal-to-noise at such lower blue power levels also made the comparison more difficult.

The Ca2 apparatus was designed to best take advantage of the atomic beam and the 431 nm blue imaging system. The blue detection zone is made to be as close to the last red zone as possible - in Ca2 this distance is shortened to just under 6 cm from the previous 15 cm. At this closer distance we would expect >80% of the excited state atoms to not have spontaneously decayed were all other factors the same.

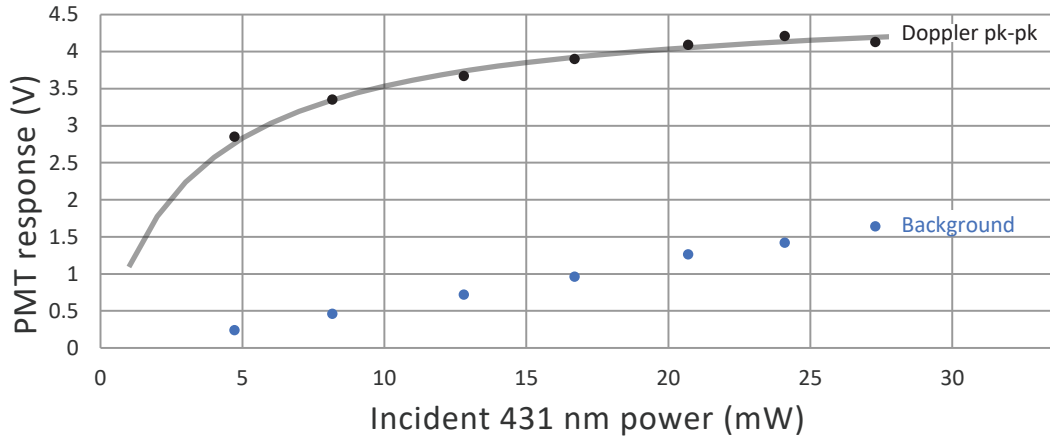
4.3.3 Blue power

The blue saturation intensity was measured for the blue laser profiles used with Ca1. The laser power needed to reach saturation was formally calculated using the conventional response function

$$\text{Atomic response} \propto A \left(\frac{P/P_0}{1 + P/P_0} \right), \quad (4.1)$$

where A is a scaling constant, found to be 4.7 in this case, P is the incident power, and P_0 is the power level where saturation is reached. Though the saturation power was found to be 3 mW, we try to run well into saturation to reduce the calcium system's sensitivity to blue intensity fluctuations without overloading stray background levels, leading to running the blue laser around 14 mW of incident power at a minimum.

Figure 4.11: The top graph displays the experimentally measured atomic response as a function of the input 431 nm laser power. The background level, which is independent of the red laser frequency, was easily recorded as the input laser power also increased. The fit to our Doppler pk-pk amplitude was used to calculate the power needed to reach saturation. See text for more.



4.4 Fluorescence collection

For the best use of the atomic signal, ideally we want to collect as many signal photons as we can while avoiding any non-fringe correlated scatter. Though the most straight-forward way to collect the atomic signal is to simply place a photo-detector downstream from the last laser-atom interaction, this is one of the least efficient ways to collect fluorescence. Photons are only emitted as an excited-state atom spontaneously decays, and we only collect ones that are emitted not only in front of the detector but also in the direction of our collection optics.

4.4.1 Solid angle collection

The solid angle is determined by the collection optics we utilize and their proximity to the point where the calcium and blue laser beams intersect. To maximize the solid angle, the optics are located as close to the fluorescence source as is practical. This required the use of very short focal length optics with the largest numerical aperture we could find. On the far side of the beam from the detector, a mirror is placed with its focal point roughly at the atomic beam. A lens on the close side of the atomic beam then condenses the atomic fluorescence onto a detector's active

area.

For the red 657 nm photon collection, the atom emits in a dipole pattern oriented relative to the imposed magnetic field. The resultant probability of collecting a red photon emitted near the focus for our optical elements turned out to be 16%. Meanwhile, emission from the 431 nm cycling transition is uniform and we collect a bit under 11%. Of course, optical elements are not perfect and neither is the fluorescence condensation onto the photodetector, so the actual values could be lower than this. Though the inherent dipole form of the 657 nm collection is more efficient, hundreds more photons are emitted per atom in the 431 nm case, far outweighing the slight solid angle disadvantage.

Though generally increasing solid angle collection is a reasonable way to increase the signal-to-noise of atomic signals, there is a limit to its advantage. Once every atom is effectively contributing at least one electron to the photoelectric current, then further increasing the solid angle will not continue to improve the system's stability performance, as the quantum projection noise will dominate over the photon shot-noise. At this level, fluctuations in atom number of the atomic beam can dominate the calcium electronic signal noise, assuming that all other noise sources that impact the atomic excitation levels are sufficiently suppressed, like intensity, phase, and pointing noise of the 657 nm laser.

4.4.2 Avoiding unwanted photons

The blue electron shelving technique that showed superior signal-to-noise also created a problem of significant background scatter. When mirror and lens imaging combinations were originally tried, we were still using an older version of the calcium apparatus where the atomic beam after the interferometer was enclosed within a glass walls. However, the unwanted reflections and scattering from the glass surfaces greatly increased the background we collected. To collect more signal photons, flat mirrors were mounted to the sides of the imaging cell, but the collection of additional scattered photons stopped the signal-to-noise from improving much.

Background scatter and photons uncorrelated with the fringe pattern are undesirable for a

few reasons. First, they raise the total number of photons incident on the detector, raising the detector's shot noise level, which makes for a noisier electronic signal. Second, they increase the locking system's sensitivity to intensity fluctuations of the contaminating laser source. So, for example, if the 431 nm scatter background is 50% of the total signal level, and it fluctuates by 10%, then photodetector's output signal fluctuates by 5%. But if the blue background was only 10% of the total signal, then the same fractional fluctuation causes only 1%. Red background scatter is easily filtered from our detectors, but it can still cause loss of signal from incoherent photon-atom interactions. Of course, these arguments assume that the atoms of the thermal beam are being interrogated by the 431 nm laser very far into saturation such that the atomic response does not change appreciably with blue power fluctuations. Lastly, any blue photons that make it into the central vacuum chamber where the Bordé interferometry takes place can then cascade through the atomic beam causing decoherence of the fringes.

4.5 Atomic beam generation and shaping

Many different aspects of our atomic beam generation system were explored before settling on final attributes for the updated Ca2 version of the experiment, including use of microtubules to better collimate the atomic beam, burnoff apertures to reduce clogging problems, a formalized 'oven turnoff' process to reduce calcium blockages, and various tests of oven temperatures.

4.5.1 Atomic flux measurements and oven temperature

Measuring the flux of an effusive thermal atomic beam with spectroscopic methods is not always an easy task, as it can be difficult to address all of the atoms given their sizable Doppler shifts. We performed numerous measurements of our calcium atomic beam flux, with most agreeing that the atomic flux of our beam at 660°C falls in the mid 10^{10} to mid 10^{11} atoms per second. At 760°C, the flux was conservatively estimated to be between mid 10^{11} and mid 10^{12} atoms per second.

These flux measurements were carried out by approximating our atomic flux from the am-

plitude of our PMT detector response. The PMT response was used to estimate the number of photons that hit the detector surface by first measuring the detector's response function for our specific wavelength. Neutral density absorptive films with independently measured transmission rates were used to create a response curve for each PMT used in these measurements, which easily allowed conversion of the amplified PMT voltage to incoming optical power at the relevant wavelengths (431 and 657 nm). The power is readily related to the number of photons per second that collide with the detector. From there, transmission and reflection efficiencies of the relevant optics were used along with the calculated solid-angle collection (p. 117) to yield the number of photons given off by the ensemble per second within our detection region. For red photon collection on Ca1, we expect only 4% of the excited state population to spontaneously decay where our optics can collect their single photon, whereas the 431 nm measurement repeatedly cycles many photons per atom from the approximately 27% of atoms not yet spontaneously decayed from the excited state. For the average speed calcium atom, a photon is emitted about every 12 ns when driven on resonance, resulting in about 550 photons being emitted in our detection region.

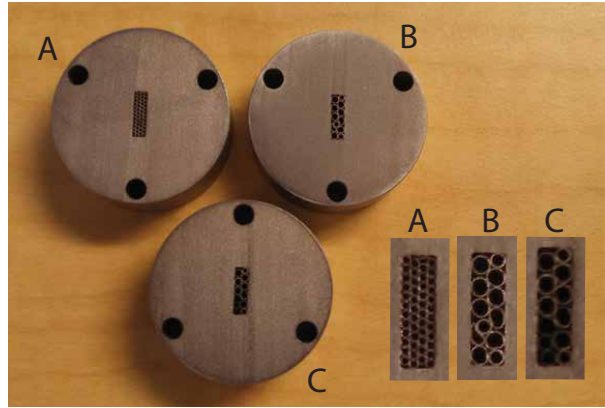
These flux calculations were unsurprisingly lower for the 431 nm measurements than the 657 nm ones. Firstly, the assumption of roughly 50% of the ensemble being in each the excited and ground states directly after the interferometer is obviously flawed due to spontaneous decay and imperfect excitation of the atomic velocity distribution. Furthermore, during the 431 nm fluorescence cycling process, the atoms emit photons isotropically, but they are stimulated by the blue laser in a directional manner. This causes the excited state atoms to receive a net momentum kick in the direction of the laser wave vector and can cause them to cycle photons less efficiently as a Doppler shift develops.

Lastly, these flux measurements are obviously dependent on the size of the atomic beam collimation apertures and the oven temperatures. Of course, changing the oven temperature to increase the flux has other consequences on the velocity distribution as discussed in previous chapters. Despite these additional consequences, measurements indicated that increasing the oven temperature would be a straight-forward way to improve the atomic system's signal-to-noise.

4.5.2 Microtubules

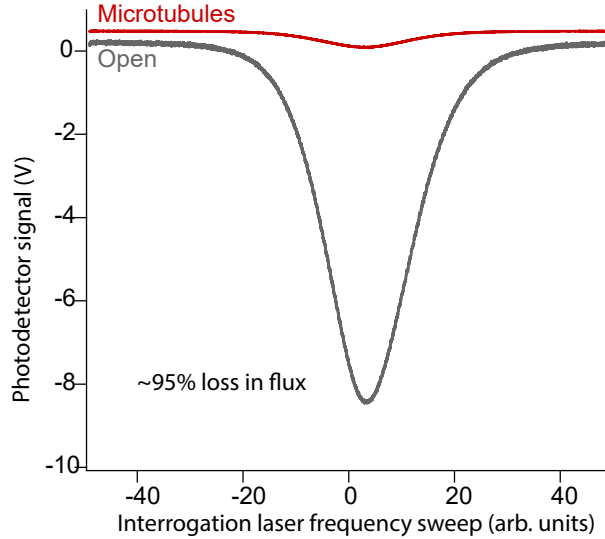
To optimize usage of the atomic beam, we explored methods of shaping the effusion pattern emerging from the oven via microtubules stacked within the oven nozzle. Photos of these microtubules can be seen in Fig. 4.12.

Figure 4.12: Three example oven nozzles packed with differing sizes of microtubules. The microtubules were hoped to improve the atomic beam collimation such that perhaps a larger atomic beam could be used without significantly widening the transverse velocity distribution. However, we found that the apertures largely responsible for transverse velocity distribution given the experimental dimensions here.



Unfortunately, the microtubules did not produce the desired results. As seen in Fig. 4.13, the huge loss in atomic flux negated any benefit they may have provided. The original hope had been that the total flux emitted from the oven would drop while the flux portion that would make it through the fine collimation apertures into the far detection zone would remain unchanged, perhaps up to the loss in aperture area.

Figure 4.13: Two single-pass laser-atom Doppler sweeps with and without microtubules for otherwise comparable conditions. Misalignment from the oven could have also contributed to making this discrepancy appear worse than it actually is, but we measured a great loss in atomic flux for the same oven nozzle aperture footprint when stuffed with microtubules.



Though we found that microtubules did not benefit the experiment, the way in which these channels were implemented could have also played a role in their ineffectiveness. If the nozzle of the oven, packed with the microtubules, were to be angled away from the far aperture just before the blue zone, it could be that we were effectively blocking a good deal of the atomic flux we wished to image. Thus, another advantage of using an open oven nozzle is that it is less sensitive to misalignment of the atomic beam apertures and nozzle. An open nozzle may then also reduce the impact of environment factors on the apparatus, such as thermal expansion caused by the ovens at many hundreds of degrees Celsius.

4.5.3 Clog reduction

Another concern with the atomic beam is one of more practical importance - clogging. Though we like to pretend otherwise when modeling the Bordé system on a thermal beam, there is actually an enormous amount of wasted atomic flux from our ovens. The 3-5 grams of dendritic calcium loaded into the ovens mostly winds up coating the interior surfaces of the vacuum chamber. One way to mitigate this is through various baffles and apertures that physically block calcium

atoms with unwanted velocity components. Those atoms usually wind up on the interior of our apparatus. Pictures of some calcium buildup and clogging are shown in Fig. 4.14.

Figure 4.14: A variety of the types of clogging experienced by our calcium system both on the oven nozzles and on the atomic beam collimation apertures.



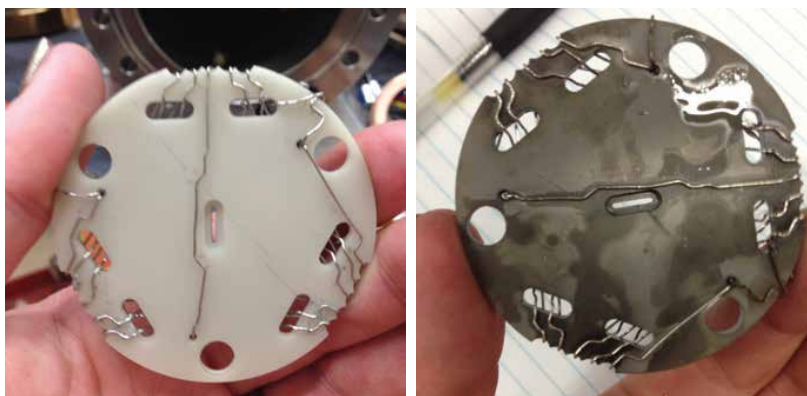
Clogging is a frustrating issue when attempting to run a precision system over many months. For our experiment, there was no way to visibly see a clog, and so first laser malfunctions, magnetic field problems, and detection issues had to be ruled out. If a clog (or absence of calcium) was concluded, the vacuum chamber would have to be opened, cleaned, and then brought back under vacuum again - a process usually taking more than a week.

Clogging was mostly confined to one of two locations. First, the nozzles on the oven would often clog. This was most likely due to lazy protocols regarding the oven on-off cycling. Often large clogs of this nature were preceded by periods where the calcium ovens were run hotter than normal, and then allowed to rapidly cool while coming back to temperature. In theory, this does not present a problem, but the higher thermal losses on the nozzle end of the ovens caused it to cool down faster than the bulk of the crucible and calcium atoms. The atoms would then begin to stick to the microtubules, eventually forming large protrusions that greatly reduced the flux, sometimes to levels where we thought calcium had simply run out.

Though the nozzle clogging could be remedied by simply paying careful attention to the oven controls over the nozzle compared to the main crucible temperature, clogging of the collimation apertures is far more difficult to overcome. One solution explored to stop aperture clogging was

through suddenly heating the aperture to very high temperatures and then allowing it to dissipate heat through the vacuum chamber walls so as to avoid large temperature swings of the apparatus. The aperture material ultimately chosen was aluminum nitride (AlN), which is a good thermal conductor yet a terrible electrical conductor. Other materials we tested include graphite and stainless steel, but the AlN with nichrome was by far the most effective. The AlN aperture plate was then machined as in Fig. 4.15, and nichrome wire was threaded in a pattern such that it did not make contact with itself. With a supply current of 5 Amps, the aperture plate, as measured by a suitable thermocouple, could reliably be brought to more than 500 °C after two to three minutes.

Figure 4.15: Prototype burnoff apertures, before and after testing in a vacuum chamber.



The use of the burnoff aperture was more complex than it wound up being worth, and we did not have the same aperture clogging problems on Ca2 that we saw in earlier versions of the apparatus. Thus, there seemed no reason to implement burn-off apertures in Ca2.

4.5.4 Atomic beam width and fringes that like uncollimated light

As has been discussed elsewhere in this dissertation (Ch. 3.4.2.1), the calcium experiment benefitted from use of slightly uncollimated 657 nm laser beams. To reduce the apertures such that the time-of-flight broadening would be broad enough to account for the expected transverse velocity distribution, it would take apertures only tens of microns wide to satisfy this condition. Such narrow apertures have been successfully demonstrated on thermal beams [3], but at the cost

of greatly reduced atom number.

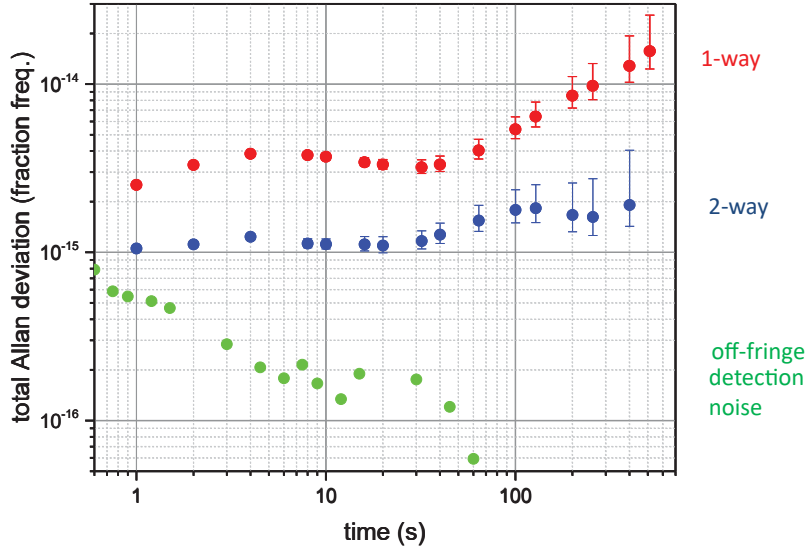
When adjusting the collimation of our 657 nm interrogation laser, I routinely found better fringe contrasts with slightly convergent or divergent beams. Starting with the most divergent beam for which we could still resolve fringes and bringing the laser towards collimation sees the fringe contrast progressively increase. However, very close to collimation, the fringe contrast suddenly drops. The signal-to-noise worsens and the fringes can disappear entirely. As the beam's focus is then brought in from the very far field, the fringes appear again, the contrast sharply increases before again slowly decreasing.

Though we were able to optimize the Bordé laser's collimation to optimally interrogate our atomic beam, it is not clear what atomic and laser beam dimensions would maximize the measurement signal-to-noise. It might be interesting to explore the far limits of the atomic beam extent possible for a very uncollimated, high power laser. A more thorough study of the atomic beam dimensions and obtainable fringe contrasts for various laser divergence levels would be particularly enlightening, though characterizing the laser divergence of an abnormally shaped Gaussian beam close to collimation can prove difficult, as can adjusting the collimation lens in small enough intervals to prove useful. These results also suggest that use of a much broader atomic beam may provide a useful route to further increasing the atom number, so long as the first-order Doppler shifted atoms are still contributing coherently to the fringe signal.

4.6 Laser reversal vs. two ovens

Though the Bordé method is quite powerful, it can still suffer from residual Doppler effects and fluctuations of the relative phase between the four interactions. These effects have been known for quite some time, and they are typically solved via some sort of $\vec{k}$ -reversal, i.e. laser beam reversal, process [119, 138]. We initially implemented a laser beam reversal system using a MEMS fiber switch, which could direct fiber coupled light into one of two output ports with a high switching rate. This method proved successful enough, as shown in Fig. 4.16, to encourage further work on utilizing multiple Bordé signals, but instead with two atomic beam sources over $\vec{k}$ -reversal.

Figure 4.16: This plot shows the differing behavior found when working with (2 way) and without (1 way) the $\vec{k}$ -reversal method for calcium Ramsey-Bordé frequency stabilization. For the Ca1 system, wherein the Bordé laser path was largely through air, the 2 way lock showed improvements at all timescales ≥ 1 s., approaching the measured off-fringe amplitude noise floor.



Moving to dual ovens on Ca2 instead of laser beam reversal as used on Ca1 was largely due to the difficulties in obtaining equivalent fringe signals from both laser orientations in the $\vec{k}$ -reversal. When working with Ca1, we found that the fringe signal is extremely sensitive to changes in the interrogation laser collimation, size, and orientation, and thus obtaining comparable fringes with two distinct laser paths was very challenging. Because the fringes from each direction of the laser reversal process wound up with different contrasts and possessed slightly varying shifts relative to each other, we were dissatisfied with the ultimate locking and drift result. Thus, the decision was made that the next generation apparatus would rely on two counter-propagating atomic beams instead of two sets of laser interactions.

Regardless of the way in which we compensated for residual Doppler shifts of the Ramsey-Bordé alignment, it was immediately clear that the use of conjugate Bordé fringes improved the long-term instability of the calcium-corrected laser frequency.

4.7 Magnetic fields

The magnetic fields used for Ca1 mostly relied upon permanent magnets placed externally to the system. Initially it was easiest to simply place small piles of standard refrigerator-strength magnets on top of the Ca1 chamber with one magnet source placed above each laser-atom interaction. Later, two large loops of magnetic wire were placed on top of and below the calcium vacuum chamber, supplying a large, more uniform Helmholtz field over the entire interferometer region. In the Ca2 apparatus, the calcium beam was located far from the chamber exterior, and thus neither of these solutions was appropriate. Instead we utilized multiple solenoid coils within the vacuum chamber.

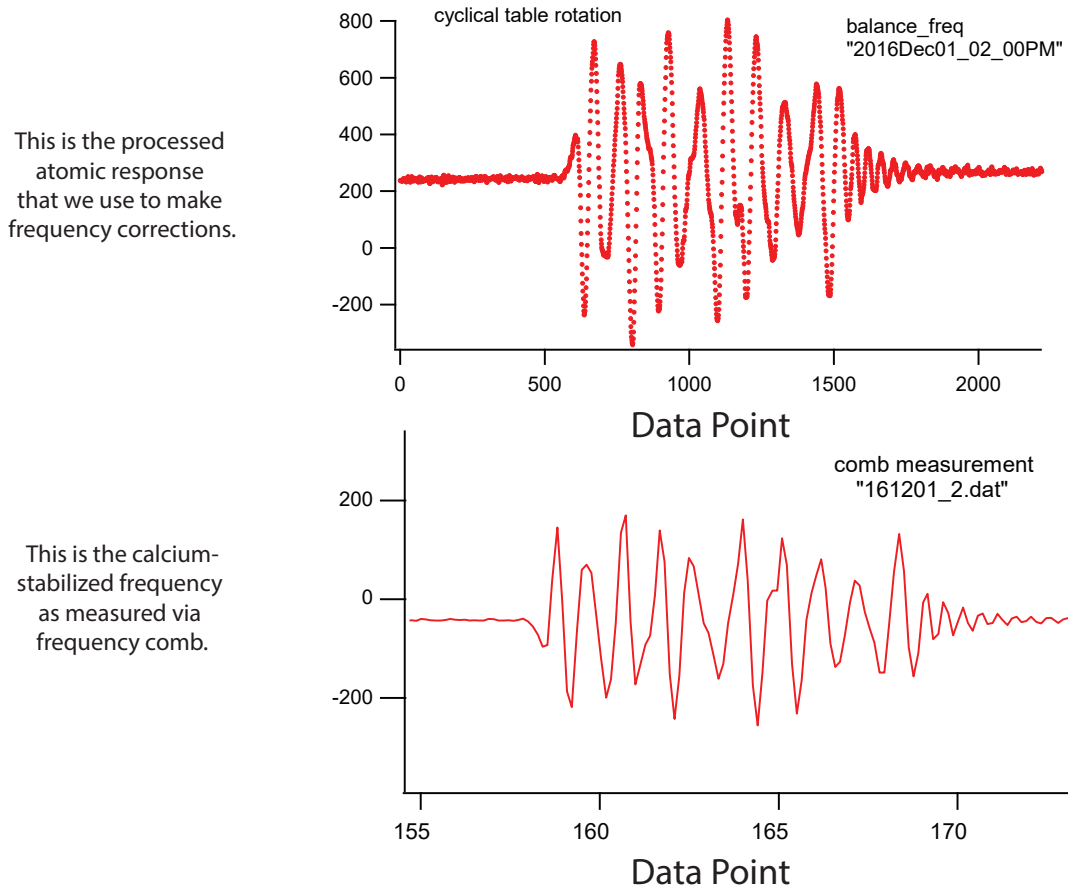
As to the magnetic field in the blue imaging zone, we observed that its magnitude and orientation were largely unimportant, so long as the appropriate polarization was found for the incoming 431 nm laser source. There was a great deal of flexibility here, and the signal was typically optimized by adjusting both the magnetic field magnitude and the laser polarization until the atomic fluorescence collection was maximized.

For all magnetic fields utilized here, the field variation over the atomic beam is slow enough that we believe Majorana spin-flip transitions [43] to be negligible. Such spin-flip transitions are typically suppressed in the adiabatic limit. When exploring the placement of magnets on Ca1, the addition of magnets to the dark zones between laser fields was not found to impact the resulting spectroscopic signal noticeably. However, the additional field component would have made the magnetic field gradient even more gentle. Were spin-flip transitions to be significant, the smoothening of the magnetic field would have reduced their probability. As no signal improvements were measurable, these transitions were not further investigated. Additionally, no significant differences were observed between the Ca1 results when using individual permanent magnetics versus the large solenoid field encompassing the interferometer region.

4.8 Angular rotations

Angular rotations of the apparatus were not intensively studied, but we did find that the system possessed a clear response to them, as seen in Fig. 4.17. The rotations were caused by firmly pushing the corner of the optics bench such that it rotated around an axis perpendicular to the bench surface. However, linear pushes and shoves of the optics bench were not nearly as sensitive, though their impact was difficult to discern without finding a way to expose the calcium apparatus to more than one type of table motion at a time.

Figure 4.17: We found that the calcium system showed clear response to rotations of the floated optical bench upon which the apparatus was seated. Such benches are commonly used to suppress vibrations, but we found the same instability performance with and without the optics bench floated, i.e. vibrations were not a limiting source of noise. Thus, we routinely ran the system with the bench grounded so as to avoid accidentally bumping the table and causing frequency excursions. The much lower measurement rate of the comb causes the differences between our frequency corrections (top) and the measured frequency excursions (bottom).



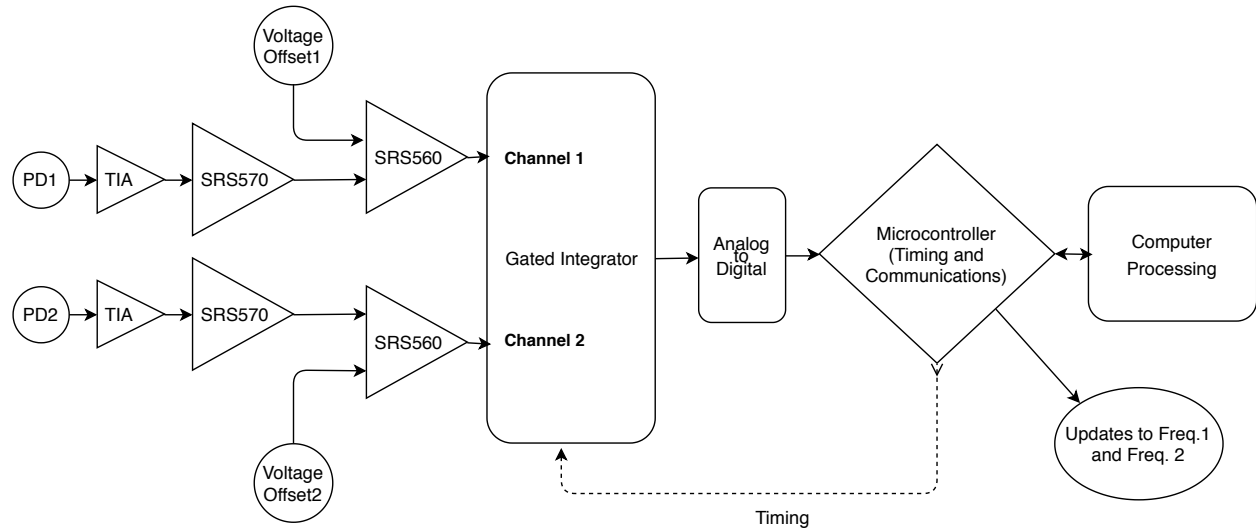
4.9 Electronic locking Systems

This section details the electronics system used to stabilize a laser's frequency to levels never before achieved by a thermal ensemble. The previous sections described motivations for the Ca2 system's main chamber and physical considerations, i.e. the 'what' portion, while this section focuses more on the 'how' aspects, i.e. how the Ca2 system is actually utilized in the experiment. Thus, an overview of the non-standardized electronics processing equipment will be given, and the locking procedure will be discussed. Photodetection will be discussed along with basic signal

processing considerations. The discussions here will be relevant for the following chapters when discussing sources of noise and limitations of our system performance.

The general flow chart of the electronic signal processing is given in Fig. 4.18, shown for two independent photodiodes, as were used in the Ca2 system. For the Ca1 system, only a single photodiode was needed, and thus the second gated integrator track was absent. For most of the time Ca1 was used, the photomultiplier tubes (PMTs) with built-in trans-impedance amplifiers (TIAs) output voltages already within range of the gated integrator, thus the SRS pre-amps were not needed. The figure caption provides the general purpose of each component, with more detailed information provided in subsequent subsections.

Figure 4.18: The collected fluorescence is converted to an electronic signal by a photodetector (PD) and immediately sent to a nearby trans-impedance amplifier (TIA). Subsequent low-noise current (SRS570) and voltage (SRS560) amplifiers, in addition to a low-noise voltage offset, prepare the signal to fit within the electronic window of the gated integrator. The signal is then converted to a digital bit value and processed to provide frequency corrections to the AOM-controlled locking parameters called Freq1 and Freq2. The entire process is coordinated by the micro-controller unit (MCU), which is responsible timing dissemination to relevant components, communications of the digitized values to the computer for visualization and human interface, and local processing of the digitized data to calculate and implement frequency corrections.



4.9.1 Photodetection and electronic signal processing

A variety of electronic subsystems are needed to go from a fluorescence measurement to a frequency correction. This all begins with the photodetector. For most of this work, we utilized photomultiplier tubes (PMTs) for our photodetectors. Immediately after the photodetector lies a built-in transimpedance amplifier (TIA), which converts the fluorescence-proportional PMT current into a reasonable voltage. The properties of the TIA are important for determining the electronic noise properties of the converted atomic signal. Additionally, a larger output signal size from the TIA is less susceptible to unwanted electrical pickup.

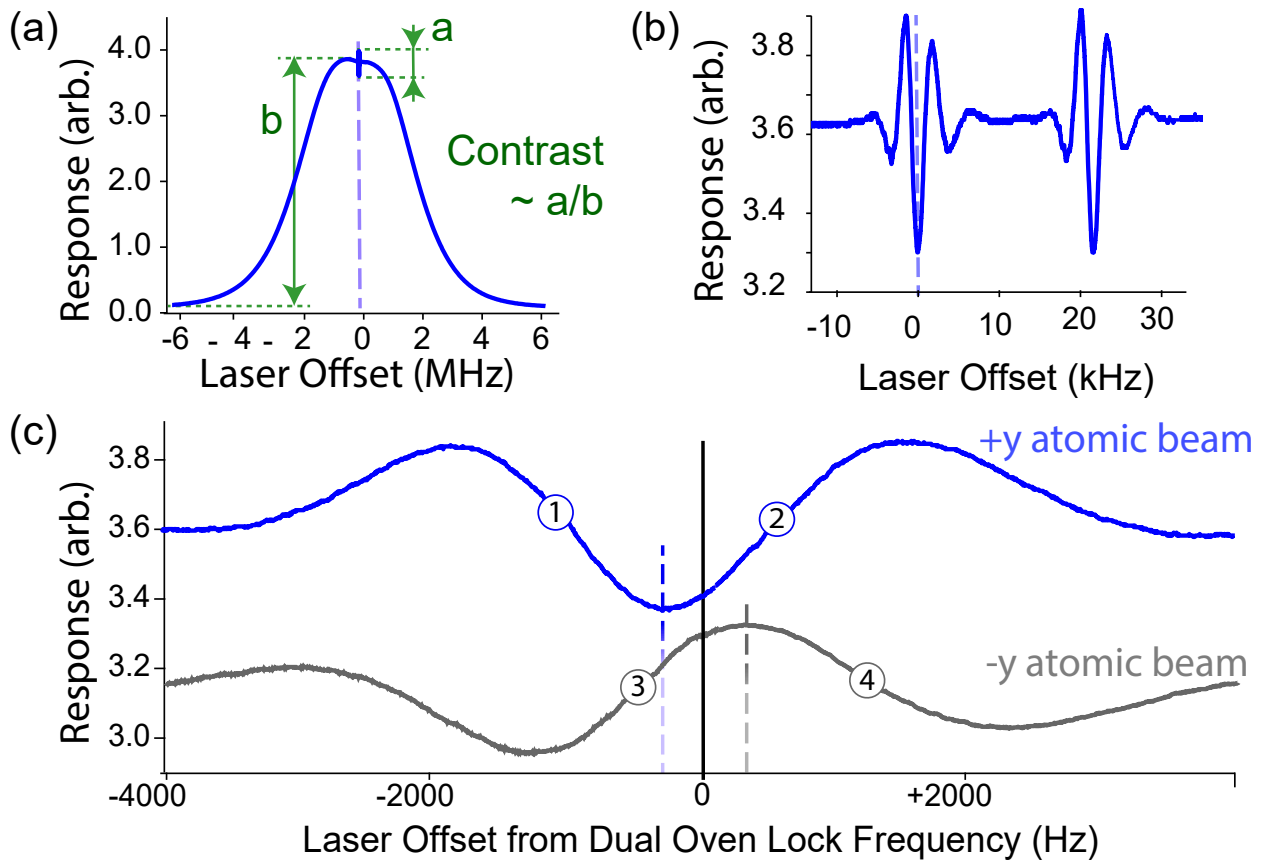
The photomultiplier tubes originally utilized worked well, but slow drifts caused by changes in the PMT responsivity and/or the transimpedance amplifier made it difficult to work with these detectors within the narrow voltage window allowed by the pre-made analog-to-digital converter. Given the large amount of atomic fluorescence collected after implementing 431 nm electron-shelving detection, the PMTs had to be run with gain levels well below the manufacturer specs. To overcome the PMT drifts, the voltage offset fed into the SRS560 pre-amp could be adjusted so as to keep the modified atomic signal within the processing window. Such adjustments typically had to be made every 30 seconds to a minute or so at first, but with better isolation of the PMT module, adjustments were reduced to every few minutes. Despite this partial solution, the offset adjustments sometimes still induced flawed frequency corrections that degraded the system's measured frequency stability over time.

The atomic signal is next processed by low-noise pre-amps, an SRS570 (current pre-amp) and SRS560 (voltage pre-amp), to bring it within the pre-chosen range for our gated integrator. The current pre-amp was only needed when using photodiodes on Ca2, but both Ca1 and Ca2 used the voltage pre-amp. The gated integrator is fed timing information from the micro-controller, and it accumulates the electronic signal for a set period of time. After the time is up, the gated integrator holds the final voltage while the analog to digital conversion samples it. This sampled voltage is converted to a number between 0 and 2^b where b is the number of bits available from

the analog-to-digital and microcontroller. The Ca1 system was limited to 12-bit digitization, which limited the resolution of our frequency corrections to the mid 10^{-16} given the signal size from our calcium atoms. Thus, one of the important changes made for Ca2 involved switching to a 16-bit ADC so as to reduce the digitization noise well below the calcium system's performance level.

The digitized values for the accumulated atomic fluorescence are then used within a programming routine to calculate and implement the desired frequency correction. The micro-controller also dictates the locking scheme, which relies on sampling the atomic response at four frequency values depicted in Fig. 4.19.

Figure 4.19: The locking scheme for our calcium system relies on measurements of the atomic response at four frequency locations total for the two fringe components. In this case, they are displayed for Ca2's dual ovens, but it is an equivalent process for laser beam reversal. Figure taken from [195].



The frequency corrections to freq2 can then be easily calculated from the digitized values, a_i

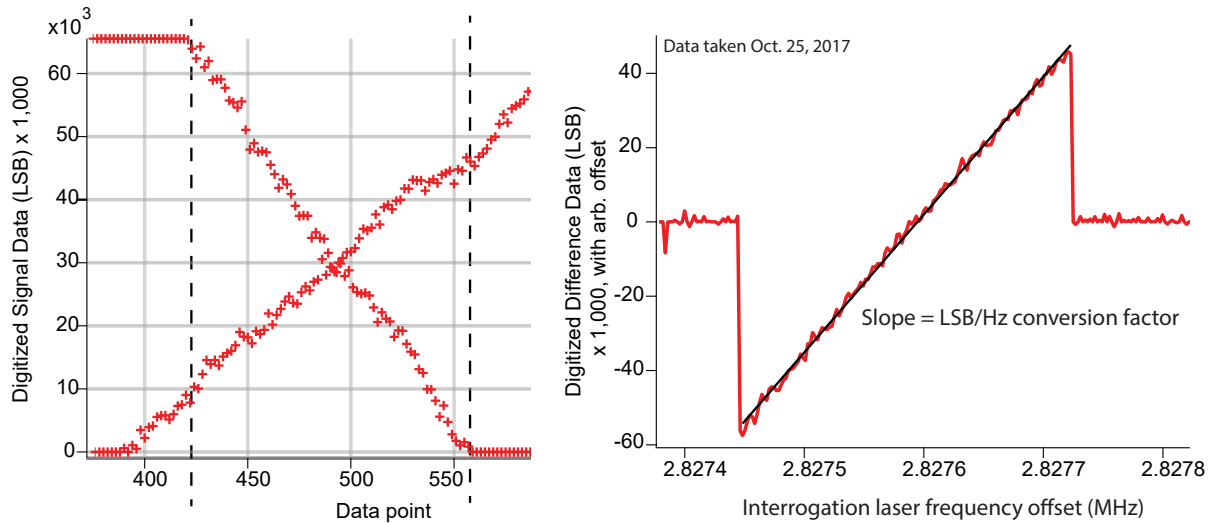
where $i = 1, 2, 3, 4$ in Fig. 4.19 corresponding to the cyclic measurements. In general, the correction to generate the calcium-stabilized optical frequency were given by $c = g[(a_2 - a_1) + (a_4 - a_3)]$ with some gain factor g and integrated. A second-integrator was also added, this one given by a scaled version of the integrated correction just mentioned, and this is also integrated over time so as to account for long-term drifts of the calcium corrections. Meanwhile, the difference in the two fringe component values, given by $c' = g'[(a_2 - a_1) - (a_4 - a_3)]$ is also tracked to compensate for the offset in the two fringe components used for interrogation, which factor into the modulation between frequency sampling points a_i that are fed to freq1 by the microcontroller.

The microcontroller also communicates the sampled data to a host computer running a Lab-view program that serves as a primitive interface primarily used for monitoring or post-processing data. The interface provides a visual feed of the incoming sampled values at frequency locations 1-4 as given in Fig. 4.19, and then separately computes the anticipated frequency corrections that the microcontroller sends to the AOMs. This interface provided essential information used for trouble shooting and understanding the frequency locking procedure, including some key values for predicting frequency instability performance, including the rather important conversion factor of least-significant-bit to Hz, i.e. the LSB/Hz value. This parameter is determined by the slope of the processed fringe signal, and allows direct conversion of fluctuations in the digitized bit values we measure into their corresponding frequency change. Thus, the LSB/Hz value is important for both calculating the noise properties of the atomic signal and other experimental noise parameters of interest.

The LSB/Hz conversion factor is easily found by recording the atomic signal response as we linearly sweep the Bordé laser frequency over the locking feature in the presence of the cyclic frequency modulation on either side of the fringe. The slope of the fringe on its steepest portion, where its frequency is typically sampled during the measurement process, would, in a perfect world, be a direct measurement of the frequency change in our laser. However, the noise processes inherent to the modulating sampling procedure inhibit the use of a continuous feedback protocol, and thus a continual sampling method with a finite integration of the atomic fluorescence is used. An example

of an LSB/Hz conversion factor calculation is given in Fig. 4.20, with more information in its caption on how such the LSB/Hz values were found for a single fringe component. The LSB/Hz value is fringe-dependent and can change based on the signal amplitude, fringe linewidth, fringe asymmetry, and fringe contrast. For frequency corrections then relying on complementary fringe signals, such as with our $\vec{k}$ -reversal and dual oven locking, obtaining comparable fringe patterns is important for simplifying the locking servo process. Otherwise, differing parameters (modulation depth, gain, etc) for the lock may be needed for each fringe.

Figure 4.20: These plots recorded the collected signal from a single oven source as the interrogation laser frequency was linearly swept by freq2. The narrow voltage range of the analog to digital conversion system (16-bit here for Ca2) is railed until the collected fluorescence comes within the window of 0 to 65535 LSB. The modulation process continues during this sweep, controlled by freq1, causing the data point of the left graph to alternate between the two distinct traces as each side of the fringe peak or trough is sampled. Where the two values are equal is the approximate locking point we strive to maintain. The right graph shows the difference values between adjacent measurements. This graph is scaled into frequency using the known step program of freq2. The slope of this plot, in black, corresponds to the data points between the black dotted lines on the left plot. The value of the slope indicates how measured changes in the digitized signal, in LSB, correlate to an expected change in the locking frequency in Hz.



The measured LSB/Hz value is extremely useful for determining the noise properties of our system, but can be misleading without also including a metric for the average noise of the system. For a digitized signal that has an RMS fluctuation of 500 LSB and a conversion factor of

100 LSB/Hz, we can conclude that the system will then show an RMS frequency fluctuation of 5 Hz, which, assuming valid after 1 s of averaging, corresponds to a fractional frequency instability of about 1×10^{-14} . The ability to easily convert the measured digital signal fluctuations into anticipated frequency fluctuations is extremely powerful. In addition to using this metric for sensitivity measurements covered in the following chapter, it is also quite useful for defining the noise contributed by roughly orthogonal noise sources - those of amplitude and those of phase.

The locked calcium system only samples atomic response on the Bordé fringes, where both changes in the DC signal amplitude and variations of the fringe phase and linewidth can contribute noise to the frequency lock. However, the frequency dependence from amplitude noise alone can be measured by instead sampling the atomic signal many 10's of kHz from the fringes but still upon the Doppler pedestal at a DC signal level comparable to when locking on the fringes. The atomic signal here can be sampled and processed as if the calcium system were going to use these digitized values for frequency corrections. The measured LSB fluctuations here then mostly correspond to amplitude changes in the collected fluorescence, as well as the electrical contamination incurred during processing. Thus, this **off-fringe** noise measurement, as it will often be called in this dissertation, provides a minimum fractional frequency instability value for the calcium system. If we were to be locking on the fringes ('on-fringe') as usual, it is clear that we should expect at least this amplitude noise contribution. Furthermore, any additional noise beyond that measured off-fringe can be attributed to fluctuations in the fringe properties or the interrogation laser frequency.

More details on the final Ca2 locking procedure and use of LSB/Hz conversion factors to assess noise contributions is given in Ch. 6.

4.9.2 Other ancillary servos in brief

In addition to the obvious calcium-stabilized frequency correction servo just described, numerous other servos were utilized to provide necessary stabilization of laser intensity, frequency, and phase. Both the 431 and 657 nm lasers utilized PDH-based frequency stabilization to Fabry-Perot cavities. Both lasers also underwent intensity stabilization before interacting with the atoms, by

comparison of a sample laser pickoff to an external low-noise voltage reference. Only the 657 nm laser required phase stabilization, which was accomplished by using back-reflected light from the flat-tipped fiber optic cable bringing the laser to the atomic vacuum apparatus. The phase of the back-reflected light is then compared to that of the laser at an earlier point in its path, prior to the fiber, and an AOM prior to the fiber is then used to actuate the appropriate response such that phase noise along the fiber is canceled.

4.9.3 Frequency evaluation

Measurement of the calcium-stabilized frequency instability for Ca1 could largely be measured by comparison against the 657 nm cavity-stabilized light. This worked when the calcium corrections, though working, could only provide stabilization at levels $\geq 2 \times 10^{-15}$ or so, i.e. at levels less stable than those of our cavity. However, the later Ca2 calcium system performance significantly surpassed that of the 657 nm Fabry-Perot cavity, thereby requiring an alternative frequency measurement method. In this case, we were conveniently able to work with the talented members of the frequency comb laboratory down the hall, notably Tara Fortier and Holly Leopardi, to whom we sent phase-stabilized calcium-corrected laser light. They could then compare the calcium frequency stability to that of the neighboring Ytterbium optical lattice clock's advanced Fabry-Perot cavity that routinely achieves fractional frequency instabilities of 2×10^{-16} at 1 s.

When the Ca2 system began to reach instability levels comparable to the Yb cavity, we were then able to measure via the frequency comb against the Yb optical lattice clock locked frequency when operable. Of course, these measurements became increasingly complex and required far more people to occur, making assessments of the calcium system progressively more difficult over time. Additionally, various measurement campaigns between our Yb lab and others at NIST and JILA precluded measurements of the calcium system for many months, as well as times when the comb or Yb systems were offline. Despite these interruptions, a great number of calcium measurements were able to be made thanks to time donated by Will McGrew, Xiaogang Zhang, Andrew Ludlow, and others who operated the Yb lattice clock for us.

Chapter 5

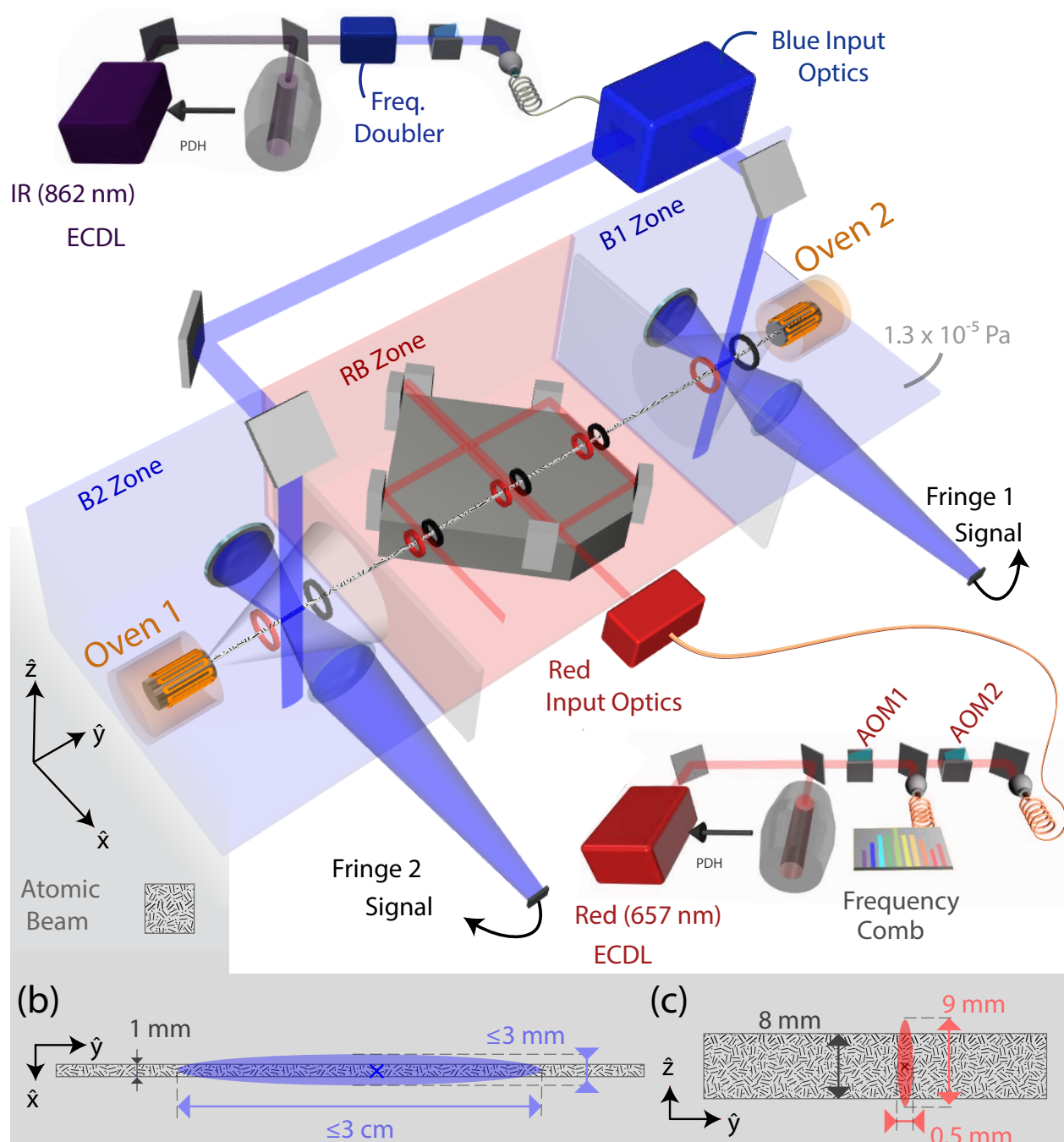
Final Ca2 design and operation

After experimentation with the initial Ca1 apparatus, it became clear that some rather drastic changes would be needed for a significant performance improvement. First, the Bordé laser's optical path needed to be better isolated from the environment. Second, we would utilize two atomic beams, overlapping and counter-propagating, to provide frequency corrections in lieu of the $\vec{k}$ -reversal method. Third, the Ca2 system would be designed to reduce all background sources of unwanted fluorescence. Fourth, the measurement electronics would be upgraded to a 16-bit micro-controller and a dual input gated integrator to account for the two photodetectors. The result of these changes became the apparatus I call Ca2 (pronounced $/s\bar{e}/\bar{a}/tu/$).

A basic, stripped back visual of the idea behind the Ca2 apparatus and relevant laser systems is shown in Fig. 5.1, which will now be discussed in detail. I begin with the upper left corner where the 431 nm laser source is generated. Infrared (IR) light from an 862 nm diode is frequency stabilized to a Fabry-Perot cavity held in vacuum via a Pound-Drever-Hall (PDH) locking scheme [75]. This frequency pre-stabilized IR light is then sent through a frequency doubler specialized for these wavelengths, which generally consists of a crystal with nonlinear optical properties such that the 862 nm laser beam can efficiently be converted to a 431 nm source¹. Prior to the purchase of the commercial Toptica doubler, I had constructed a home-built doubling cavity with some

¹ For the interested experimentalist, the commercially purchased IR diode and blue doubling system from Toptica is also capable of tuning to the 423 nm transition from the calcium ground state, often used as a cooling transition. It would be informative to see a direct comparison of 431 versus 423 nm state-readout based instabilities to see how my earlier assumptions about the two methods stand up to hard data.

Figure 5.1: Overview of the Ca2 system, adapted from [195]. See text for full description.



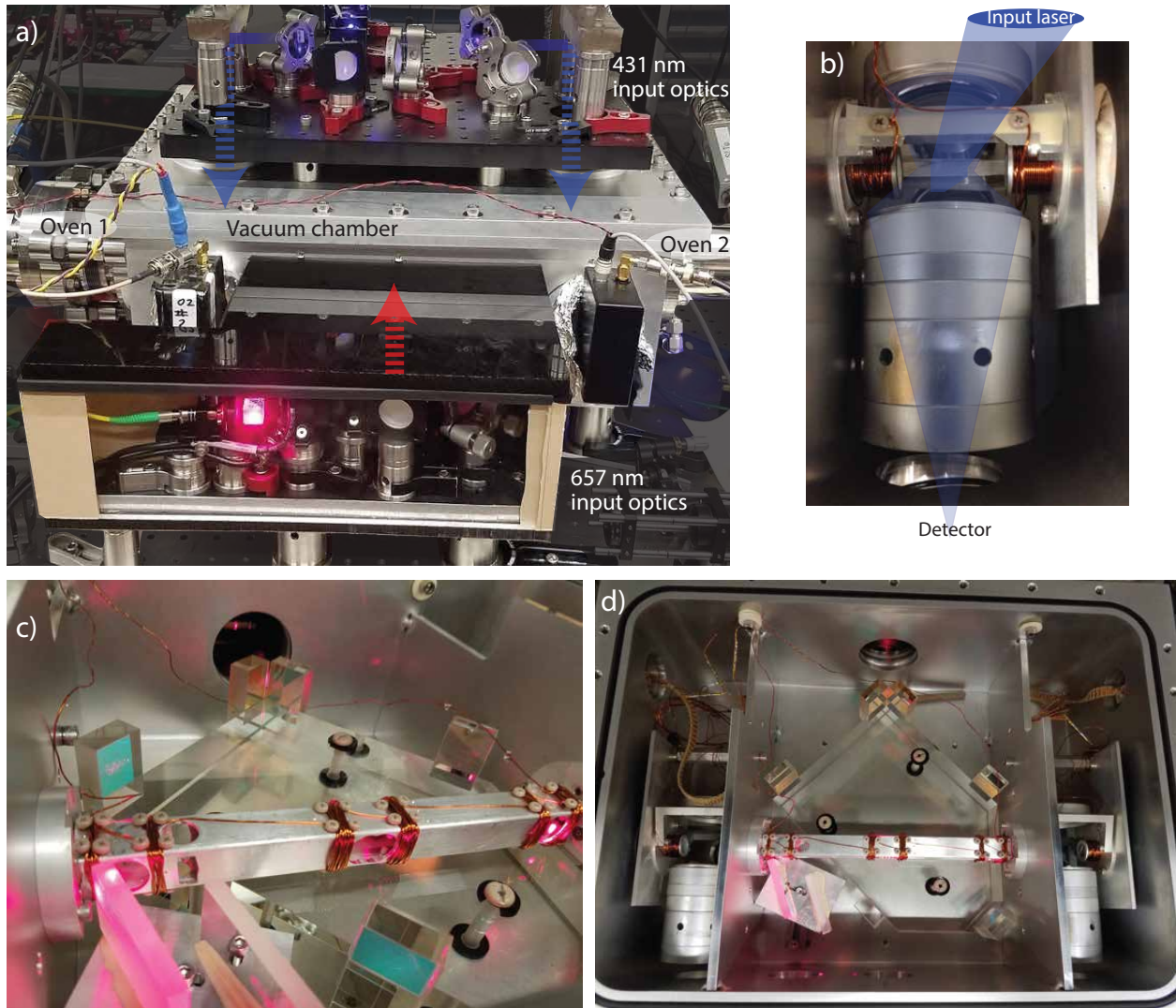
success.² Numerous locking schemes were tested, including transmission- then reflection-based Hänsch-Couillaud methods, which did not require in-path optical modulation of the laser light, thus allowing high IR power to reach the doubler. However, thermal issues with the crystal with >1 W of IR power incident on the cavity coupling mirror caused these locks to be less reliable than was practical. A standard PDH lock was later incorporated that was more consistent, but regular adjustments of the doubling cavity were still needed. The commercial system purchased later was far more reliable and rarely required adjustment.

The doubling cavity output is then single-passed through an acousto-optic modulator (AOM), and the zeroth order is coupled into a fiber. This AOM is aligned to divert 431 nm laser into its first-order diffraction dependent on the RF driving power, acting as an intensity stabilization actuator. The 431 nm laser is then fiber coupled and sent to the input optics breadboard atop the calcium chamber (Fig. ??.) As the blue interrogation beam at the atoms was elongated, I found sending 100's of mW of 431 nm light to the Ca2 chamber useful, but also dangerous for our fibers. Using specialty fiber with end caps reduced the damage they received from small misalignments and back-reflection of the blue laser.

From here, the blue laser of Fig. 5.1 was sent to its input optics assembly, which collimates the beam into an approximately 1.5 cm diameter Gaussian profile using adjustable lenses. The blue laser then passes through a pickoff, which directs a small percent of the total 431 nm power into two reflections on the front and back surface of the pickoff. To avoid possible etaloning, one of the reflections is blocked while the other is directed onto a photodiode to act as the in-loop blue intensity servo monitor. Light after the pickoff is passed through an elliptical beam shaper and then a non-polarizing 50:50 beam splitting cube, and each of the arms is directed toward its respective vacuum chamber entry window. Two mirrors in each arm allow for precision placement of the blue laser relative to the atomic beam, with one being mounted at the appropriate 45 degree angle to direct the beam vertically downward into the chamber. Between these two mirrors is placed a

² For home-built doubler systems like these, both for crystal choice and locking schemes, I highly recommend consultation of the text *Compact Blue-Green Lasers* [219], which was originally lent to me by colleague Shaun Burd. Thanks Shaun!

Figure 5.2: Photos of the Ca2 apparatus. a) The final assembled Ca2 system, with the blue optics board seen in place on top of the vacuum chamber, and the red input optics within a sealed plastic compartment. b) The blue detection region of oven 2, showing the blue zone solenoids wrapped around tubes that pass the calcium atomic beam. The input laser and fluorescence collection are artistically rendered. c) The RB-zone of the vacuum chamber, showing the ULE optics, the calcium beam tube, and the magnetic field coils attached to the beam tube. d) A top-down view of the populated calcium vacuum chamber. Ovens are to the left and right of the figure (not pictured).



half-wave plate for polarization adjustments.

The red (657 nm) laser system is depicted in the bottom right of the figure. A 657 nm diode laser source is frequency stabilized to a temperature and vibration controlled Fabry-Perot cavity housed within a vacuum environment. The 657 nm light is actually sent to the cavity via fiber (unlike its depiction here), and most optical elements for the standard PDH lock along with the cavity are mounted on a bread board within an acoustic shield. The remaining light not utilized for the PDH is passed through an AOM, called AOM1 here, which passes the first order diffraction component. AOM1 is also responsible for actuating the calcium frequency corrections. After AOM1, some of the light is passed to the frequency comb measurement room down the hall via phase-compensated optical fiber. The remaining light is passed through a second AOM, AOM2, where the first order diffracted portion also accounts for the frequency modulation of our locking scheme. This light is then passed to an optical fiber and delivered to the red input optics. This fiber is also phase-compensated via AOM2 utilizing the final flat-face fiber tip reflection.

The red input optics layout for Ca2 were originally made with long focal length lenses mounted to the optics bench to reach the height of the vacuum chamber's input windows. However, we found that extremely thick and cumbersome opto-mechanical posts were required to properly maintain the laser optical path when it was required to be many tens of centimeters from the optical bench. A second iteration fastened the input optics to the front of the chamber using pre-tapped holes planning in the chamber face in addition to posts directly under it, and this is depicted in Fig. 5.2a. A third iteration of the input optics further shortened the red optical path-length in air and is depicted in the next chapter on the Ca2 noise analysis.

Both the red and blue laser system output beams are directed into the vacuum chamber through windows anti-reflection coated windows for both the 431 and 657 nm wavelengths. To avoid etalons, all windows and viewports are made from wedged window substrates and are furthermore mounted at an angle relative to the chamber face. The windows and chamber lid are sealed via viton o-rings and 1/4-20 machine screws. The vacuum chamber is nominally made from 3/16" aluminum with welded baffles. In addition to the blue entry and exit windows, the red entry and

exit windows, and the detection viewports, a window was also installed directly over the ULE retroreflector such that the first and second reflections within the vacuum chamber could be easily monitored. Additional conflat ports were also provided to allow for electrical feedthroughs, vacuum gauges, the ovens, and vacuum pumps. The ion pump is directly mounted behind the ULE piece, visible in Fig. 5.2 parts c and d. The baffle walls possess large cut-outs near the ion pump area, and were planned to allow ample vacuum conductance between the vacuum chamber partitions.

The ovens generating our calcium beams were customized from basic molecular beam epitaxy sources with separate heating elements for the calcium crucible body and exit nozzle. The nozzle is run at a higher temperature than the crucible body to reduce clogging. The ovens were designed with radiative shields and water cooling so as to allow the vacuum chamber body to remain at a much lower temperature than the ovens.

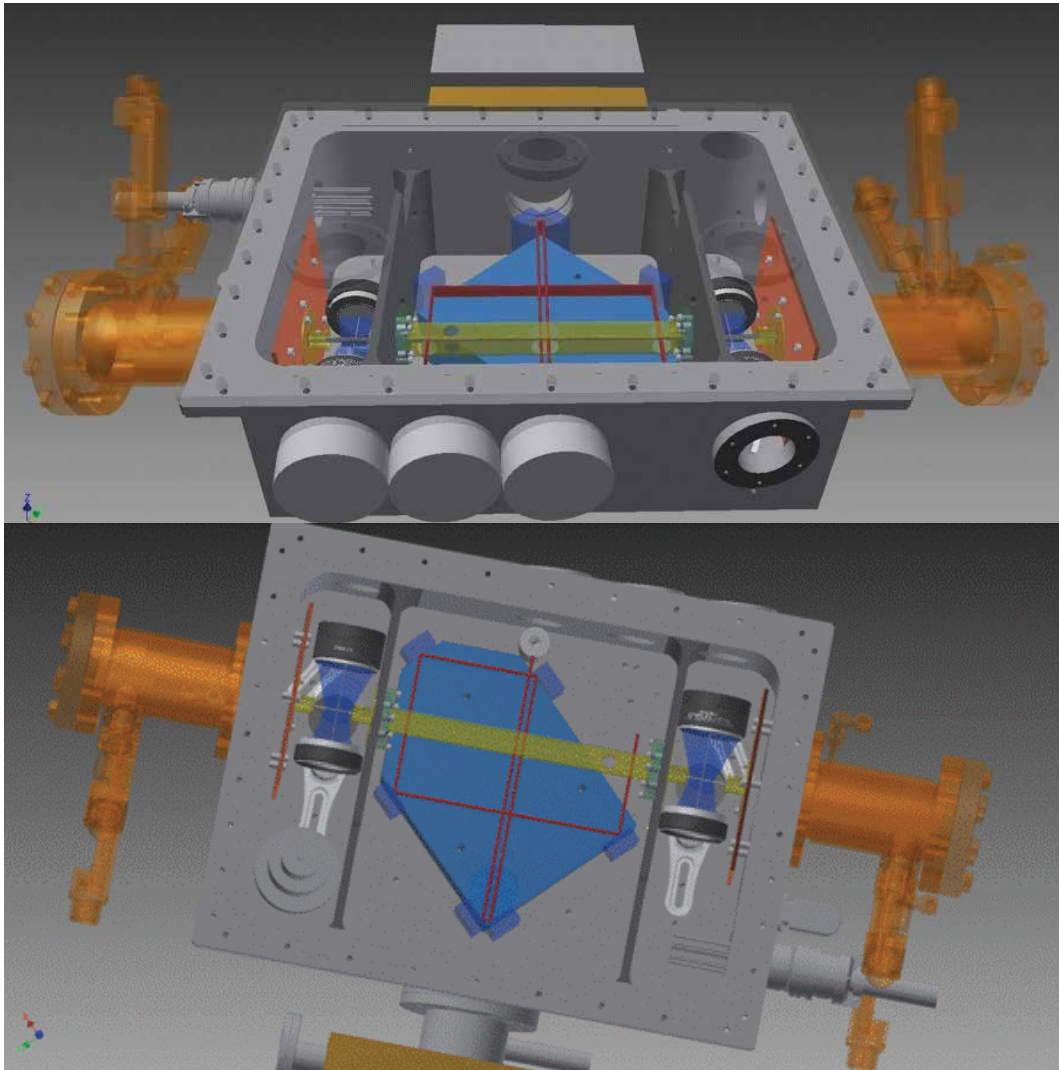
Some individual design aspects that contributed to the success of the Ca2 apparatus will now be discussed in more detail.

5.1 Vacuum chamber

I modeled the entire vacuum chamber and its contents in 3-dimensional software from the screws to the opto-mechanics and everything in between, as seen in Fig. 5.3. Numerous design iterations were created before settling on the final arrangement that was affordable to machine, small enough to fit in a commercially purchased acoustic isolation enclosure, and reasonable to work with given the size of tools and hands.

As to attributes of the vacuum chamber, the Ca2 vacuum chamber was designed with vacuum conductance, apparatus size, and functionality in mind. To simplify the apparatus, many aspects of the design were created to be multifunctional. For instance, the two interior baffle walls served to block photon cross-contamination between the red and blue zones, provided mounting surfaces for the fine apertures, beam tube (yellow in Fig. 5.3), and blue zone magnetic field coils. The baffle walls show significant cut-outs near the back wall of the chamber, allowing ample vacuum conductance to the ion pump while discouraging particles from contaminating the sensitive optical

Figure 5.3: The vacuum chamber walls are given in gray, the Bordé retro-reflector in blue, calcium ovens in orange, and the laser paths in their respective colors. The rectangular yellow tube above the ULE reflector allows the atomic beam to pass through while protecting the optics from stray atoms. It also supported magnetic field coils, which were wrapped around it. Numerous baffles can be seen that try to restrict unwanted calcium deposition while still allowing ample vacuum conductance. The ion pump is mounted on the back end of the chamber, and windows and feed-throughs on the apparatus lid are transparent in the top image.



surfaces in the Ramsey-Bordé central zone. Additional tapped holes in these large baffle walls also served to fix electrical wires in the vacuum, and the walls itself acted as a heat sink connecting to the main chamber body so as to lessen the possibility of overheating from the solenoid coils or from calcium deposition. A second set of baffles was also included, highlighted in orange in Fig. 5.3. These baffles blocked the vast majority of calcium spray off-axis from the final calcium beam while also providing an initial coarse aperture with a 1 cm diameter through which the calcium atoms could pass. These baffles were held approximately 1 inch from the side walls, allowing ample vacuum conductance to the oven interiors and crucible. Additionally, these plates provided mounting surfaces for solenoid coils needed for imaging in the blue zones while also acting as a heat sink for both the solenoid coils, hot calcium deposition, and residual thermal radiation not blocked by the crucible caps.

The dimensions of the vacuum chamber were designed to be as minimal as possible, which resulted in numerous close clearances. To avoid potential pitfalls, all parts to be placed in the vacuum chamber were also modeled, including the various screws, opto-mechanics, and mounting hardware. Access with tools like screwdrivers were also considered. Additional tapped holes were also provided along the interior chamber walls for routing electrical wires within the vacuum chamber and for ease of potential future modifications. Eventually numerous resistance temperature detectors (RTD's) were mounted within the vacuum chamber to monitor temperature changes and avoid reaching temperatures that might degrade high-vacuum materials.

To reduce the potential for deleterious etalons, all vacuum chamber windows were mounted at slight angles to the chamber face. Also in this vein, all windows were wedged at small angles. To reduce spurious laser scattering, all laser beams were passed back out of the chamber before termination in a beam dump and all optical surfaces were coated for either anti-reflection or high-reflection, dependent on their use. To aid in alignment of the interrogation laser, a window was also provided directly above the retro-reflector segment nearest the second and third laser interaction. This window also provided a convenient view into the status of the chamber interior such as for monitoring calcium deposition on the ULE reflector, ensuring all parts were still in place after

pumping, and making the experiment more accessible to tour groups.

To give some flexibility to the vacuum system, additional conflat connections were built into the chambers side walls and lid in convenient locations. Some of these were later used to pass RTD sensor signals to and from the vacuum environment or for the addition of pressure gauges. To allow easy access to the chamber interior when at atmosphere, the entire lid of the chamber could be lifted off. The lid sealed to the main body with a large Viton o-ring 36 1/4-20 machine screws, which fastened to tapped holes in a lip around the top of the main vacuum chamber body. Whereas all other vacuum connections were made with copper conflat flanges, the use of viton for the lid could limit the obtainable vacuum pressures even with higher vacuum pumping rates.

5.1.1 Vacuum procedures

When the Ca2 chamber was opened, such as to replenish calcium or adjust internal vacuum chamber components, the apparatus interior would be thoroughly cleaned. Within a few hours of breaking vacuum, calcium deposition would begin to oxidize, turning an opaque white color and flaking off easily. A clean room vacuum with miniature nozzle attachments was used to remove any visible debris from the chamber and ovens. The oven crucibles would be removed after detaching the ovens from the main chamber and soaked in a water bath until all visible calcium deposits were gone. Sometimes this also required the use of vinegar or light pressure from a soft cloth. All parts to go into vacuum were carefully cleaned, first in a sonicated bath of Simple Green household cleaner, and then a warmed methanol sonication bath. Parts were then left to dry sufficiently on UHV compatible foil and stored in air-tight containers with desiccant until ready to be installed shortly before sealing the vacuum chamber. On each opening, the reflection coefficients of the windows and in-vacuum optics would be tested, and any surface showing significant deviation from previous values was then carefully cleaned with methanol. Some surfaces that experienced heavy calcium deposition were most easily cleaned in vacuum by using a razor blade to pry up the corner of the deposited calcium plaque prior to its oxidation. Often such thick plaques could then be easily pulled off the chamber surfaces as a single piece without risking further damage to the chamber

surfaces.

To pump down, the vacuum chamber would first be sealed and a combination roughing/turbo pump run. If the turbo pump could come up to speed and no obvious leaks were noticed, then RTDs were attached at three locations on the chamber's exterior and one on the ion pump. The chamber and its attached ion pump (with all input optics assemblies removed) were then wrapped in a layer of aluminum foil followed by heating strips. A final layer of foil then encased the entire apparatus. The exterior chamber temperature was brought to approximately 100 °C by the variac-driven heating strips while the ion pump was brought closer to 150 °C. The system would be baked like this while under the roughing pump for three days or so before the vacuum levels would reach ion pump range. The chamber pressure spikes quickly when the calcium ovens are initially turned on, so often the ovens would be increased to near 500 °C or more at this point while still roughing so as to remove much of their outgassing before relying on the ion pump alone. Once the apparatus seemed ready and stable, the roughing pump was detached and the ion pump was left to do the remaining work. Over the course the next two days or so the oven temperatures would be slowly stepped beyond normal operating temperature while the ion pump performance was monitored to ensure that the vacuum chamber pressure never dipped above the ion pump's limitations. Typical pump down and bake-out required approximately nine days to return the Ca2 apparatus to nominal vacuum operating conditions between $1-2 \times 10^{-7}$ Torr.

When preparing to break vacuum, the calcium ovens were reduced in temperature over an afternoon to reduce the possibilities of additional clogging that might interfere with our system assessments upon opening. The nozzle was always kept at a higher temperature than the crucible body, usually with a differential of approximately 20 °C. Larger differentials could not be supported by the thermal conductivity of the crucible between the nozzle and main body elements without the higher temperature nozzle ultimately doing all of the work. All electrical feedthroughs to the system were then turned off and the chamber grounded in case of shorts or electrostatic buildup. The lid and/or ovens would then be removed for the desired maintenance.

5.2 657 nm laser system

The final red 657 nm Ramsey-Bordé interrogation laser used for the Ca2 apparatus was very similar to when Ca1 was operating but with a few modifications. The original diode and ECDL were replaced with a commercially available system and controller. The slave diode was also replaced with a commercially available laser amplifier that had been tested in tandem with the slave diode on Ca1. All were purchased from Toptica Photonics. The laser was run with both phase and intensity stabilization shortly before entering the Ca2 chamber.

5.2.1 Mode competition

One issue that became clear when operating the new system was that of frequency mode contamination, wherein the laser beam would run slightly multi-mode. While the total laser power was actually stabilized, fluctuations in power distribution between the primary and competing modes could be observed on the calcium fluorescence signal. Verification of mode-competition in the interrogation laser was eventual accomplished through the use of a 1.5 GHz FSR scanning Fabry-Perot cavity purchased from Thorlabs. This device possessed just enough resolution to see conjugate fluctuations of a small frequency mode near the primary one. Re-calibration of the laser locking parameters was able to reduce the mode competition to unnoticeable levels when a mode-hop free tuning range of the 657 nm laser source surpassed approximately 8 GHz. Such calibrations were done by externally driving the PZT with a triangular wave function while monitoring the transmission from the undriven (stationary) 1.5 GHz cavity. Mode hops, when present, could be clearly seen on the scope trace from the transmission when triggering on the PZT's driving waveform. The impact of the mode competition on our atoms was assessed by adjusting the diode current slightly, forcing the driver's PZT response to compensate to remain locked. When the tuning range was ≥ 8 GHz, no change in the atomic fluorescence could be seen as the PZT was adjusted. When the tuning range was < 8 GHz, the same process would typically cause clearly correlated changes in the atomic response level that were large enough to be a cause of either

long-term frequency drift or short-term frequency fluctuations.

5.2.2 Optical path in vacuum

The decision was made to enclose the great majority of the 657 nm's optical path within the vacuum chamber so as to eliminate the air-induced fluctuations in phase, pointing, and wavefront discussed in the previous chapter. Using the precisely measured angular tolerances and data on ideal Ramsey dark time spacings discussed in the previous chapter, my adviser, Andrew Ludlow, then proposed a monolithic retro-reflecting structure that could direct the laser entirely within vacuum. Furthermore, to reduce potential variations in the optical path length from components moving or vibrating, this structure was to be immutable³. Soon after its initial installation, the in-vacuum ULE reflector was found to yield Bordé fringes from our atomic beams. Furthermore, with the reduced degrees of freedom in aligning the interrogation laser, fringes could easily be found within a few minutes (assuming the lasers were behaving and near the correct frequencies). Optimized contrast values between 13 and 19% were routinely obtained, outperforming the more typical 8 to 12% values seen with Ca1.

The ease of optical alignment with the monolithic reflector was also aided by a few design elements afforded by my extensive 3D modeling of the Ca2 apparatus. The exact desired location of the 657 nm laser on the input window was well-known, as was its final location on the exit port window. With a broad frequency sweep over 10 MHz or so, the input 657 nm beam could be quickly adjusted to maximize the Doppler within seconds. Then fine alignment to resolve fringes could be easily done on a narrowed sweep covering only 10's of kHz near the expected fringe frequency, roughly centered on the Doppler peak. With far less time now dedicated to alignment

³ The time between when the monolithic structure design was finalized and when it was actually implemented was a stressful time. Concerns over the measurements of our previous retro-reflector, which was found to direct beams on paths far less parallel than the measured angular sensitivities, kept doubt alive. Without a way to measure the laser beam parallelism within the Ca1 vacuum, there was no way to know if the Bordé fringes actually had the best contrast when the interrogation laser paths possessed some small but finite angular offset. The entire Ca2 system was developed around this central ULE monolithic reflector, and if it were to fail, then the rest of the Ca2 development done thus far might have been futile. Creating our own in-vacuum retro-reflectors from commercially available opto-mechanics might take years or be impossible given (1) the mechanical changes as vacuum pressures lowered and (2) the large wait times between vacuum breaks to make adjustments.

of the Ramsey-Bordé apparatus, more complicated and interesting measurements could be made. Additionally, the fringes were more consistent from run to run, making long-term comparisons of the system performance easier.

The final angular tolerances for the ULE structure were specified at 1 arcsecond ($5 \mu\text{rad}$). The main body and mirrors were hewn from a large block of ULE. The mirror pieces were then partially high-reflection coated for 657 nm (only on the relevant surfaces that would be reflecting) and then optically contacted to the base. The final ULE piece was placed in the central Ca2 vacuum chamber and held in place with three long polyether-ether-ketone (PEEK) plastic screws that I vented by hand. They screwed directly into the Ca2 apparatus base plate, though Viton o-rings were placed between the ULE and the aluminum walls. Initially the Ca2 apparatus was sensitive to rotations of the optics bench, but calculations of the expected Doppler shifts and Sagnac effect did not correspond well to what was measured in the laboratory. Upon re-opening the chamber, the ULE piece was additionally glued in place with UHV compatible epoxy. After this, the rotation sensitivity dropped greatly, though no rigorous tests of angular rotations were subsequently performed.

5.2.3 Maintaining optical surfaces

A reasonable concern when enclosing optics in a high vacuum (2×10^{-7} Torr here) near the thermal source is deposition on sensitive optical surfaces. However, no significant calcium deposition on the ULE retro-reflecting surfaces was seen over about 2 years of running Ca2 regularly. I assume this was facilitated by enclosing the vast majority of the atomic beam within a hollow tube while in the Ramsey-Bordé zone. Small apertures in the tube allowed for the laser beams to pass undisturbed. Calcium deposition was found inside this tube, but was minimal. The vacuum conductance openings between the chamber divisions were placed near the far back wall of the chamber, by the vacuum pump connection. Thus, particles or renegade calcium atoms from the outer blue detection zones were less likely to wind up near sensitive optics. Conveniently, the apertures in the atomic beam tube also aided in the laser alignment.

To monitor potentially unseen calcium deposition on the retro-reflector surfaces, the reflection efficiency of each surface was measured during most vacuum breaks. The only time a significant drop in reflectance was seen came after a solenoid housed in one of the blue zones malfunctioned. The coil was driven with too much current, causing more heating than anticipated, until insulation coatings failed, resulting in burn out of the coil and substantial vacuum contamination. An orange-brown chemical deposition was seen on the chamber walls around the coil and roughly followed a path from the burned-out element to the ion pump as seen on the underside of the vacuum chamber lid. These depositions were extremely difficult to remove chemically. The two mirrors nearest the ion pump connection were the most impacted. These elements were cleaned by first wrapping the contaminated mirrors (still attached to the ULE piece within the chamber) with a methanol soaked cleanroom cloth followed by gentle wiping. After cleaning, all retro-reflection elements returned to reflection losses $\leq 1.5\%$ when operated at their standard 45° incidence. These power losses caused slight variations in the total power at each atom-laser interaction and could have introduced wavefront deformities (phase distortions) between the interaction zones, all of which could impact the Bordé fringe lineshape.

Lastly, the 657 nm input window to the vacuum chamber wound up being important for obtaining our high fringe contrasts. This window was most often exposed to mechanical damage from probes and power meters as well as oils and residues from human hands and dust from the laboratory. Maintenance of this window and regular, careful cleaning were extremely important for obtaining high contrast fringes. All other optical surfaces between the fiber exit and the input window were housed within enclosures that protected them from such contamination.

5.3 Calcium beams

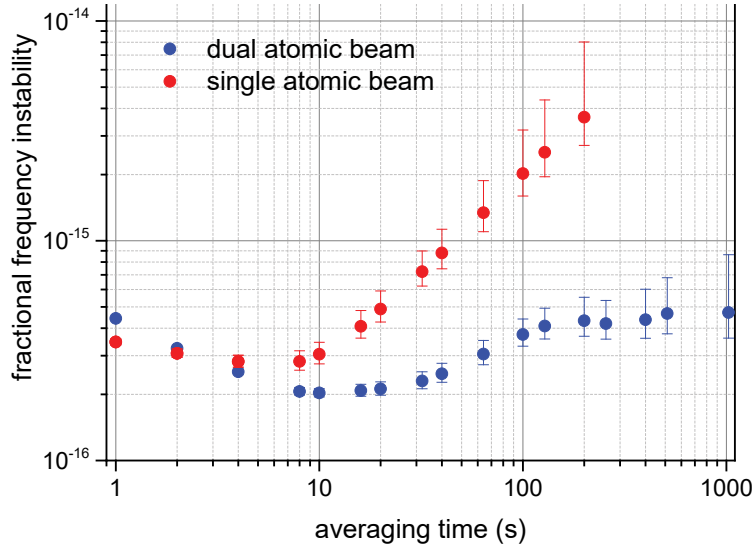
One of the most obvious differences between the Ca1 and Ca2 systems was the choice to utilize two ovens for Ca2 instead of relying on $\vec{k}$ -reversal to cancel residual first-order Doppler shifts. With the two ovens, achieving nearly identical fringes from both ovens was extremely easy, as the two atomic beams utilized the same interrogation laser and experienced the same laser-interactions,

albeit in reverse order. Because only one laser path was required, far fewer optics were needed to prepare the interrogation laser. With dual ovens, no power was lost to fiber coupling through the MEMs switch used for $\vec{k}$ -reversal, and no time delays were incurred from waiting for the laser intensity and phase servos to settle. Because the atomic beam dimensions are mostly determined by the fine apertures and oven nozzles, forcing the ovens to utilize the same fine collimation apertures ensured very similar atomic beams - far more similar than we could make two 657 nm laser beams with Ca1.

Based on measurements from Ca1, the ovens of Ca2 were designed for higher temperatures, able to surpass the melting point of calcium around 842 °C. Beyond 760 °C, our system should possess atomic flux values beyond 10^{13} atoms per second, theoretically corresponding to a quantum projection noise at 1 s of only 1×10^{-18} via Eqn. 1.1, even accounting for the broader fringe linewidth accompanying the higher oven temperatures.

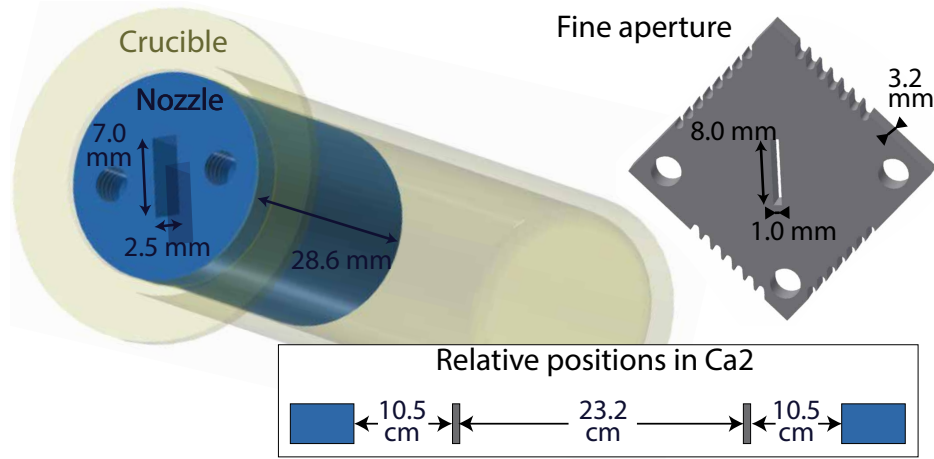
An early graph comparing the Allan deviations of Ca2 stabilization based on a single atomic beam versus dual atomic beams is given in Fig. 5.4. It clearly indicates that use of both ovens' Bordé fringe signals provides better long-term frequency stability than a single fringe does. Though, at timescales of order 1 s and below, it is still not clear whether or not the dual atomic beams actually provided significant improvements, though the ambiguity here could be attributed to non-ideal locking parameters for the dual ovens during this early stage of development. Unfortunately, uncertainty limitations of the frequency comb's averaging intervals prevented exploration of this phenomenon for timescales near the Ca2 cycle time.

Figure 5.4: Instability results for single versus dual atomic beam locking schemes. The dual beams clearly show suppression of longer-term fluctuations over many periods of averaging. This data was taken with the Ca2 system, but is roughly equivalent to k-reversal.



The atomic beam dimensions were most strictly determined by two fine apertures housed in the walls between the blue and red zones of the Ca2 apparatus. To accommodate misalignment between the ovens and the apertures, the oven nozzles possessed a wider aperture. The nozzle and apertures are shown with their respective dimensions and relative spacing in Fig. 5.5. For some time an adjustable baffle was also incorporated between one of the ovens and the main chamber, which allowed the angle of the oven to be adjusted relative to the chamber. Very little sensitivity to this angle was observed, both in terms of the atomic flux levels and the Bordé fringe frequency, implying that the fine apertures do define the majority of the atomic beam's transverse velocity distribution and that oven alignment was not an enormous concern.

Figure 5.5: Depicted are the Ca2 oven nozzles and fine apertures, which, together, limit the transverse velocity distribution of the calcium beam. The tapped holes on the nozzle face accommodated a machine screw to make removal of the nozzle from the crucible easier after use. The jagged edges on the fine aperture were intended for wrapping nichrome wire in case the aperture needed heating to remove clogging calcium deposition, which was luckily never needed.



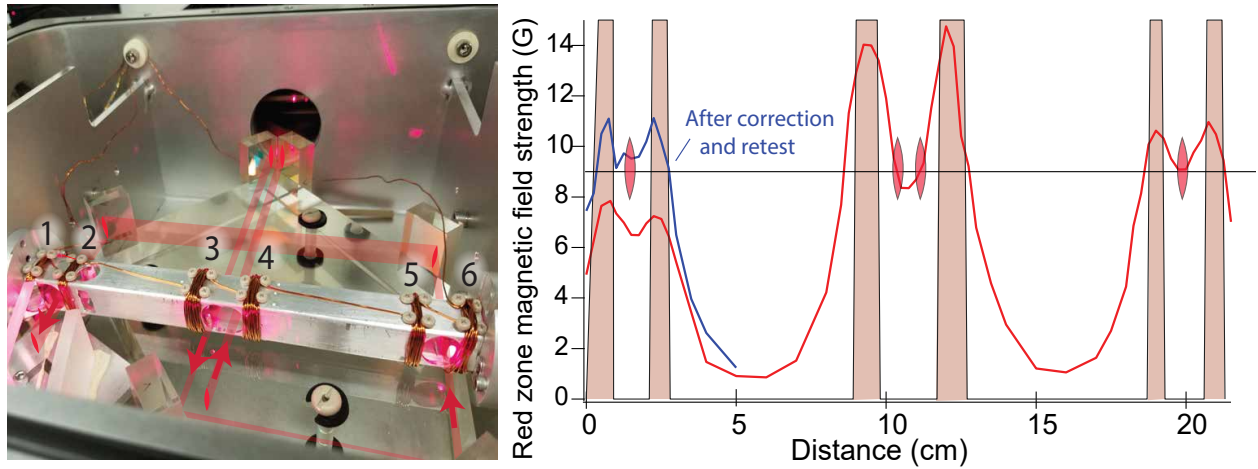
5.4 Magnetic fields

The magnetic fields for both the red and blue zones were generated by pairs of Helmholtz oriented solenoid coils, six coils total in the central red zone and four total for the blue zones, arranged to create parallel magnetic fields along the atomic beam axis. The coils were powered by a low-noise current source fed into the vacuum environment, with the Kapton insulated wire affixed to the interior chamber walls to reduce sympathetic vibrations and act as a heat sink. The magnetic field peak is fairly uniform over the laser-atom interaction in the red zones, but varies significantly over the much longer blue interactions.

As to the red zone, I remade the red-zone magnetic field coil assembly numerous times during the development of the Ca2 system. Because of the ideally zero distance between the two central Ramsey-Bordé laser interactions, one coil had to be used for both of the inner laser interactions (See Fig. 5.6a). Thus, coils 3 and 4 over this region had to possess additional turns so that the magnetic field at the inner laser-interaction zones required more turns to generate an equal field as the outer coils. An early measurement of the magnetic field strength along the atomic beam propagation locations is given in Fig. 5.6b for an arbitrary current choice near the anticipated

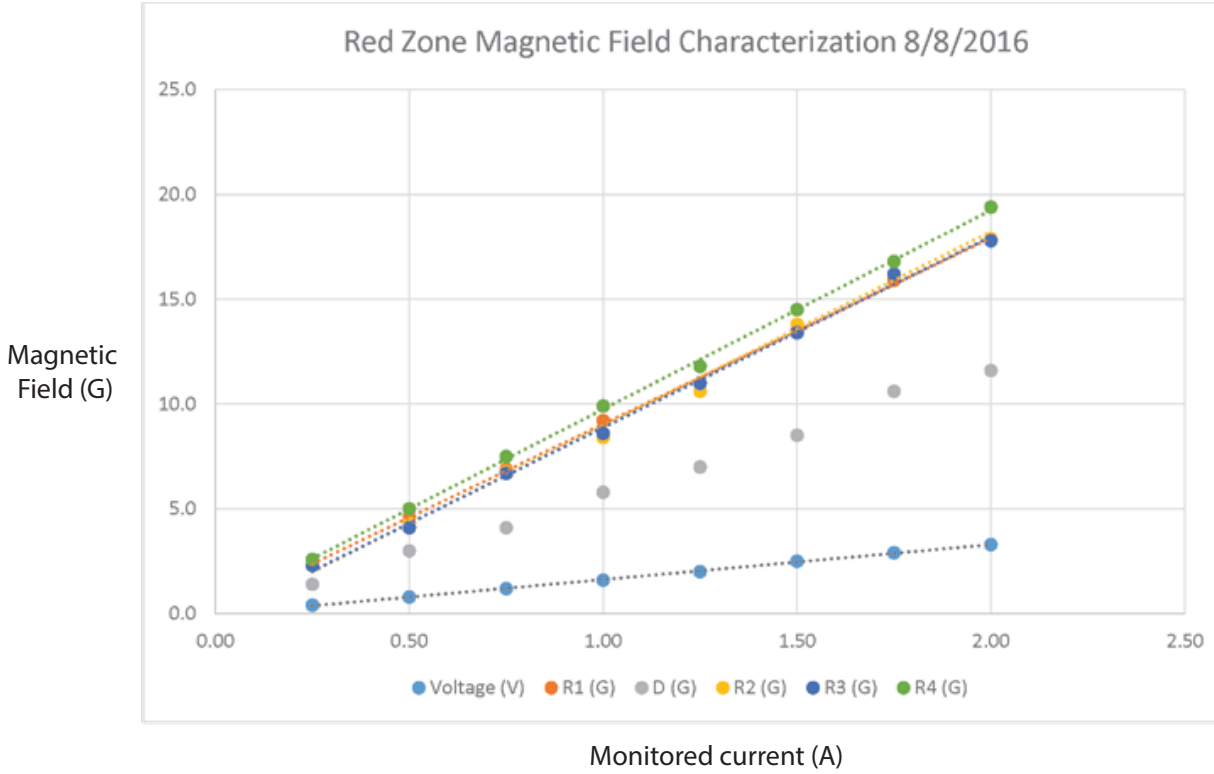
driving current. Screws made from UHV-compatible PEEK plastic held the coils in place. The tube on which all of the coils are wrapped served not only to provide the coil center, but also to protect the optical elements from calcium spray.

Figure 5.6: On the left is a photo of the three pairs of Helmholtz-oriented solenoids, mounted to the calcium beam tube and held in place with PEEK screws. The six individual coils are labeled by numbers 1 to 6, and the laser beam path has been superimposed for clarity. On the right, measurements of the magnetic field are given as a function of distance down the atom tube on which the solenoid coils are mounted. The locations of the coils are indicated by the filled in rectangles while the laser interactions are indicated by the red laser beam profile. Example of a coil correction is also given after it was initially found to have too low a field.



Once the field strength was consistent at the laser-atom interaction zones, their response was measured versus the driving current, as can be seen in Fig. 5.7. Typical field values utilized were near 8 Gauss, as determined by increasing the driving current until fluorescence of the Bordé fringe maximized.

Figure 5.7: The magnetic field measured at each of the four 657 nm laser-atom interactions, given by $R1$ thru $R4$, all given in Gauss. The field at D was intended as a single-pass Doppler interrogation zone for measurement of atomic flux. The field in zone $R4$ is clearly offset, and in a future iteration would be re-wrapped so as to provide a more consistent magnetic field at all of the interaction zones.



Each blue zone magnetic field consisted of two, much larger solenoid coils. To avoid blocking the blue signal photons, the coils had to be placed further away from the laser-atom interaction region, thus required many more turns to generate the desired field with reasonably low current levels. The coils were mounted on hollow spindles through which the atomic beam passed. The spindles possessed a large flat face for mounting onto their respective baffles to improve thermal conductance. The blue zone fields were initially wired in series and powered by a shared current source, but later they were run individually to allow slight differences in magnetic field power such that each blue zone was independently optimized.

Similar data was taken for the blue fields as the red, with the final fields also peaking around 8 Gauss at the center of the laser-atom intersection. This field value produced the best fringes

as found when assessing the fringe amplitude and contrast for a wide range of 431 nm linear polarization angles relative to the magnetic field for a variety of field amplitudes.

5.5 431 nm laser

The blue shelving detection of Ca2 built upon the initial ideas tested with Ca1. Namely, the blue laser profile was additionally stretched along the direction of the atomic beam to extend the time during which excited-state $(4s4p)^3P_1$ calcium atoms are rapidly cycled to and from the doubly excited $(4p^2)^3P_0$, pumping out photons for our detector all the while. This increases the number of photons we collect per atom, which, as mentioned in Chapter 2, is useful for decreasing certain types of noise processes. However, detecting signals like the Bordé fringes from our thermal beam, which are relatively small in amplitude and frequency space compared to the Doppler background on which they sit, can be easily dwarfed by excess laser noise. This was the case with Ca1, where the background photons scattered off the vacuum chamber windows comprised the majority of our signal, except when using laser profiles much smaller than those used in Ca2.

Ideally, every signal atom needs to contribute at least one electron to the frequency correction, even after accounting for all conversion losses to the electronic signal and solid angle collection efficiency. In this limit, quantum projection noise should form the ultimate limit of the frequency stabilization system, assuming other technical noise sources were minimal. The blue 431 nm system is what really let us explore this new regime of operation by enabling us to sample more than a photon per atom. At this point, the oven temperature can be increased, which increases the atomic flux. The blue system utilized here was overkill given the other noise sources in our signal processing electronics, meaning that the quantum projection noise fell below our measured frequency instability.

To ensure we collect as many signal photons as possible from our atomic beam, the blue electron shelving laser is extended along the atomic beam dimension to cycle as many photons as possible in front of the atoms. In the Ca2 system, the blue beam has been tested with lengths from about 1 cm to nearly 4 cm. A nearly linear change in atomic response was seen with the increased

length, so long as the beam power was also increased to maintain the same average intensity, close to the atom beam's saturation limit. The 431 nm transition is quite broad for calcium - more than 30 MHz, but the accumulated recoil kicks during the cycling can in theory impart enough of a velocity change in the atom that it becomes non-resonant with our 431 nm source. To test whether or not this was the case, the blue laser was retro-reflected back through the atomic beam. This did improve the signal size somewhat with lower powers, but no more than would have been improved with doubling the single-pass blue laser power. A variety of wave-plates were also tested below the Ca2 chamber to adjust the 431 nm polarization before passing back through the atomic beam. No polarizations were found that produced substantially improved results. When the blue beam was run well into saturation, retro-reflection ceased to improve the signal amplitude but did continue to increase the background and cause worry about etaloning. This was the case for all blue beam shapes tested, with the longest test being on a 4 cm long laser profile along the atomic beam direction.

Ultimately, the blue beam found to be the most advantageous for Ca2 was elongated to approximately 3 cm and came to a gentle focus at the atomic beam. Its profile was only slightly wider than the atomic beam. Longer beams caused more scattering than desired.

5.6 Signal collection optics

For Ca2, the windows through which all lasers passed were intentionally positioned relatively far from the atomic source we were imaging. To further reduce laser scatter, the 431 nm laser enters the chamber through a view port in the lid and exits through the bottom, on an axis orthogonal to the average atomic velocity and also to the imaging axis, as depicted in Fig. 5.1 and 5.3. Thus, using a standard lens system, a real image of the atomic beam could be formed beyond the view port and outside the vacuum chamber. This let us readily adjust and experiment with optics external to the vacuum chamber that could further wrangle signal photons onto the detector. In the end, we decided upon a mirror on the far side of the imaging window, whose focal point lay at the atomic source being imaged, and a lens, with a focal length half that of its distance to

the atomic beam. Because the atomic beam possesses a finite depth and significant length off the optical axis, the image formed was distorted. Furthermore, because the mirror also creates a real image, misalignment of the mirror causes two images to form, offset along the optical axis.

To ensure proper positioning of the blue collection optics within the vacuum chamber, aside from careful measurement based upon the CAD model of the chamber, I also created a test source for the fluorescence from a thin series array of small surface-mount LEDs with wavelength near that of the 431 nm source. These LEDs were mounted such that they were held at the height of the atomic beam. To ensure proper alignment of the LED array along the imaging axis, a small red LED source placed in the opposing oven crucible was directed through the fine apertures and into the blue detection zone being tested. The blue LED array was then positioned to fall along the assumed atomic beam axis given by the red laser path. The imaging optics could then be adjusted such that the mirror formed an image of the LED array upon itself. The lens was then positioned so that a nice external image of the array was formed on the chamber exterior. This method helped align the in-vacuum optics with more certainty than basic geometrics measurements could. Of course, changes in refraction angles from the differing indices of refraction between vacuum and atmosphere were not accounted for in this model, thus slight adjustments to the optical component positions were then made to account for this.

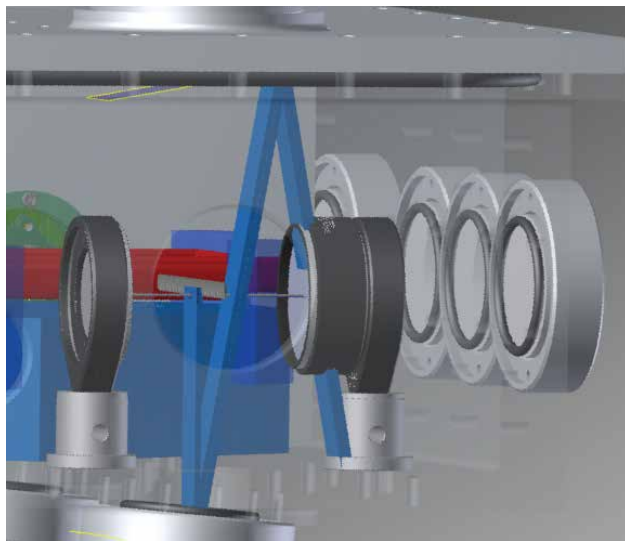
To maximize the solid angle collection of photons from the atomic beam source, optical elements were chosen to have the largest numerical aperture (NA) possible through selection of relatively large 2" optics with short focal lengths so as to be close to the atomic beam. Eventually, the single lens was replaced with a two-lens system, wherein the first lens was placed at a focal length from the atomic beam. A second lens then condensed the fairly parallel rays from the first lens to its focal point exterior to the vacuum, wherein a large area photodetector was placed. Due to imperfect alignments and the finite volume atomic beam source, additional optics exterior to the vacuum were found to increase the number of signal photons incident on the photodetector, such as a small focal length lens placed directly in front of the photodetector.

To reduce unwanted scatter from reaching the photodetector active area, not only were

relevant surfaces in the vacuum chamber coated with Acktar ultra-black absorptive film, but an aperture was also inserted external to the vacuum chamber. The calcium system could be easily run with a large 657 nm frequency sweep, causing the blue signal image to flash on and off whereas unwanted scatter remained unchanged. With laboratory lights off, the blue signal fluorescence could be easily assessed by eye and large sources of uncorrelated 431 nm photons that would otherwise reach our detector were physically blocked.

Lastly, to minimize specular reflection off of the vacuum chamber viewports through which the 431 nm laser entered and exited, the locations of additional reflections internal to the Ca2 vacuum chamber were modeled in the existing 3D design environment (Fig. 5.8), and the most probable scatter locations were further coated with absorptive Acktar ultra-black UHV-compatible adhesive foil.

Figure 5.8: In an effort to further reduce the blue background noise caused by spurious reflections, the next few vacuum-internal reflections of the blue laser beam were approximated to reduce their impact on fluorescence collection.



5.7 Photodetection

A primary concern for a photodetector used with an imaging system like in Ca2 is the active area of the photodetector, as the optical image formed is extended. Large area photodetectors

have become more popular and accessible in recent years, as well as detectors with high quantum efficiency for detection of blue optical wavelengths. We initially utilized PMT's, which, as discussed in earlier chapters, were troublesome given the large optical powers beyond PMT specifications that we collected as well as trouble with fluctuating DC offsets or responsivity of the detectors. Numerous commercial PMTs with built-in trans-impedance amplifiers (TIAs) were tested with both Ca1 and Ca2, and these problems were found in all of them. Some of these issues could have stemmed from supplying the TIA gain-control voltage with batteries, which, while very quiet, also lost charge over time.

As the total collected signal fluorescence grew with improvements to our imaging, the PMTs were pushed further from their specified operating conditions. Eventually, the measured white noise level of the detectors, as measured when a white light source (flashlight) stimulated an equivalent PMT response, rose to be at or near the entire calcium system's measured noise level. Normally one would expect that the increased fringe amplitude would also increase the LSB/Hz scaling factor and cause an overall reduction in noise. Whether due to degradation of the PMT itself, problems with the TIA controls, or electrical issues caused from new servo impedances or pickup, using PMTs became increasingly problematic and the noise from our detection system on seemed to limit the system performance at short time-scales.

In the time between when the PMTs were originally selected and when they became a source of concern for our experiment, Hamamatsu had released a new line of large area photodiodes with high blue responsivity. These photodiodes possessed promising specifications for our system when used in tandem with a home-built TIA⁴. One large difference between the photodiode and PMT approaches to photodetection came through bandwidth. The photodetection bandwidth was much lower than that of the PMT, which required additional considerations for our measurement timing. The photodiode approach seemed quite promising, but was ultimately limited by electronic pickup caused by locating the SRS-570 current pre-amps far from the diodes. The diodes were re-housed

⁴ This process was greatly aided by reading Hobbe's text entitled *Building Electro-Optics* [121] and the use of Analog Device's online Photodiode Circuit Design Wizard, which suggests appropriate parts for the TIA and predicts its operational properties like noise levels for particular bandwidths.

with a home-built TIA within the same metal enclosure, but the calcium experiment closed before the new housing could be properly tested.

Though a combination of improved measurement timing, described in the next chapter, and improved noise properties of the photodiodes over the PMTs, the 1 s Allan deviation instability was able to be brought down from $\approx 6 \times 10^{-16}$ to 2×10^{-16} .

Chapter 6

Final results: Ca2 noise and sensitivity measurements

This chapter will detail the various noise contributions of the optimized Ca2 calcium system and culminate in a final discussion of the thermal beam frequency reference performance. Some of the noise contributions include:

657 nm laser noise

- Linewidth
- Phase
- Drift
- Intensity
- In-air path

431 nm laser noise

- Frequency
- Phase
- Drift
- Intensity

Magnetic fields

- Earth's field
- Red-zone magnetic fields
- Blue-zone magnetic fields
- Electronics

Atom source

- Oven temperature
- Atomic beam collimation
- Aperture impact
- Sputtering
- Background gas collisions

Vacuum chamber

- Vacuum pressure

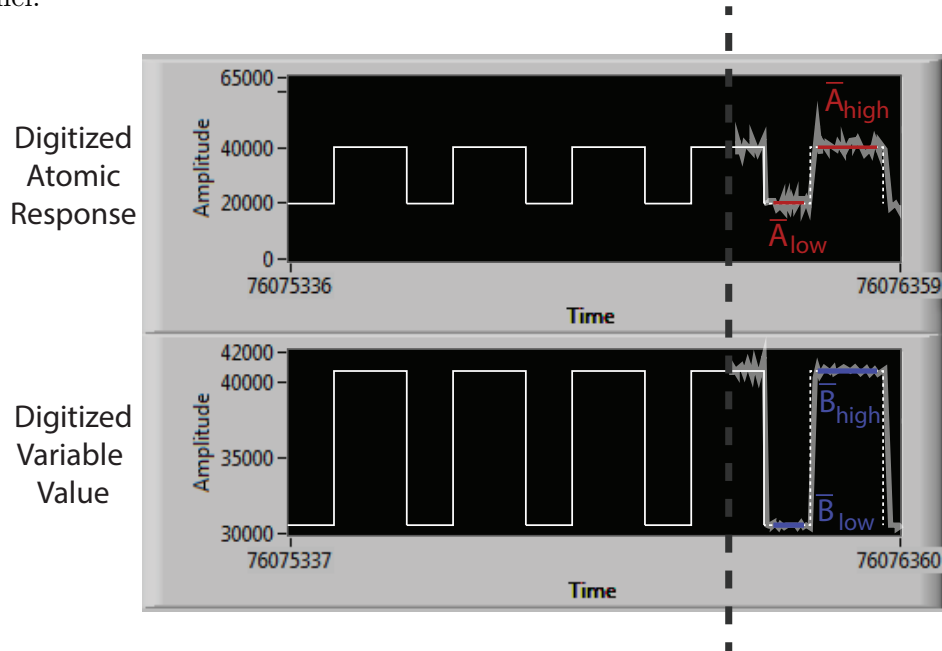
External effects

- Temperature
- Pressure
- Vibrations
- Rotations

6.1 Methodology

Most sensitivity measurements of the calcium system presented in this chapter utilize a direct-scaling noise measurement facilitated by the two channels of the 16-bit analog-to-digital conversion (ADC) voltage sampling electronics, which will now be explained in detail. The noise source being investigated is parameterized by some voltage metric, which I will call the test or external variable in this discussion. Simultaneous measurements of the test variable and the calcium photodetector output are performed during a large, square-wave type modulation, as depicted in Fig. 6.1. Often the atomic signal displays an overall linear drift due to the 657 nm cavity, which is easily subtracted from the data array. Spurious values are often seen at the beginning and end of the step, as the external variable was often actuated by hand, and thus data values at the very beginning and end of the square wave are typically omitted.

Figure 6.1: The direct-scaling noise analysis method relies on measuring the relative scaling factor between the test variable and the calcium detector response, as measured on our 16-bit ADC system. This is valid so long as the step size within both channels greatly exceeds the noise of either channel.



Each data segment is then averaged and labeled by some x_i for the photodetector signal

and y_i for the test variable. To account for variations in the human-actuated square wave, the sensitivity factor, S , is calculated for each step as

$$S_i = \left| \frac{x_{i+1} - x_i}{y_{i+1} - y_i} \right|. \quad (6.1)$$

The individual scaling factors are then averaged to yield a final sensitivity factor, which relates a change in the test variable, Y_{test} , to a change in the digitized photodetector signal, Y_{Ca} , given by

$$S = \frac{Y_{Ca}}{Y_{ext}}. \quad (6.2)$$

The sensitivity factor can be calculated using the atomic response either off- or on-fringe, corresponding generally to amplitude or amplitude, frequency, and phase fluctuations, respectively. To arrive at the test variable's noise contribution, an extended measurement of the stationary value is taken, scaled by the sensitivity factor S , and further scaled by the LSB/Hz factor. The Allan deviation of this data represents the fractional frequency noise that can be attributed to the test variable.

6.1.1 Ca2 measurement electronics notes

Clearly, the noise assessment method here relies heavily on our 16-bit digitized micro-controller measurements communicated to the Labview front panel, as was introduced in the Ca1 chapter. As such, some basic notes will now be given on specifics as applied to Ca2, building off the previously established terminology and subsystems. The Ca2 version of the processing electronics primarily differed from Ca1 in use of a 16-bit ADC instead of the previous 8-bit. Both systems were developed by NIST colleague Richard Fox.

For the Ca2 iteration, voltages between values of approximately 4.60 – 4.74 V can be digitized for one ADC channel while the other goes from 4.58–4.71 V. These windows originally chosen to fall near typical PMT voltages seen with Ca1 when using blue detection. The improvements in imaging and photon number from Ca2 quickly came to require low-noise pre-amps with additional voltage offsets in order to fall within the ADC window. The impact of this choice can be seen by considering its impact on the calcium frequency servo's locking capability. Were the 1.6 kHz

FWHM fringes to take up the entire window, then the average measurement LSB would correspond to a fractional frequency change of $(1.6 \text{ kHz})/[(2^{16} \text{ LSB})(456 \text{ THz})] = 5 \times 10^{-17}$, which was well below the calcium system performance at the time.

Though it might initially seem that these electronics would provide sufficient resolution for our measurements while allowing the entire fringe signal to fit within the measurement window, this was not the case in practice. To achieve sufficient frequency resolution from the ADC, the fringe pk-pk amplitude had to be much larger than the ADC window to account for noise inherent to the pre-processing electronics prior to the ADC. A flow chart of the electronic chain is depicted in Fig. 4.18 back on page 130. As a battery provides the quietest electronic source we might hope to mimic (shot-noise limited by quantization of the electron charge), replacing the signal in the flow chart just after the low-noise SRS570 current pre-amp with a comparable battery voltage shows the practical measurement limitations. The standard deviation of the processed battery typically converged around 40 LSB. Yet a 40 LSB fluctuation in the atomic signal for this example would then also correspond to a fractional frequency change of 2×10^{-15} , far above the performance we ultimately achieved.

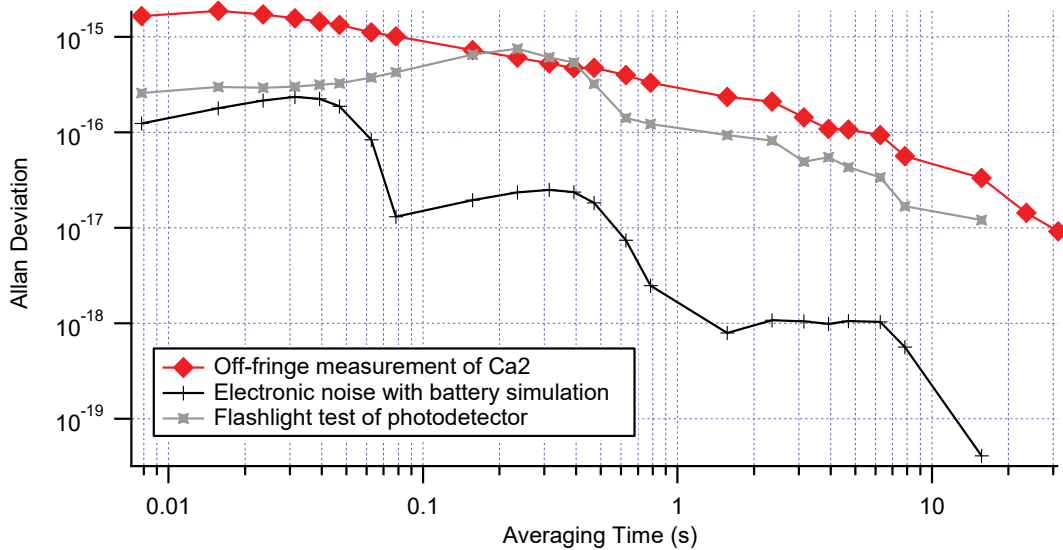
The small and offset voltage window of the ADC thus required the fringe signal to be greatly amplified and offset in order to achieve the necessary fringe peak-to-peak amplitude (thus resolution) and still fall within the ADC window. This accounts for why so many of the fringe images given in this dissertation are either digitized values from Cal's 8-bit configuration, taken from low-resolution oscilloscope traces, or very zoomed in images of small portions of fringes.

6.1.2 Off-fringe, i.e. amplitude noise considerations

One of the most helpful tools when working with the calcium system is the ability to roughly divide noise processes into those that change either the amplitude or the frequency/phase of the fringes. The amplitude portion includes all noise that affects the level of the Doppler-pedestal to the outside of the fringe components. The smallest amplitude noise contribution we might hope to achieve from our atomic ensemble can be simulated using a white light source, e.g. a flashlight,

to incite the same photodetector response level as when operating with Ca2. These flashlight tests thus mimic the photon shot-noise level of our detection scheme. They originally estimated fractional frequency instabilities at one second in the mid to low 10^{-17} regime. However, as time went on, the white noise level of the PMTs increased beyond what would be expected. Furthermore, this increase was seen with nearly all of the PMTs tested. A plot of the off-fringe noise, photon shot noise, and battery simulated electronic shot noise are given in Fig. 6.3.

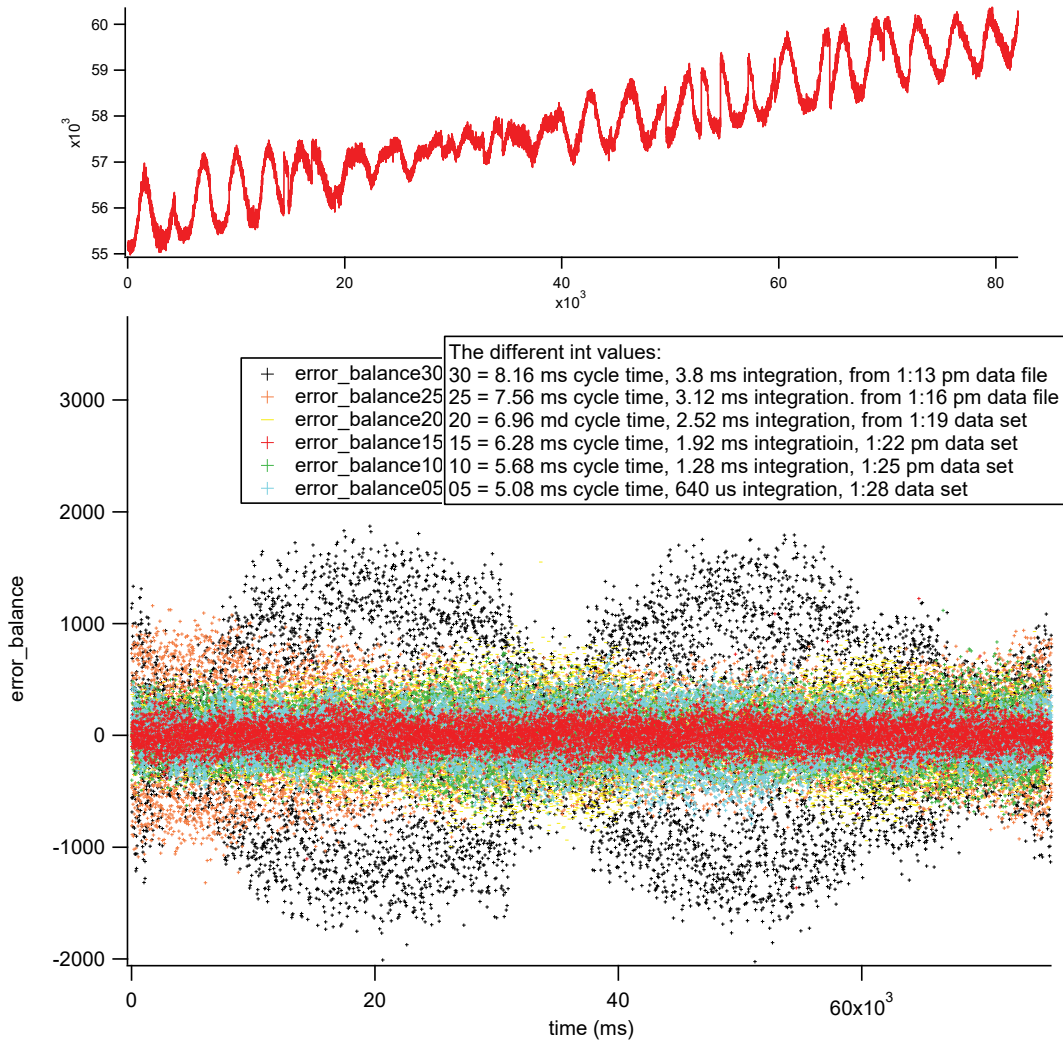
Figure 6.2: Allan deviations indicating the fractional frequency instability contributions from the off-fringe measurement (red), flashlight test of the detector (gray), and of the electronic noise simulated with a battery voltage. The amplitude noise contribution closely followed the simulated photo-detector signal. Data taken 5/15/2018.



The exact reason for the increase in the photodetector and measurement system's amplitude noise was never fully understood, though similar $> 10^{-16}$ at 1 s performance was seen with both the PMTs and photodiodes. The flashlight noise performance would intermittently dip back into the 10^{-17} decade for brief periods of time but never remained there long enough to take advantage of. Malfunctioning electronic components, ground loops, and increased electrical pickup could all have come into play. At first it was not easy to see the electronic noise on the atomic signal, as the cycle timing from the MCU would routinely fluctuate on the μ s scale. These timing errors masked

what became clear noise aliasing, which varied with the cycle time and integration period as seen in Fig. 6.3. The flashlight measurements displayed similar oscillatory behavior as well.

Figure 6.3: The top graph displays the time series of digitized photodiode voltages in LSB that were collected while Ca2 was locked with a single oven with point number on the horizontal axis. The bottom displays the difference values of adjacent photodiode points, which are roughly proportional to the frequency correction. Clearly, the cycle and integration time impact the noise content of the frequency corrections. Data taken 5/31/2018.



Lastly, as more and more signal photons were collected from the atoms, the photodiode white noise level should have increased somewhat because the total number of charge carriers also

increased. However, the larger LSB/Hz scaling factor from collecting more photons should have caused the total photodetector noise contribution to be the same or lower.

Assessment of noise sources will now be given and conclusions drawn about the most direct way that Ca2 could have further improved its performance. These largely rely on measurements of the amplitude noise contribution of these sources, as the goal at the time was to find and reduce the noise source impeding performance below 10^{-16} . However, relevant frequency and phase concerns are also addressed.

6.2 657 nm Ramsey-Bordé laser noise

The 657 nm laser system used for Ramsey-Bordé interrogation can introduce noise through a wide variety of mechanisms. Some of these were rather subtle and difficult to assess, such as mode-competition within the laser spectrum. Because the properties of the laser determine the process through which atoms go in Ramsey-Bordé, any variations in the laser's frequency, phase, or form can impact the fringes used for frequency corrections.

6.2.1 Frequency content

The level of necessary pre-stabilization for the Ramsey-Bordé interrogation laser can be seen in the requirement that the laser phase variation during the measurement be $\ll \pi$, or else coherence would be lost). The average calcium atom from our ovens is moving somewhere around 600 m/s, and thus takes approximately 300 μ s to traverse our interferometer. Thus, a laser linewidth much less than $\approx 1/(300 \mu\text{s}) = 3 \text{ kHz}$ is needed for fringes to remain coherent for atoms at that speed. Of course, linewidths far more narrow than this are clearly more ideal, and the laser linewidth was seen to clearly have an impact on the obtainable Ca2 frequency instability. To test this, the 657 nm laser instability could easily be increased by nearly a perfect decade, to 2×10^{-14} at 1 s. The Ca2 instability during such periods was impacted, also by nearly an order of magnitude. Stabilization of the 657 nm laser to the superior Yb cavity via frequency comb would have been an interesting

test to see if improving the 657 nm instability would have had significant impact.

Issues with frequency mode fluctuations of the 657 nm laser plagued the experiment for some time, as discussed in Ch. 5.2.1 back on p. 147, capable of bringing the noise the Ca2 instability into the 10^{-15} decade at times. With a mode-hop free tuning range ≥ 8 GHz, the mode competition incited by PZT changes of the diode laser ECDL was found to be small enough to fall below the electronic shot-noise of our system. Though retaining such a large tuning region required regular adjustments, the system could be run for many hours without needing to tweak the 657 nm interrogation laser. Often if the calcium system was underperforming, relocking the red laser to its cavity or minutely adjusting the ECDL grating position would return typical Ca2 frequency stabilization performance in the mid to low 10^{-16} 's.

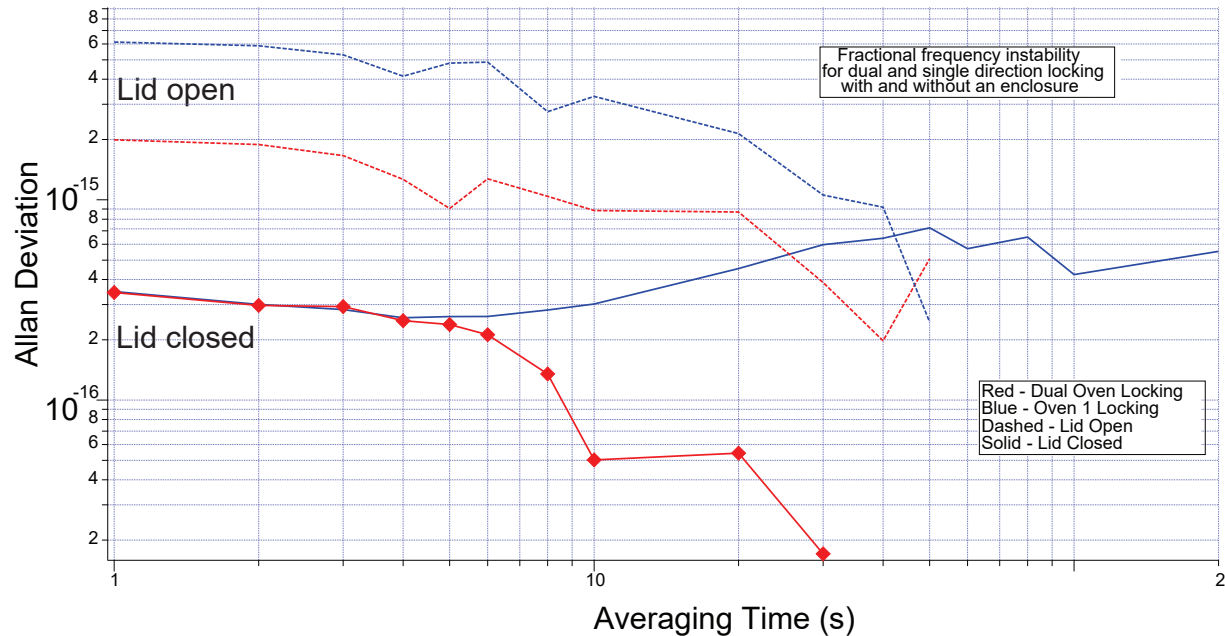
6.2.2 Air fluctuations

Phase noise on the interrogation laser is particularly difficult to assess. The original Ca2 interrogation laser path encompassed approximately 20" of in-air path uncompensated by the phase lock, and performance during this time was largely still limited to the low 10^{-15} decade at 1 s of averaging. This path was eventually shortened by a factor of two, and performance in the high 10^{-16} range was achieved at 1 s without making other changes to the Ca2 system. When an acrylic enclosure was then made for this optical path by post-doc Roger Brown, who worked part-time in the Ca2 lab for a few months, the calcium stabilization performance dropped to the mid 10^{-16} level at 1 s. However, reducing the path length in air much further than this was impractical with commercial optomechanics.

Using the input optics assembly based in commercial optomechanics, we were able to draw some interesting conclusions as to how air impacted the interrogation laser's ability to interact with the atomic beam. These instability results are depicted in Fig. 6.4 to compare Ca2's resulting stabilization performance with or without a lid on the 657 nm input optics chamber as well as how dual versus single oven locking handled these changes. Looking at the open lid case, it is clear that some air fluctuations occur on timescales ≤ 40 seconds that could be clearly compensated for with

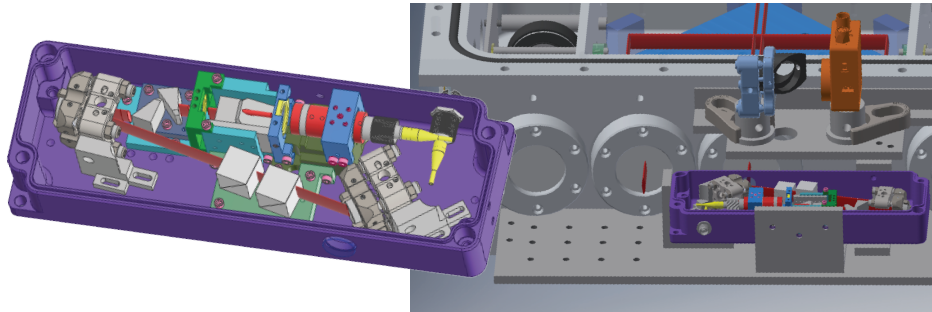
dual ovens. The disparity of the 1 s ADEV values here implies that fluctuations are also significant below 1 s, and that the dual oven locking can still compensate for them somewhat. However, exposing this input optics assembly to laboratory atmosphere still limited the Ca2 performance, as the lid closed case is significantly improved. With the lid closed, the noise between 1–4 s (and likely for a range of time <1 s) is common to both single and dual oven locking schemes and very close to the measured off-fringe amplitude noise. Thus, it is entirely feasible that the air fluctuations contribute noise below the lid-closed levels for the dual oven case over all times and the single-oven case for short times. The likely cause of the upturn in the lid-closed, single-oven case is the temperature increase caused by sealing off the input optics, wherein the laser warms its environment over time. This causes the opto-mechanics to expand slightly, inducing a residual first-order Doppler shift. Such a shift can be compensated for with the dual ovens, but cannot with the single oven using our measurement protocols

Figure 6.4: Comparison of four instability measurements of Ca2 for both dual and single oven locking schemes also both with and without a lid on the 657 nm input optics enclosure. The short-term disparity between dual and single ovens without a sealed enclosure indicates that air currents can be somewhat mitigated by dual ovens. However, with the enclosure open the instability of the system was clearly limited by air currents. See text for more.



Because sealing the input optics enclosure had such a dramatic impact on the Ca2 stabilization performance, and because the air fluctuations had previously been a limiting noise source, it seemed prudent to further reduce the input optics path so as to ensure that air fluctuations were no longer problematic. The final result of this endeavor is shown in Fig. 6.5, which contains all of the same optical elements, but fit within a significantly more compact enclosure of $\sim 200 \text{ cm}^3$ with an optical path length just over half the length of our earlier one. Despite obtaining similar fringes and running the calcium system as similarly as possible to the earlier instability measurements, we did not see improved Ca2 frequency stability performance. Thus we were able to conclude that air currents were not the limiting noise source, and so must contribute noise $< 2 \times 10^{16}$ at one second for our Bordé fringes. Furthermore, the smaller input optics chamber was less sensitive to whether or not the enclosure had its lid on, typically producing nearly identical Allan deviations for their resultant fractional frequency instabilities. This suggests that, with the lid on, the air fluctuations in the optical path length for the reduced input optics chamber should contribute noise well below the Ca2 system performance level.

Figure 6.5: An angled fiber delivers the cavity-pre-stabilized 657 nm light to the input optics chamber, where a butt-coupling with another angled fiber leads to a flat faced fiber tip attached to an adjustable collimation element. The small back-reflection off the flat fiber face is used to generate the phase-noise cancellation error signal. The laser beam exiting the collimation element is immediately passed through three anamorphic prism pairs, which were hand aligned to maximize the fringe contrast through controlling the vertical and horizontal beam waist dimensions. Two prism pairs are used to extend the vertical beam dimension, as they are able to transmit a far greater portion of the 657 nm power than a single prism pair could do. Next the laser is reflected off the first steering mirror, passed through a polarizing beam splitter then a pickoff for the intensity servo light. Then the final steering mirror directs the laser out through an anti-reflection coated, wedged window.



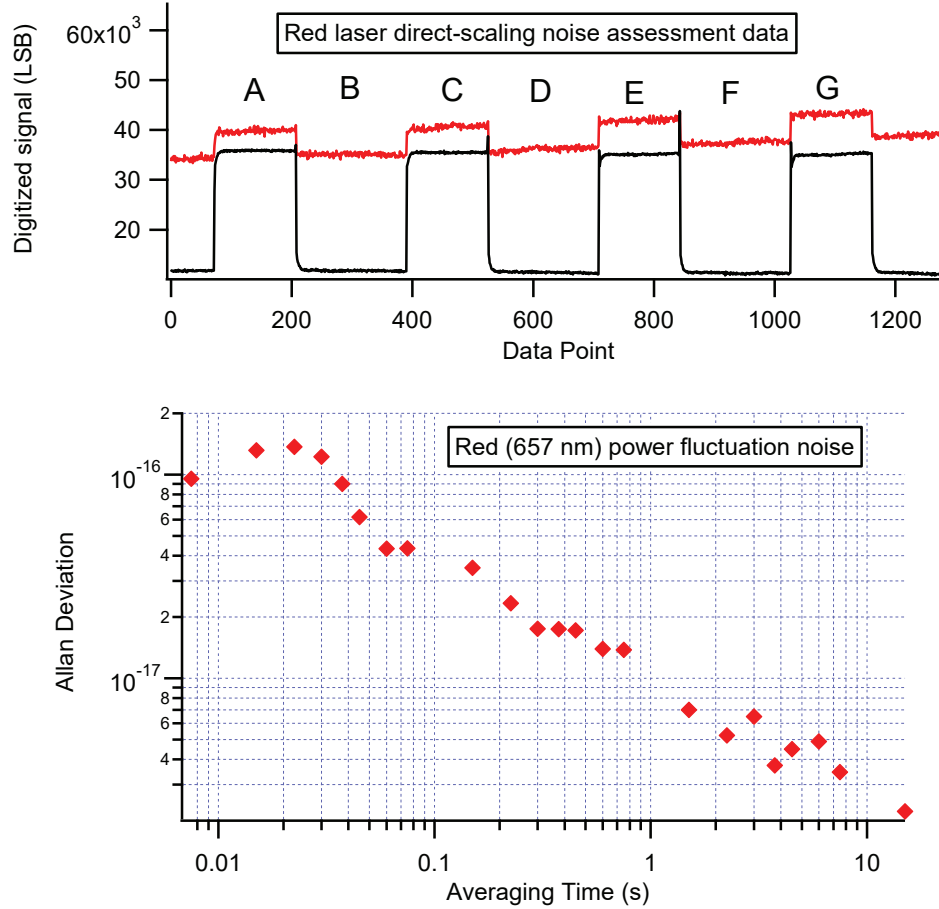
Another option, which was never implemented with much success, was to flood the normally air-filled ($n=1.000292$) optical path enclosure with gaseous helium ($n=1.000035$) [116] to reduce the laser's sensitivity to its atmosphere. However, no difference was found between a helium flooded beam enclosure and standard air-filled enclosures. Difficulties caused by temperature swings from cold helium from a compressed gas cylinder made it difficult to draw strict conclusions. This difficulty was still found even when the helium was first brought to room temperature in under-inflated mylar balloons before being forced into the shortened input optics chamber of Fig. 6.5.

6.2.3 Intensity

The red laser intensity is one of the easiest noise contributions to assess with a well-calibrated laser intensity servo. In this case, a pickoff from the red input optics chamber is compared to a low-noise voltage reference, and a correction proportional to their difference is applied via an AOM. The intensity servo boards designed by Jeff Sherman also include TTL inputs that allow switching between two set points. This function was used to provide the step function to make a very easy direct scaling measurement of the red intensity noise. To reduce the calcium atomic beam's intensity sensitivity, the laser power was typically held at 25 mW, far into the saturation regime.

The 657 nm intensity noise contribution is the first noise source assessed in this chapter with the direct-scaling method, and thus for the red intensity measurement, the square wave modulation is depicted in Fig. 6.6. The red laser noise can be seen to be quieter than the total system noise just through its square wave modulation, as the intensity step size is much larger than the atomic response and displays superior signal-to-noise. The scaling factor was still calculated and applied to a separate long measurement of the un-modulated intensity metric and additionally scaled to frequency units with the LSB/Hz conversion factor before being Allan deviated.

Figure 6.6: The direct-scaling data to calculate the sensitivity factor is given in the upper graph wherein the digitized LSB values are recorded for the calcium response (red) and an out-of-loop monitor of the red power level (black), in this case as the monitored voltage of our intensity servo. Below is the corresponding ADEV for the red intensity contribution scaled by the sensitivity factor and LSB/Hz value measured for the fringes just prior to the measurement. Thus Allan deviation then indicates that fluctuations in the red power levels are not a limiting amplitude noise process for our system.



Thus, with the direct-scaling noise measurement, ascertaining a red-intensity fluctuation contribution to amplitude noise at one second of approximately 1×10^{-17} was a fairly straight forward process.

The additional impact of the 657 nm laser power on the fringe frequency was measured with a similar technique, by modulating the optical power and monitoring the locked calcium frequency. The sensitivity factor was found to be 9×10^{-15} per mW.

6.2.4 Angular sensitivities

To mitigate the problems caused by the 657 nm laser traveling through air, the Ca2 system was designed around a monolithic block of ultra-low expansion glass with optically contacted mirrors. However, because the angular sensitivity of the Ramsey-Bordé fringes was extremely sensitive in our experience, first the angular tolerances of the system had to be tested. Of course, it is very difficult to change the angle of a laser beam without also causing some finite displacement. Thus, our angular sensitivity measurements most likely overestimate the effect, as the displacement would additionally impact the fringe contrast. However, tests were carried out anyway using the input optics steering mirrors, which were adjusted repeatedly by ± 150 μrad for the vertical and ± 200 μrad for the horizontal while the calcium-stabilized frequency was monitored via frequency comb. Vertical sensitivity was measured at 3.5×10^{-16} per μrad while horizontal was $< 5 \times 10^{-17}$ per μrad .

6.3 431 nm laser noise

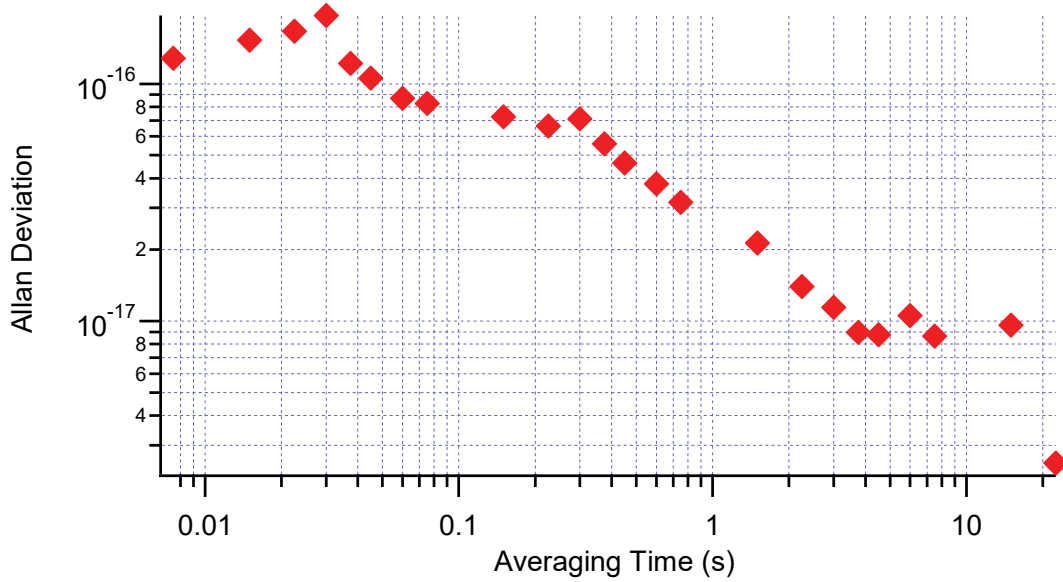
The blue laser's interaction with the atomic beam does not rely on the laser phase or coherent processes, and thus many of the complications inherent to the previously discussed red laser system are not present here. The main parameters of concern for the blue laser are its frequency, physical orientation, and intensity.

The IR source that was doubled to generate 431 nm light was able to be assessed using the frequency comb system. The cavity-stabilized IR source was found to have a drift of approximately 200 Hz/s, with a 1 s Allan deviation fractional frequency instability of 8.5×10^{-14} and a 0.1 ms instability of 9.9×10^{-14} . As to how this might impact the calcium frequency corrections, in a 5 ms interrogation time, the IR could be expected to drift less than 2 Hz (so less than 4 Hz for the blue). Such a small change upon the ≈ 40 MHz transition resonance was calculated to contribute noise at the 5×10^{-20} level at one second.

The impact of blue intensity fluctuations were assessed similarly to the red intensity measurement previously discussed. At one second, blue intensity noise was found to contribute noise

at a level of only 2.8×10^{-17} , far below the total calcium system's measured noise. A plot of the full Allan deviation is given in Fig. 6.7. The measured fractional frequency sensitivity to blue power was found to be 4.7×10^{-15} per mW.

Figure 6.7: Direct-scaling method noise assessment of blue intensity fluctuations on the off-fringe measured amplitude noise of the calcium system. Data taken March 29, 2018.



Air fluctuations in the blue laser path were not found to contribute noise at the 10^{-16} level or above, as inclusion of an enclosure around the blue input optics resulted in no changes to the calcium frequency correction performance.

6.4 Magnetic fields

The magnetic fields experienced by atoms in the calcium Ramsey-Bordé system all fall well within the weak-field regime. We utilize the first-order Zeeman shift to spectroscopically separate the 3P_1 ($m=\pm 1$) levels away from the desired $m=0$ level for Ramsey-Bordé. However, the $m=0$ level does have some non-zero quadratic sensitivity to magnetic fields, which must be considered for the magnetic-field sensitivity of the 657 nm transition. Meanwhile, the first-order shift for the ($m = \pm 1$) levels is drastically larger, making easy to spectroscopically separate their resonances

from the ($m=0$) component.

The magnetic field shifts for the ($4s, 4p$) $m = \pm 1, 0$ have been calculated for the intermediate magnetic field regime by Beverini, Maccioni, and Strumia in [27] as

$$\delta\nu [(4s, 4p)^3P_1(m = +1)] = \mu_0 g_1 B - \frac{1}{4}\epsilon_2 B^2 + \frac{3}{5}wB^2, \quad (6.3)$$

$$\delta\nu [(4s, 4p)^3P_1(m = 0)] = \frac{1}{3}(2\epsilon_1 - \epsilon_2)B^2 + \frac{4}{5}wB^2, \quad (6.4)$$

$$\delta\nu [(4s, 4p)^3P_1(m = -1)] = -\mu_0 g_1 B - \frac{1}{4}\epsilon_2 B^2 + \frac{3}{5}wB^2, \quad (6.5)$$

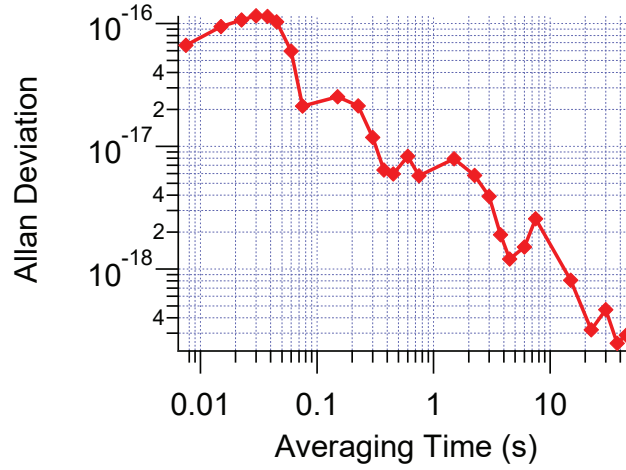
where $\epsilon_1 \approx 125$ MHz/T² and $\epsilon_2 \approx 61.6...$ MHz/T² are calculated from the fine-structure energies in [244] and experimentally verified in [27]. The factor $\mu_0 = eh/2m = 13.996...$ GHz/T is the Bohr magneton, and $w = 5.58 \times 10^5$ Hz/T² is a coupling parameter found via Hartree-Fock calculations [27].

For our experiment, the red zone magnetic field was found to produce the optimum fringe signal and contrast with a magnitude near 700 μ T, which yields a predicted shift of 31 Hz for the ($m=0$) state. In general, the second order coefficient for the calcium clock transition on 1S_0 – $^3P_1(m=0)$ is approximately 64 MHz/T² [206]. Thus, to maintain performance at the 1×10^{-16} level (ie. to less than 0.0456 Hz), then the magnetic field fluctuations need to be limited below about 27 μ T in a single measurement period, or to better than a 3.9 percent fluctuation.

The current passing through the solenoid coils was sampled and processed with low-noise current and voltage pre-amps (models SRS560 and SRS570) to reasonably fall within the ADC voltage window. Not all standard voltage or current controls in a laboratory setting would be quiet enough for the task, and multiple current sources were tested before we found one satisfactory.

The resulting red magnetic field noise contribution, parameterized for digitization by the driving voltage (related to the driving current via the fixed circuit resistance) can be seen in the Allan deviation of Figure 6.8. Additionally, it can be noted that the distinct oscillations in the Allan deviation are indicative of some oscillatory noise process. The 1 s noise contribution with this method was found to be 6.5×10^{-18} , indicating that the red magnetic field is sufficiently quiet.

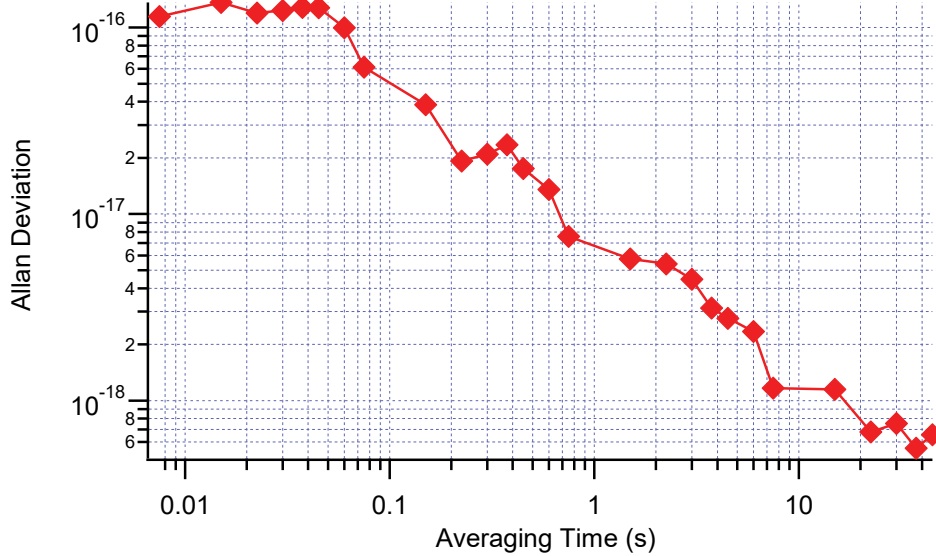
Figure 6.8: The magnetic fields in the Ramsey-Bordé zone of the Ca2 apparatus were found to contribute off-fringe noise at very low levels far below the total noise of the calcium system.



The frequency impact of the red magnetic field was also measured, by monitoring the induced frequency fluctuation to the locked calcium system via frequency comb. The sensitivity was found to be 1.8×10^{-15} per Gauss or 1.8×10^{-11} per Tesla.

The blue magnetic field calculations are somewhat less clear than the red, as the field strength in addition to laser's linear polarization angle and intensity couple into the atomic signal. However, the direct-scaling method made calculation of the blue-zone magnetic field noise contribution to the off-fringe signal. The one second contribution fell at 7.5×10^{-18} , and the full ADEV is given in Fig. 6.9.

Figure 6.9: The magnetic fields in the blue detection zones of the Ca2 apparatus were found to contribute off-fringe noise at very low levels far below the total noise of the calcium system.



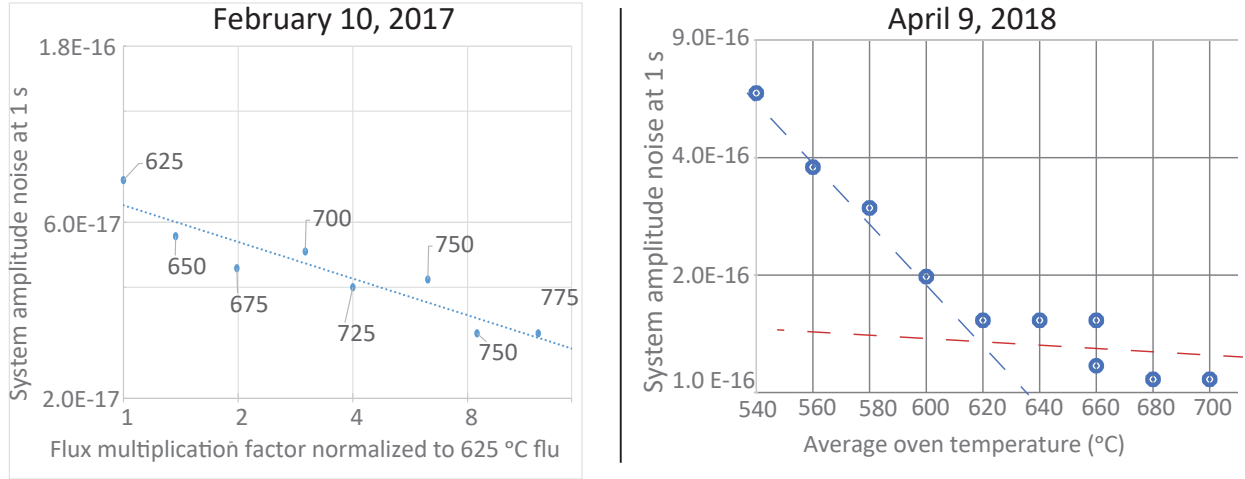
The frequency impact of the blue magnetic field was also measured, by monitoring the induced frequency fluctuation to the locked calcium system via frequency comb. The sensitivity was found to be 1×10^{-14} per Gauss.

6.5 Atomic beam and ovens

Though sometimes it is useful to run the calcium ovens at different temperatures, we generally ran the calcium ovens with the nozzle end (where calcium exits) at 650 °C and crucible body servo'd to 600 °C. However, this caused the front heater to do the majority of the work, and later the standard became a nozzle at 635 °C and crucible at 615 °C. This prevented clogging and we could still generally treat the calcium temperature as being the average, 625 °C.

Whereas the oven temperature versus off-fringe amplitude noise had once been measured to be far below the calcium-stabilized laser's fractional frequency instability, over the course of a year the system came to display a disparate response. This is depicted in Fig. 6.10 for similar data taken over a year apart. Clearly, some fundamental noise properties of the Ca2 measurement at 1 s had changed between these two tests.

Figure 6.10: Two measurements of the off-fringe Ca2 system noise versus oven temperature taken 14 months apart. The earlier plot, actually plotted against measured flux change, displayed a roughly linearly decrease in noise, whereas the later plot shows that both the noise was greater for the same oven temperatures and that the linear trend ceased somewhere around 620 °C with a comparatively flat response between $1\text{--}2 \times 10^{-16}$. This plateau was common to most off-fringe calcium measurement. Thus, for the cycle times used in the later plot, the Ca2 quantum projection noise appears to fall below the flicker floor near 10^{-16} , prompting me to look into shortening the cycle time.



As the source of the short-term noise near 10^{-16} could not be reduced by improving strengthening the atomic signal, there wasn't much of a way forward in terms of reducing this noise by changing the Ca2 apparatus or its lasers. However, if the calcium system were to make frequency corrections faster, then perhaps it could reach this limit more quickly. And clearly, for oven temperatures above about 640 °C, we could sacrifice some of the atom number and still hope to reach the existing flicker floor (the red trace of Fig. 6.10). The earlier section discussing the electronics highlighted some of the noise problems encountered while trouble-shooting this problem.

The calcium ovens possess a reasonably large thermal load, and thus temperature swings of the calcium source within the oven crucible take time. At 625 °C, an oven temperature fluctuation of $+0.1$ °C would induce a second-order Doppler shift of Bordé fringes by 0.04 Hz, or $\sim 8.8 \times 10^{-17}$, given by the change in fringe location for the most probable atomic speed in both cases. However, temperature fluctuations occur slowly compared to the cycle time of our measurements (4 - 30 ms), so should not be a significant noise contribution at short time scales.

Other concerns with atomic beams include sputtering, density variations, and collisions, none of which were found to be limiting factors. The calcium ovens are run in the laminar flow regime, and no differences in a single oven's locking frequency were found between when the second counter-propagating oven had significant flux levels and when it did not.

The other area of concern is in the transverse velocity distribution fluctuations, as determined by the fine apertures on either side of the central Ramsey-Bordé zone and earlier discussed in Ch. 3.4.2.1 on page 93. Motion of these apertures during the experiment, such as might be caused by thermal expansion or contraction of the vacuum chamber walls or the apertures themselves, is not thought to be a limiting noise contribution to the experiment, but could be a source of long-term drift.

6.6 Vacuum pressure

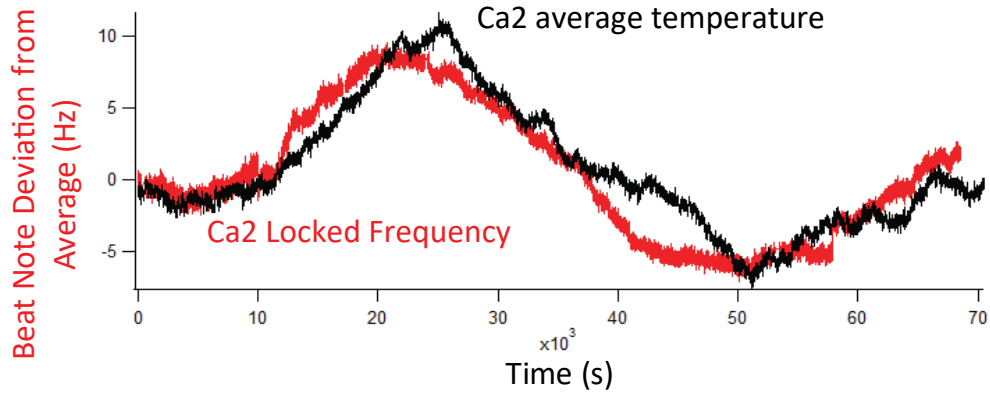
Measuring the vacuum pressure impact on the Ca2 frequency stabilization performance was difficult as it could only be done with the experiment by turning the ion pump off for a fixed period of time and then analyzing the frequency excursions as the pump returned the system to its initial conditions. However, the impact of pressure tests often took many hours to full dissipate. The measured sensitivity, of 1.6 Hz per 10^{-7} Torr, was larger than hoped for, but monitoring of the Ca2 pressure during normal operation did not indicate fluctuations large enough to critically impact the instability performance.

6.7 Temperature shielding and long-term measurements

During spring of 2017, with the help of visiting Ph.D. student Todd Sheerin, a thermal shield was tested on the Ca2 apparatus for a few weeks, during which some of our best data was collected. The temperatures of RTD's affixed to various locations on the Ca2 apparatus and thermal shield were monitored and compared against the calcium-stabilized frequency response. Prior to this, monitoring the temperature swings of the apparatus were found to correlate strongly with the longer-term frequency fluctuations also seen via frequency comb measurements. The temperature

stabilization system was implemented in a short period of time as a proof of principle and overall did significantly reduce the variation in the Ramsey-Bordé stabilized laser frequency. For example, with the temperature shield, the residual thermal fluctuations still correlated well with the Ca2 frequency deviations, as seen in Fig. 6.11 over nearly 20 hours of data.

Figure 6.11: Over nearly 20 hours of data, the Ca2-stabilized frequency deviated by less than 15 Hz while the temperature swing encompassed an approximately 50 mK frequency range.



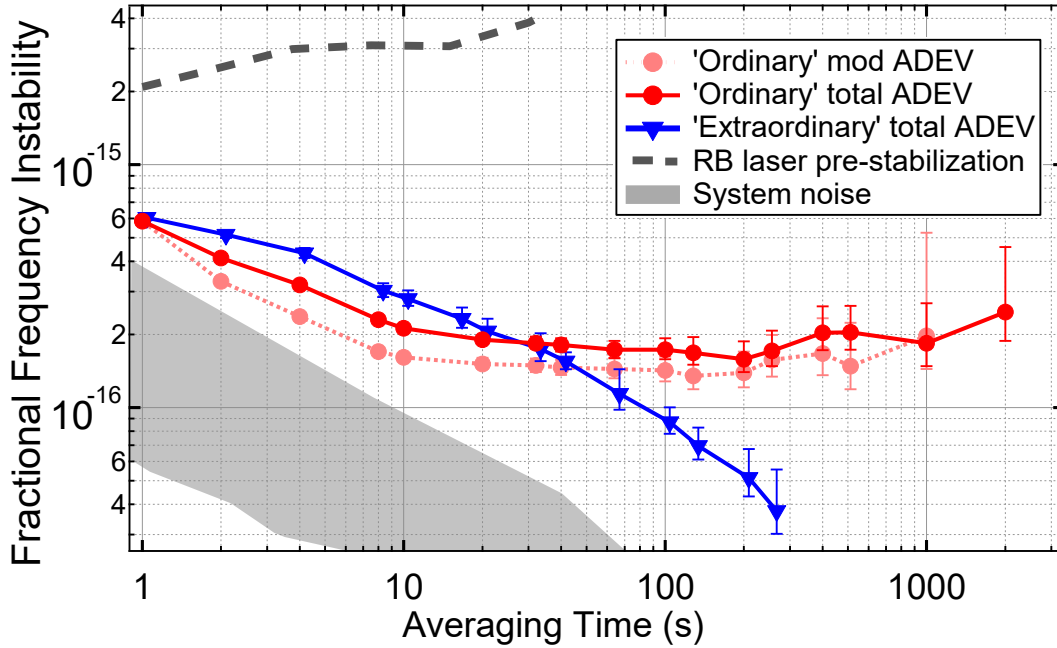
With the removal of the approximated sinusoid of the frequency variation, the subsequent Allan deviation never reaches into 10^{-15} . Numerous smaller sections of this long data set also exhibit very good noise properties for our system, with typical performance of $4\text{--}6 \times 10^{-16}$ at 1 s and averaging to between $1\text{--}2 \times 10^{-16}$. These tests made it clear that temperature drifts of the Ca2 apparatus could easily be responsible for the long-term rise in the long-term frequency instability.

Also during this time period, basic temperature sensitivity of the Ca2 apparatus was measured by attached heaters to the apparatus and monitoring the change in locked calcium frequency. A sensitivity was calculated at approximately 10^{-16} per 6 mK. Since temperature stabilization below the mK level is routinely obtained for cavity and other thermally sensitive systems, regular performance averaging to the 10^{-17} decade should be feasible.

6.8 Representative instability results

From many years of data, I present in Fig. 6.12 two plots that are representative of the two performance regimes we repeatedly saw from Ca2. These two plots are also discussed in detail within the journal article submission underway [195].

Figure 6.12: Two Ca2-stabilized frequency measurements are given here with 1σ error bars. The system performance regularly seen when Ca2 was running properly is given by the ‘Ordinary’ (red circles) data set, whereas ‘Extraordinary’ (blue triangles) highlights shorter data segments exhibiting exceptional performance, averaging below 10^{-16} . The RB laser pre-stabilization is shown after a 2 Hz/s drift removal, though this drift is present during calcium system operation. The measured range of frequency instability contributions from detection noise sources falls within the gray region.



The ‘ordinary’ data set of this plot displays the performance usually obtained when running the calcium system for any extended period of time before changing drift levels cause the Allan deviation to turn up at long time scales. The most characteristic feature of this plot is perhaps the extended flicker noise floor between $1\text{--}2 \times 10^{-16}$. While the cause of this noise is not entirely certain, the ability of the shorter data sets to sometimes surpass this limit does provide clues as to the flicker noise source’s origin, which we believe to be caused by thermal fluctuations of the Ca2 chamber.

Particularly, the thermal sensitivity was visually most correlated with Ca2 frequency fluctuations when the temperature of the 657 nm input optics assembly was monitored. Changes in these optics can easily couple into the fringe properties, as discussed in the earlier angular deviations section.

Numerous Ca2-stabilized frequency measurements were found to average well into the 10^{-17} decade, similar to the 'extraordinary' data set of Fig. 6.12. These sets shared a few commonalities. Firstly, all lasted for periods of time no longer than 20 minutes, with 10 minutes being far more common before the Allan deviation result would begin to rise to the flicker floor of the 'ordinary' data. Second, these sets were particularly steady in their drift levels over all timescales, which is what largely enabled them to continue averaging down into the 10^{-17} decade for longer averaging intervals than were typically seen. We theorize that these extraordinary data periods happened to occur when the Ca2 system was in a state of thermal equilibrium or at a turning point in a temperature cycle, which is why such data was less commonly seen and able to avoid the low 10^{-16} flicker floor. This assertion is further supported by extraordinary-type data being found more regularly at the end of longer measurement days, wherein the Ca2 system had already been running continuously for many hours.

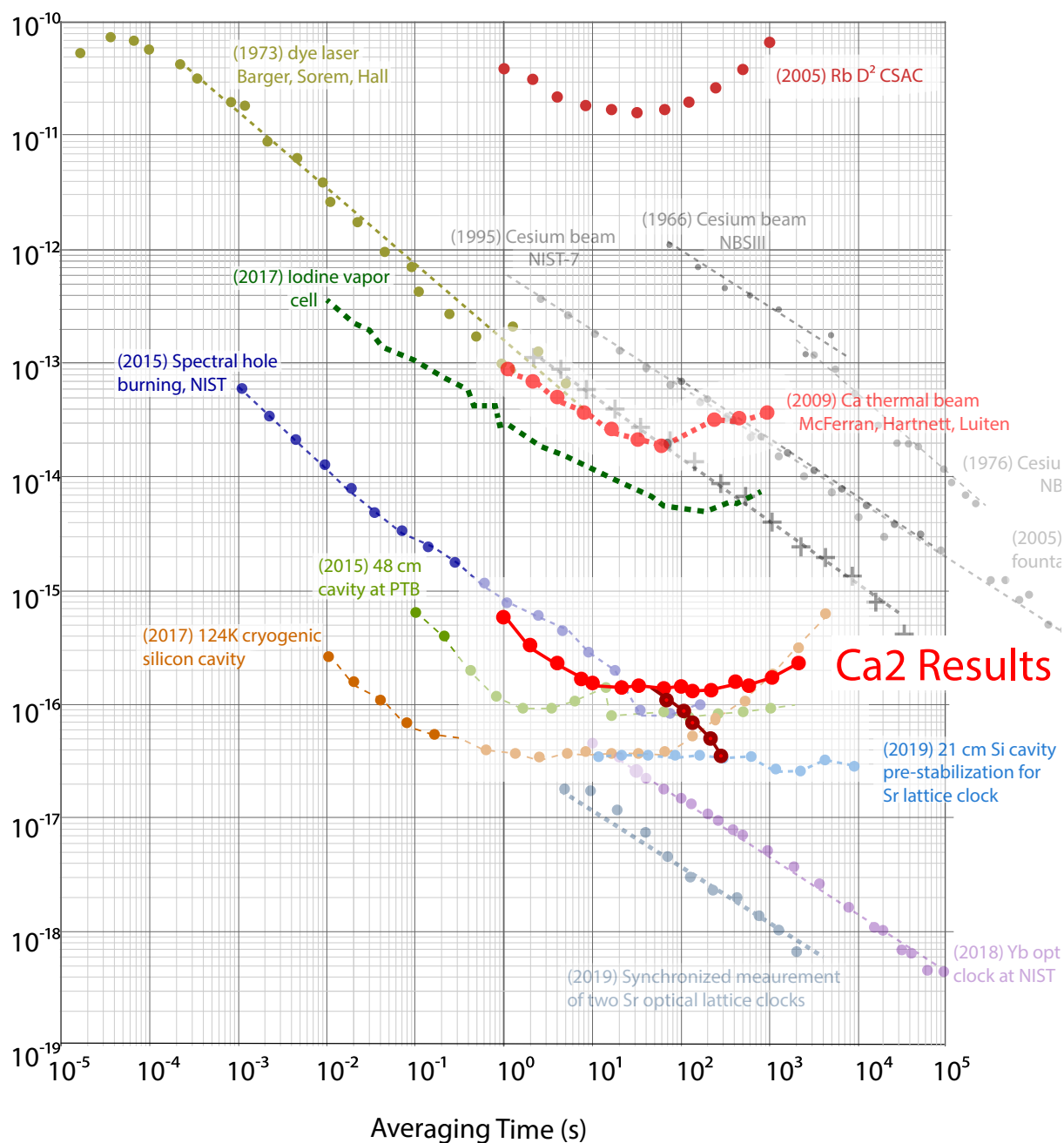
The type of thermal stabilization that might be needed for the Ca2 system to regularly average into the low 10^{-17} fractional frequency instability decade is nothing extreme compared to the temperature stabilization common to other ultra-stable frequency sources. Notably, advanced Fabry-Perot cavity systems often rely on multiple layers of thermal shielding and temperature stabilization to maintain a stable environment. To offer a baseline, commercially sold cavities offer temperature stabilization at or below a few mK, which would clearly benefit the Ca2 system here.

Additionally, we must emphasize that the Ramsey-Bordé method utilized here was quite effective without the use of vibration isolation, as is also common and typically required for advanced cavity systems. Similar results to those presented here were regularly seen both with and without the optics bench floated. Moving the Ca2 apparatus onto a vibration isolation platform within an acoustic shield did nothing to improve its performance, implying that the system's sensitivity to these is fairly low and that their noise contributions fall below the obtained system performance.

To put these results in the context of the field, I return to the earlier Fig. 2.12, given here again but with the Ca2 results included as Fig. 6.13.

Figure 6.13: The final Ca2 instability results presented here show improvements of 1-2 orders of magnitude over other thermal ensemble results. Furthermore, the frequency stability obtained from Ca2 is comparable to many advanced cavity systems but far less sensitive to temperature and vibrational effects.

Fractional
Frequency
Instability*



The obtained Ca2 fractional frequency instabilities presented here represent an improvement over the best comparable thermal beam Bordé-Ramsey results of [180] by a factor of more than 200. The Ca2 results are more than an order of magnitude improved over other ‘simple’ atomic systems not relying on slowing or trapping of their atoms. Furthermore, with the low quantum projection noise afforded by applying Bordé-Ramsey spectroscopy to a high-flux thermal beam, additional improvements to the calcium system performance are feasible. Lastly, the long-term frequency stability and reasonable accuracy of our atomically referenced interferometer offer a unique performance regime inaccessible to optical cavities.

Chapter 7

Future Prospects

7.1 Improving upon Ca2

Though there is room for improvement over Ca2's performance, the Ca2 system was still able to demonstrate extremely impressive frequency stabilization at levels never before seen from a thermal ensemble and that rival many advanced cavity stabilization systems used today.

How I would design a Ca3

The changes between Ca1 and Ca2 were largely influenced by their practicality given the small number of people actively involved in the calcium experiment. Using what was learned from Ca2, a fictional Ca3 apparatus is now considered through a series of possible minor modifications to the existing Ca2 system.

If more flux or a lower atomic shot noise limit were desired, the atomic beam could be redesigned. It is entirely feasible that a wider and taller calcium beam could be utilized, as these aspects were not tested with either Ca1 or Ca2. Additionally, an array of calcium beams could be potentially utilized to take advantage of the $1/\sqrt{N}$ instability scaling. By having each atomic beam pass through its own set of fine apertures, which could be made deeper than those utilized in Ca2, the velocity distributions of the atomic beam could be unaltered while improving the total number of signal atoms. This change could be carried out initially by having new crucible nozzles and fine apertures machined, which would still be compatible with the existing Ca2 apparatus. Yet one could easily imagine a much larger scaling up of this method.

Another set of improvements may come from shortening the optical path length of the 657 nm laser source within the vacuum, as the optical path length far greater than the beam's Rayleigh range accentuates optical aberrations and causes the laser profile to differ between the four laser-atom interactions of the interferometer. In theory, a monolithic ULE retro-reflector could be made that is more compact, with all reflecting surfaces closer to the atomic beam. However, machining of such a structure with the same angular tolerances utilized in Ca2 could be technically challenging. Yet the benefits of the smaller apparatus size may be worth considering for commercial applications.

One particularly interesting change worth considering would be to incorporate an adjustable phase element in the interrogation laser path. The equal and opposite phase relation between the two ovens' fringes would allow the relative laser phase to be set such that the locking features of both ovens align in frequency space. Though some finite offset might still exist between the spectroscopic peaks or troughs, such offsets could be accounted for in post-processing of the photodetector signal. Ultimately, aligning the fringe features in frequency space would allow both fringes to be interrogated at the same time. We could thus make simultaneous measurements of both ovens rather than interleaved measurements, which improves the QPN of our atomic signal. The difference frequency between the fringe components could then be actuated with the phase element instead of the current AOM use, keeping the fringes overlapped in frequency space throughout long measurement campaigns.

Some evidence suggests that further lowering the vacuum pressure levels may be beneficial to the system's locking properties, as some frequency dependence on the vacuum pressure was clearly seen. If the pressure were lowered far beyond the 10^{-7} Torr level at which Ca2 was run, then the corresponding noise from its fluctuations or from background gases would also be reduced. However, this may be somewhat redundant if the 657 nm optical path were also shortened.

The solid angle collection of 431 nm photons from the atomic beam could also be greatly improved. Larger optical elements would be one obvious step forward. A half domed elliptical mirror could be designed to curve around the atomic beam, almost as if it were half an integrating sphere with additional apertures for the atomic beam. Something similar could be done on the lens

side, perhaps with another mirrored integrating hemisphere as before but with the addition of a lens element along the imaging axis. Not only would such a system greatly increase the collected solid angle, but it would cause many more blue photons to pass through the atomic beam at a wide range of angles, potentially allowing better 431 nm photon cycling across our velocity distribution while also requiring less input power (though this effect would be very small compared to the 431 nm laser powers typically used in Ca2). Imaging of the fluorescence would be easier were the windows leading to the photodetectors closer to the atomic source. A shorter optical path along the imaging axis could reduce the severity of off-axis aberrations at the plane of the detector, thus condensing into a smaller and more predictable area for photodetection. Of course, ample photons are already collected per atom, but the more efficient collections scheme would allow for use of a wider range of detector quantum efficiencies and cause less problems were the atomic velocity distribution to be shifted toward higher temperatures.

Some additional elements to facilitate certain measurements could also be incorporated into a fictional Ca3 vacuum chamber. Notably, incorporation of an atomic beam block would have allowed easier and more concrete assessment of the dual atomic beam interactions. In Ca2 this could only be assessed by changing the oven temperatures, which couple to numerous other systematics. Incorporating adjustable-width fine apertures that could be controlled within the vacuum environment could allow the optimal atomic beam dimensions to be more quickly analyzed. Lastly, the use of a flux-measurement device in vacuum could allow concrete measurements of atom number or even be used to assess the atomic velocity distribution. Some of these devices rely on monitoring the response of a sensitive crystal resonator that predictably changes in frequency as mass is deposited on it, allowing calculation of the atomic deposition rate, thus the flux. Additionally, beam flux measurements relying on absorption from the 1S_0 - 1P_1 Ca cooling transition at 423 nm have been quite successful at analyzing the atomic beam flux and velocity distribution [188, 228].

As was explored in the previous chapter, the short-term Ca2 fractional frequency instability could benefit from decreased cycle times without being limited by atom number. The minimum cycle time should fall shortly above the time it takes for an average speed atom to traverse

the interferometer and reach the detection zone, which, for 600 m/s calcium atoms, is around 600 μ s for our apparatus, which would be approximately 10 times faster than most Ca2 integration times. Even were we able to integrate for such a short period, the frequency correction cycle would be dominated by dark time due to the finite millisecond-level communications with the Labview program. This overhead could be reduced if the relevant variables were reduced to their minimum required bit size or replaced with mechanical binary switches, which would entail the transfer of less data. As ADC and MCU technology is improved, both the bit resolution and the communication protocols could be expedited. Furthermore, with the use of a higher bit digitization, a much larger portion of the calcium spectroscopic signal could be observed, which would be useful for assessment and characterization.

Clearly, some issues with pickup and electronic noise exist within the measurement system used here, which were measured at levels that limit Ca2 performance at short time-scales. Careful design of the photodetection and trans-impedance amplification stages with ample shielding from the environment and very short connections would be important.

Phase control possibilities

In the Ca2 system of this dissertation, we had no direct control over the interrogation laser's relative phase, which determines the Bordé fringe phase. Yet some benefits could be gained were the experiment able to control the fringe phase somewhat, as alluded to in the previous section. In addition to unlocking the ability to simultaneously measure the fringes from both ovens, phase control could also allow better assessment of the fringe envelope. The fringe envelope provides information about the velocity distribution and other dephasing mechanisms, which could be useful for system assessments and verification of experimental parameters. Furthermore, because the fringe envelope is independent of the fringe phase, the envelope could provide a useful metric for measuring and discerning phase fluctuations in the laser path. With a single atomic beam, the fringe peak relative to the envelope peak could also be used to track and compensate for variations in the optical path without the need for a second oven. Ultimately, such measurement could also be

used to improve the frequency accuracy of single-oven systems, as uncertainty in the fringe phase can easily couple in uncertainties at the kHz level. Lastly, were the atomic system able to assess the fringe envelope and adjust the fringe phase such that the locking feature occurs at the very peak of the envelope, then the signal-to-noise properties of the measurement would be maximized for the given system parameters.

In addition to the inclusion of some phase control element within the single traveling laser beam path, similar control could be exerted instead with two or more independent but phase-locked traveling waves. Such a change could also allow multiple laser geometries to be tested, such as four parallel beams, which provides a different type of interferometer. Actuating the laser phase with traditional phase-lock servos might also afford easier and faster control than could be found with an in-beam phase actuator, along with reducing the possibilities for the in-beam optic to add unwanted noise through vibrations or thermal effects. However, dividing the laser path into multiple individual paths would require significantly more total laser power. Yet, the additional benefits from shorter overall laser paths could also enable narrower 657 nm waists and more uniform laser interactions at each of the four laser-atom interactions. Ultimately this might translate to better fringe contrast or higher signal-to-noise within the atomic beam.

Aside from the average phase control options just discussed, control could also be exerted over the phase distortions caused by wavefront deformities. Explorations of such MEMs-based control systems have shown promise, such as in the atom interferometry experiments of Mark Kasevich [157]. In theory, by establishing highly planar wavefronts, fringe contrast could be improved and motional effects limited.

Atomic beam improvements

There remains a great deal of flexibility in atomic beam sources compatible with Ramsey-Bordé techniques. Improvements to an effusive thermal beam like used here generally aim to increase the atomic flux and/or make the velocity distribution more manageable. One of the earliest examples of this was the use of a supersonic SF_6 molecule beam, which had fairly uniform velocity distribution

[39]. This allowed them to improve the frequency accuracy of the resolved optical Ramsey fringes. Other methods involve slowing the thermal beam, such as with a Zeeman slower or radiative cooling. Some other atomic species that are heavier than calcium could also run at lower temperatures, resulting in slower velocity distributions with similar flux levels. In fact, many atomic and molecular ensembles have been studied that could provide particularly enticing flux levels near our thermal beam levels [67, 237]. Strontium sources generally seem well-suited for lower temperature oven regimes than our calcium system while still obtaining comparable flux levels [230]. Discharge beam sources could allow for ten times the atomic flux but come at the cost of size for such fast moving atoms to have reasonable Ramsey times [44].

Another approach to increasing the atom number without sacrificing fringe properties could be obtained via an array of atomic beams, equally spaced and interrogated by the same lasers in a similar configuration to that used here. Though the instability reduction by $1/\sqrt{N}$ isn't as favorable as a pure linear decrease, it would still provide a fairly straight-forward avenue toward reduced atomic shot noise. Such an array may also be used so that the oven temperatures could be run lower, though the vapor pressure curves for calcium and physical complications of adding additional atomic beams would prohibit this being reasonable in extreme cases.

Measuring away from DC

One common way to reduce noise in precision measurements involves moving the measurement away from a static, DC value. As most noise contribution decrease with higher frequencies (as was discussed in Chapter 2), measuring a the system response in a narrow frequency band away from DC often reduces the total noise of the measurement. For the calcium system, this could be done by chopping or pulsing the blue laser beam used for the fluorescent measurement. The typical cycle time of an excited state atom to emit a photon is less than 10 ns, meaning that we might expect to be able oscillate the blue laser power on and off at a rate at 10's of MHz, far above DC.

Of course, such a measurement would then also require high-speed photodetectors capable of resolving such a high frequency signal. Yet large area photodetectors like those used in the

current Ca2 system possess an inherently large capacitance, which can reduce the ability of the photodiode to respond quickly. It is not immediately clear that the noise reduction from moving the measurement to higher frequencies would be worth the loss in photon number. However, if high-bandwidth atomic systems like Ca2 further pursued, it could be that moving away from DC measurements may provide the most direct route to reducing measurement or electronic noise contributions at short-times.

7.2 Tests of exotic phenomena

One particularly exciting new application of atomic clocks in recent years has been in the area of dark matter detection. Very little is known about dark matter, but theorists have been able to propose a few candidates that could represent dark matter. One of these assumes that dark matter is composed of axionic particles, which they would expect to clump into sheets or large masses throughout the universe. The presence of such dark matter could possibly couple to the fine structure constant or other physical variables, which could then be detected by ultra-sensitive atomic clocks and frequency sources, such as the Ca2 apparatus. To our knowledge, Ca2 is one of the highest bandwidth atomic systems capable of achieving fractional frequency instabilities in the $\leq 10^{-16}$, which would allow it to sample a unique parameter space of dark matter detection.

Another interesting use for the calcium Bordé-Ramsey apparatus lies in its potential to search for King's plot nonlinearities. These plots rely on measurement of at least four even isotopes of a given atomic species. In the case of ^{40}Ca , numerous stable isotopes exist in reasonable abundances, including ^{42}Ca , ^{43}Ca , ^{44}Ca , and ^{46}Ca with ^{42}Ca and ^{44}Ca being the most abundant of the isotopes. The isotopes are fairly well studied [122]. Furthermore, because the Bordé-Ramsey measurement does not require cooling or trapping, utilizing the calcium isotopes is fairly straight-forward. Measuring the $^1\text{S}_0$ - $^3\text{P}_1$ ($m=0$) transition frequency along with either the $^1\text{S}_0$ - $^1\text{P}_1$ or $^3\text{P}_1$ - $^3\text{P}_0$ transition frequency for three or more of the calcium isotopes is expected to yield a linear relation between the relative measured frequency ratio. Discrepancies from the linear trend could indicate physics beyond the standard model, and the uncertainty in the linearity provides an upper boundary as to

the scope of possible new physics. Such King’s plot nonlinearity studies have been the subject of recent papers focusing on the utility of atomic clocks for such measurements [22, 79].

7.3 Position, navigation, and timing

The promising long-term instability potential of the Ca2 interferometer could be useful for future time dissemination systems or even as a gyroscope for navigation. For instance, GPS holdover is becoming increasingly important as technology relies more on GPS in a time when such systems can be easily spoofed or disabled. Systems like Ca2 could be important for future defense infrastructure warding against GPS or timing disruptions, ranging from the military to the electrical grid. Similarly, local ultra-precise networks are being developed that might one day allow emergency responders to pinpoint the exact room or floor from which a cellphone made a call. While Ca2 is not the most promising for its accuracy, it would be a great optical regime candidate source to act as hydrogen masers do for microwave atomic clocks defining modern day time standards. Such an optical flywheel would be essential to a future optical frequency standard.

Atom interferometers have also been well-explored as gyroscopes [103, 78, 53, 140]. Though point-source atom interferometry type experiments [72, 126] are more typical, the thermal beam orientation used here did demonstrate obvious rotational sensitivity. Though not severe enough to limit the frequency instability results ultimately obtained, rotations of the optics bench did visibly couple into the locking frequency of the calcium apparatus (Ch. 4.17, p. 129). Obviously more study would be required to understand the sensitivity and utility of such an orientation, but, given the relatively high bandwidth compared to many atomic sources [227], Ramsey-Bordé may be worth additional attention. The Ramsey-Bordé thermal beam system has been well-tested for rotational measurements, which readily couple to the atomic signal via the Sagnac effect, which was first studied on atom interferometers (called atom-wave gyros) similar to ours back in 1991 [216]. One could imagine a Ramsey-Bordé type instrument that relied on multiple orthogonal atomic beam sources (or perhaps a very dense vapor cell), which could then be used to precisely measure generalized rotations in space.

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Appendix A

The 1984 Bordé *et al* PRA Publication Excerpt

This appendix is a reprint of the 1984 Bordé *et al* article that is frequently referenced throughout this dissertation. It is provided here for clarify and as a reference. Only pages 1-7 of the 13 page paper are included, as the remaining information largely discusses the pseudospin representation and photon echo processes that were not directly utilized in modeling the Ramsey-Bordé interferometer.

Optical Ramsey fringes with traveling waves

Ch. J. Bordé, Ch. Salomon, S. Avrillier, A. Van Lerberghe, and Ch. Bréant

*Laboratoire de Physique des Lasers (Laboratoire No. 282 associé au Centre National de la Recherche Scientifique),
Université de Paris—Nord, F-93430 Villetaneuse, France*

D. Bassi

Istituto per la Ricerca Scientifica e Tecnologica, I-38050 Povo, Italy

G. Scoles

Department of Chemistry, University of Waterloo, Waterloo, Ontario, Canada N2L 3G1

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We present a new interaction geometry for optical Ramsey fringes comprised of four traveling waves instead of the three usual standing waves. First, we demonstrate experimentally that the new method leads to an improved contrast, using the optothermal detection of the vibrational excitation of SF_6 in a supersonic beam illuminated by a waveguide CO_2 laser. Second, we give a simple theoretical description of the method, using evolution matrices of spinors and pseudospin-vector representations of these spinors. Finally, we introduce strong-field density-matrix diagrams to discuss the differences between the various interaction geometries as well as between the Ramsey fringes and the usual stimulated photon echoes.

I. INTRODUCTION

As soon as the finite transit time was identified as the basic limitation to ultra-high resolution in saturation spectroscopy, it was suggested¹ to associate sub-Doppler techniques with the method of spatially separated fields introduced by Ramsey in the microwave domain around 1950.² The first realistic proposals came from Novosibirsk for saturation spectroscopy³ and for Doppler-free two-photon spectroscopy.⁴ In saturation spectroscopy the Russian method uses three equidistant standing waves and the early experimental demonstrations have used this interaction geometry.⁵⁻⁹ In an independent theoretical study, one of us showed that fringes resulted from successive interactions with four traveling waves^{10,11} and it became clear that one could also obtain fringes with four spatially separated traveling waves with a number of advantages over the three standing-waves method. In the first part of this paper, we present an experimental comparison between the standing waves and the traveling-waves interaction geometries, using an optothermal detection technique¹² to monitor the vibrational excitation in a supersonic beam of SF_6 . In the second part of the paper we give a detailed description of the new method in terms of spinors and of pseudospin trajectory. The difference with usual stimulated photon echoes¹³ is emphasized. A new graphical representation, the strong-field density-matrix diagrams, is introduced to discuss further this difference and to provide a direct comparison with the standing-waves case as well as with the mixed case introduced by Shimoda and co-workers.¹⁴

We shall see that with four traveling waves the physical origin of optical Ramsey fringes becomes especially simple to understand. Let us recall, that in the usual Ramsey interaction scheme, the induced polarization precesses

freely between two field zones. As a result of the phase comparison between this polarization and the field in the second zone, final populations oscillate as a function of detuning and time of flight between the two field zones. Unfortunately, in the optical domain, the distribution of first-order Doppler shifts averages out the fringes. To solve this problem, our interaction geometry is such that the previous usual Ramsey operation is simply repeated with a second pair of laser beams traveling in a direction opposite to that of the first pair. The populations which oscillate as a function of laser detuning and velocity after interaction with the first pair, give rise to two distinct contributions through interaction with the second pair. One for which the Doppler dephasing is canceled and for which only the oscillatory behavior with detuning subsists; this is the Ramsey fringes signal. One which loses its oscillatory behavior with detuning but for which the Doppler dephasing is doubled; this is the backward stimulated photon echo which averages to zero by velocity integration.

Ideally, a succession of $\pi/2$ pulses then results clearly in maximum signal for the fringes. As we shall see in detail the situation is far more complicated with standing waves for which the simultaneous interaction with oppositely traveling waves reduces the fringe contrast.

II. EXPERIMENTAL RESULTS

In order to investigate optical Ramsey fringes experimentally, we have used a supersonic SF_6 beam illuminated either by a triple standing wave or by four traveling waves from a waveguide CO_2 laser. The schematic diagram of the molecular beam apparatus can be found in Ref. 15. The supersonic beam is obtained by adiabatic expansion of 7 vol % SF_6 -He mixture through a 50 μm nozzle fol-

lowed by a 0.5-mm cross-sectional diameter skimmer located about 1 cm downstream from the nozzle. The supersonic beam then enters the interaction chamber where the background pressure is of the order of 3×10^{-6} Torr the presence of beam. The flow velocity of the SF_6 molecules estimated from the observed Ramsey fringe period is 940 m/s, in good agreement with time-of-flight measurements in similar conditions.¹⁶ In the first experiment the beam crossed the three standing waves generated from a single Gaussian beam (beam waist radius 6 mm) by a set of three slits 1 mm wide and 5 mm apart from each other and by a retroreflector [see Fig. 1(a)].

Before the introduction of this optical setup inside the interaction chamber, the laser beam waist had been carefully positioned on the molecular beam by optimizing the Rabi oscillations contrast as described in Refs. 15 and 17. In a second series of experiments two slits only were illuminated and the two transmitted waves were reflected by the triadron with a spatial offset equal to 15 mm in order to generate two pairs of traveling waves [see Fig. 1(b)].

The three components A_1, F_1, E of the ν_3 band $P(4)$ manifold can be reached with a waveguide CO_2 laser oscillating on the $P(16)$ CO_2 line at $10.55 \mu\text{m}$. This laser was slaved by a frequency offset—locking technique to a conventional low-pressure CO_2 laser which was itself locked to the $Q(43) F_1^8$ saturation resonance of SF_6 . This technique provides both a very good short-term spectral width (≈ 10 Hz) and the possibility to scan the frequency with an auxiliary RF synthesizer.¹⁸

After their interaction with the three or four light beams the molecules hit the liquid He-cooled surface of a silicon bolometer ($1 \times 5 \text{ mm}^2$) where they leave their internal energy. Since the laser beam is chopped, only the modulated rovibrational contribution of this internal energy is detected as a signal by standard lock-in techniques.¹² The bolometer is placed 48 cm away from the skimmer. Its electrical characteristics at 4.2 K are a 4×10^4 V/W responsivity and a NEP (noise equivalent power) of $8 \times 10^{-13} \text{ W}/\sqrt{\text{Hz}}$ at 30 Hz. However, in working conditions with a molecular beam, this NEP was observed to increase by one order of magnitude. Nevertheless as illustrated in Figs. 1(a) and 1(b) beautiful fringes have been obtained with both interaction geometries. These two figures also illustrate the large difference in fringe contrast (factor 2 to 3) which is obtained between these two cases. This difference is most easily understood in a theoretical approach, using transformation matrices, which we outline below.

III. THEORETICAL FRAMEWORK

Any realistic theory of optical Ramsey fringes should take into account transit effects, relaxation, first and second-order Doppler broadening and shifts as well as the recoil effect. All these effects are built in the following approach and no restrictive assumption is made concerning their respective importance. The individual atomic response is calculated as a function of molecular velocity and then averaged over initial formation conditions (velocity distribution or more generally phase-space distribution in a molecular beam).

We consider a two-level system interacting with a number of traveling waves which all have the same frequency but which may propagate in two opposite directions (+ or −). In the molecular frame of reference defined by $\vec{v} = \vec{p}_0/M\gamma$ for molecules formed in the lower level a with a momentum $\vec{p}_0$ [$\gamma = (1 - v^2/c^2)^{-1/2}$] we shall write the electric fields as

$$\vec{\mathcal{E}}(\vec{r}, t) = \text{Re} \hat{e}^\pm E^\pm U^\pm [v_x(t - t_1)] \times \exp[i(\gamma\omega t \mp kv_x t \mp kz + \varphi^\pm)], \quad (1)$$

where $\hat{e}$, E , U , and φ represent, respectively, the polarization vector, the amplitude, the transverse dependence and the phase of each wave. The fields are propagating along the $\hat{z}$ axis and for the sake of simplicity, in this paper, we shall ignore the y direction (molecular flow along the $\hat{x}$ axis) and assume that the function U is real and symmetric about the time t_1 . These fields couple together a large number of energy-momentum states as illustrated in Fig. 2.

In the $|\vec{r}\rangle$ representation, the wave-function components for each state $\alpha = a, b$ can be written as a discrete sum (Fourier series) where each component $\alpha_m(\vec{r}, t)$ corresponds to an energy-momentum state¹⁹

$$\langle \alpha | \psi(\vec{r}, t) \rangle = \sum_m \langle \alpha_m | \psi(\vec{r}, t) \rangle = \sum_m \alpha_m(\vec{r}, t), \quad (2)$$

where

$$\langle \alpha_m | \psi(\vec{r}, t) \rangle = \int_{m\hbar\vec{k}} d^3p \langle \vec{r} | \vec{p} \rangle \langle \vec{p} | \langle \alpha | \psi(t) \rangle$$

is an $\vec{r}$ wave packet centered about each momentum $m\hbar\vec{k}$ (m is an integer corresponding to the net number of momentum quanta exchanged with the field from $\vec{p}_0$). The molecular system can be described either by the spinor

$$\begin{pmatrix} \langle b | \psi(\vec{r}, t) \rangle \\ \langle a | \psi(\vec{r}, t) \rangle \end{pmatrix} = \begin{pmatrix} \sum_m b_m(\vec{r}, t) \\ \sum_m a_m(\vec{r}, t) \end{pmatrix}$$

or by a vector with an infinite number of components written as

$$\begin{pmatrix} \vdots \\ a_{m+2} \\ b_{m+1} \\ a_m \\ b_{m-1} \\ \vdots \end{pmatrix}.$$

The first description which was used in Refs. 20 and 21 is easier to deal with (2×2 transformation matrices) but is not as general as the second; for example, owing to the recoil effect a different energy

$$E_a + m^2(\hbar k)^2/2M = E_a + m^2\hbar\delta$$

is associated with each α_m and this correction is obviously easier to introduce in the second approach; the first approach requires successive rotating frame transformation which can be avoided in the second. In this paper we therefore emphasize this second formalism.

The Schrödinger equation can be written in matrix form

$$\begin{pmatrix} \vdots \\ a_{m+2} \\ b_{m+1} \\ a_m \\ b_{m-1} \\ \vdots \end{pmatrix} \frac{\partial}{\partial t} = \begin{pmatrix} \dots & -i\frac{E_a}{\hbar} - i(m+2)\delta - \frac{\gamma_a}{2} & 0 & 0 & \dots \\ \dots & 0 & -i\frac{E_b}{\hbar} - i(m+1)\delta - \frac{\gamma_b}{2} & 0 & \dots \\ \dots & 0 & 0 & -i\frac{E_a}{\hbar} - im^2\delta - \frac{\gamma_a}{2} & 0 \\ \dots & 0 & 0 & 0 & -i\frac{E_b}{\hbar} - i(m-1)\delta - \frac{\gamma_b}{2} & \dots \end{pmatrix} + i \begin{pmatrix} \vdots \\ \Omega_{ba}^- U^- e^{-i(\gamma_{\text{rot}} + kz + kv_z t + \varphi^-)} & 0 & \Omega_{ba}^+ U^+ e^{-i(\gamma_{\text{rot}} - kz - kv_z t + \varphi^+)} & 0 & \dots \\ \dots & 0 & 0 & \Omega_{ba}^- U^- e^{-i(\gamma_{\text{rot}} + kz + kv_z t + \varphi^-)} & 0 \\ \dots & 0 & \Omega_{ba}^+ U^+ e^{-i(\gamma_{\text{rot}} - kz - kv_z t + \varphi^+)} & 0 & \Omega_{ba}^- U^- e^{-i(\gamma_{\text{rot}} + kz + kv_z t + \varphi^-)} & \dots \\ \dots & 0 & 0 & \Omega_{ba}^- U^- e^{-i(\gamma_{\text{rot}} + kz + kv_z t + \varphi^-)} & 0 & \dots \end{pmatrix} \begin{pmatrix} \vdots \\ a_{m+2} \\ b_{m+1} \\ a_m \\ b_{m-1} \\ \vdots \end{pmatrix} \quad (3)$$

where we have made the rotating-wave approximation, where $\delta = \hbar k^2 / 2M$, where $\Omega_{ba}^\pm = \langle b | \hat{\mu} \cdot \hat{e}^{\pm*} | a \rangle E^\pm / 2\hbar$ are Rabi pulsations, and where γ_a and γ_b are relaxation constants associated with levels a and b . The following unitary transformation can be used to obtain a Hamiltonian matrix independent of space and time (apart from the slow time dependence of U):

$$\begin{pmatrix} \vdots \\ \tilde{a}_{m+2} \\ \tilde{b}_{m+1} \\ \tilde{a}_m \\ \tilde{b}_{m-1} \\ \vdots \end{pmatrix} = \exp \left[\frac{i}{2} (E_b + E_a) t / \hbar \right] \begin{pmatrix} \dots & e^{i/2[-\gamma_{\text{rot}} - 2(m+2)k(z+v_z t)]} & 0 & 0 & \dots \\ \dots & 0 & e^{i/2[\gamma_{\text{rot}} - 2(m+1)k(z+v_z t)]} & 0 & \dots \\ \dots & 0 & 0 & e^{i/2[-\gamma_{\text{rot}} - 2mk(z+v_z t)]} & 0 \\ \dots & 0 & 0 & 0 & e^{i/2[\gamma_{\text{rot}} - 2(m-1)k(z+v_z t)]} & \dots \end{pmatrix} \begin{pmatrix} \vdots \\ a_{m+2} \\ b_{m+1} \\ a_m \\ b_{m-1} \\ \vdots \end{pmatrix}, \quad (4)$$

The evolution equation for the transformed vector is

$$\frac{\partial}{\partial t} \begin{bmatrix} \vdots \\ \tilde{a}_{m+2} \\ \tilde{b}_{m+1} \\ \tilde{a}_m \\ \tilde{b}_{m-1} \\ \vdots \end{bmatrix} = \begin{bmatrix} \cdots & -\frac{i}{2}(\gamma\omega - \omega_0) - i(m+2)^2\delta - \frac{\gamma_a}{2} - i(m+2)kv_z & & 0 \\ & & 0 & \frac{i}{2}(\gamma\omega - \omega_0) - i(m+1)^2\delta - \frac{\gamma_b}{2} - i(m+1)kv_z \\ & & 0 & 0 \\ & & 0 & 0 \\ & 0 & 0 & 0 \\ & 0 & 0 & 0 \\ & -\frac{i}{2}(\gamma\omega - \omega_0) - im^2\delta - \frac{\gamma_a}{2} - imkv_z & 0 & 0 \\ & 0 & \frac{i}{2}(\gamma\omega - \omega_0) - i(m-1)^2\delta - \frac{\gamma_b}{2} - i(m-1)kv_z & \cdots \end{bmatrix} + i \begin{bmatrix} \cdots & 0 & \Omega_{ba}^- U^- e^{-i\varphi^-} & 0 & 0 & \cdots \\ \cdots & \Omega_{ba}^- U^- e^{-i\varphi^-} & 0 & \Omega_{ba}^+ U^+ e^{-i\varphi^+} & 0 & \cdots \\ \cdots & 0 & \Omega_{ba}^+ U^+ e^{+i\varphi^+} & 0 & \Omega_{ba}^- U^- e^{i\varphi^-} & \cdots \\ \cdots & 0 & 0 & \Omega_{ba}^- U^- e^{-i\varphi^-} & 0 & \cdots \end{bmatrix} \begin{bmatrix} \vdots \\ \tilde{a}_{m+2} \\ \tilde{b}_{m+1} \\ \tilde{a}_m \\ \tilde{b}_{m-1} \\ \vdots \end{bmatrix}, \quad (5)$$

where $\omega_0 = (E_b - E_a)/\hbar$.

IV. CASE OF SPATIALLY SEPARATED TRAVELING WAVES

In the case of interactions with purely traveling waves, the above Hamiltonian matrix can be broken into 2×2 sub-blocks and we may consider independently the evolution of spinors such that

$$\begin{bmatrix} \tilde{b}_{m\pm 1} \\ \tilde{a}_m \end{bmatrix}$$

for which the Schrödinger equation reduces to

$$\frac{\partial}{\partial t} \begin{bmatrix} \tilde{b}_{m\pm 1} \\ \tilde{a}_m \end{bmatrix} = \frac{i}{2} \left[\vec{\Omega} \cdot \vec{\sigma} + \Omega_0 \sigma_0 \right] \begin{bmatrix} \tilde{b}_{m\pm 1} \\ \tilde{a}_m \end{bmatrix}, \quad (6)$$

where the choice of sign is imposed by the direction of the wave and where we have introduced the Pauli matrices σ_0 ,



FIG. 1. (a) Ramsey pattern obtained for the $P(4) F_1$ SF_6 line with three standing waves (interaction geometry illustrated by the inset). The horizontal scale is linear in frequency and one fringe period corresponds to 94 kHz. The laser power was adjusted for optimum fringe contrast. The signal was recorded in a single one-minute sweep with a 0.1 s time constant and a 30 Hz modulation frequency of the laser amplitude. (b) Same as (a) with four traveling waves.

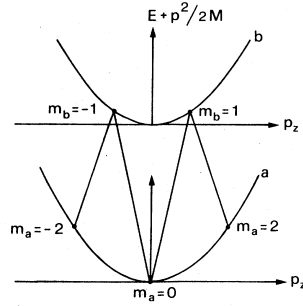


FIG. 2. Energy-momentum states of a two-level system coupled by counter-propagating laser beams.

$\vec{\sigma}$ and the effective field

$$\vec{\Omega} \begin{cases} \Omega_1 = 2\Omega_{ba}^\pm U^\pm \cos\varphi^\pm, \\ \Omega_2 = 2\Omega_{ba}^\pm U^\pm \sin\varphi^\pm, \\ \Omega_3 = \Delta \mp kv_z - [(m \pm 1)^2 - m^2]\delta + i(\gamma_b - \gamma_a)/2, \end{cases} \quad (7)$$

$$\Omega_0 = -[(m \pm 1)^2 + m^2]\delta - (2m \pm 1)kv_z + i\gamma_{ba}, \quad (8)$$

and where $\Delta = \gamma\omega - \omega_0$ and $\gamma_{ba} = (\gamma_b + \gamma_a)/2$.

If the field amplitude is constant in each field zone, $U^\pm \equiv 1$ and Eq. (6) is easily integrated from time t_0 to time t

$$\begin{bmatrix} \tilde{b}_{m \pm 1}(t) \\ \tilde{a}_m(t) \end{bmatrix} = \exp \left[\frac{i}{2} \Omega_0 \sigma_0 (t - t_0) \right] \exp \left[\frac{i}{2} \vec{\Omega} \cdot \vec{\sigma} (t - t_0) \right] \begin{bmatrix} \tilde{b}_{m \pm 1}(t_0) \\ \tilde{a}_m(t_0) \end{bmatrix}, \quad (9)$$

where the operator $\exp[i/2 \vec{\Omega} \cdot \vec{\sigma} (t - t_0)]$ is easily expressed as [parametrization formula of the group $SL(2C)$]

$$\exp \left[\frac{i}{2} \vec{\Omega} \cdot \vec{\sigma} (t - t_0) \right] = \sigma_0 \cos \left[\frac{\Omega(t - t_0)}{2} \right] + i \hat{n} \cdot \vec{\sigma} \sin \left[\frac{\Omega(t - t_0)}{2} \right] = \begin{bmatrix} A & B e^{-i\varphi^\pm} \\ C e^{i\varphi^\pm} & D \end{bmatrix} \quad (10)$$

with

$$\hat{n} = \vec{\Omega} / \Omega \quad (11)$$

and

$$\Omega^2 = \vec{\Omega}^2 = \Omega_3^2 + 4\Omega_{ba}^2 \quad (12)$$

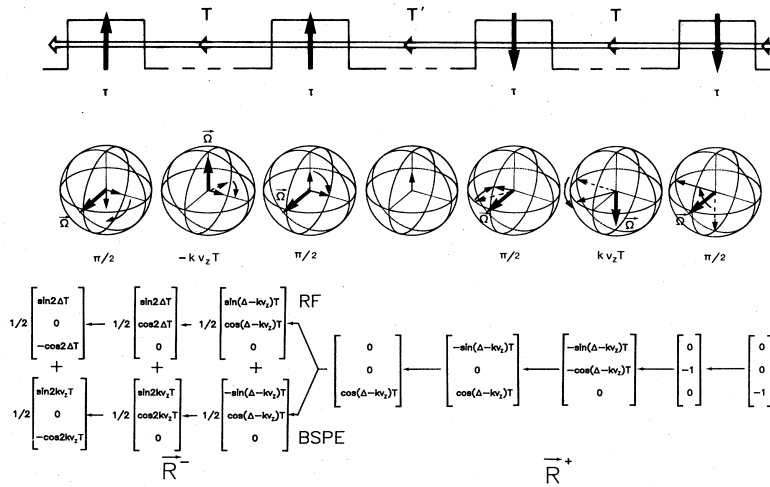


FIG. 3. Principle and illustration of Ramsey fringes (RF) with four traveling waves. At each step we give the three components of the relevant pseudospin $\vec{R}^\pm$. The backward stimulated photon echo (BSPE) would correspond to a vector $\vec{R}_{adv}$ (not represented) which doubles its Doppler dephasing.

and where

$$A = \cos \left[\frac{\Omega(t-t_0)}{2} \right] + i \frac{\Omega_3}{\Omega} \sin \left[\frac{\Omega(t-t_0)}{2} \right], \quad D = \cos \left[\frac{\Omega(t-t_0)}{2} \right] - i \frac{\Omega_3}{\Omega} \sin \left[\frac{\Omega(t-t_0)}{2} \right], \quad (13)$$

$$B = C = 2i \frac{\Omega ba}{\Omega} \sin \left[\frac{\Omega(t-t_0)}{2} \right].$$

We recover a 2×2 formalism which leads to very simple line-shape calculations by straightforward multiplication of 2×2 matrices corresponding to successive interaction or field-free zones. Let us now illustrate this with the case of the four traveling waves represented in Fig. 3.

In the first phase the interaction with the first two (+) traveling waves gives after the central dark zone

$$\begin{pmatrix} \tilde{b}_1 \\ \tilde{a}_0 \end{pmatrix} = \exp \left[\frac{i}{2} \Omega_0 (T' + T + 2\tau) \right] \begin{pmatrix} e^{i\Omega_3 T'/2} & 0 \\ 0 & e^{-i\Omega_3 T'/2} \end{pmatrix} \begin{pmatrix} A_2 & B_2 e^{-i\varphi_2^+} \\ C_2 e^{i\varphi_2^+} & D_2 \end{pmatrix} \begin{pmatrix} e^{i\Omega_3 T/2} & 0 \\ 0 & e^{-i\Omega_3 T/2} \end{pmatrix} \begin{pmatrix} A_1 & B_1 e^{-i\varphi_1^+} \\ C_1 e^{i\varphi_1^+} & D_1 \end{pmatrix} \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \quad (14)$$

where $\Omega_3 = \Delta - kv_z - \delta + i(\gamma_b - \gamma_a)/2$, $\Omega_0 = -\delta - kv_z + i\gamma_{ba}$ [see (7) and (8) with $m=0$]. Let us observe that a calculation of $\tilde{b}_1, \tilde{b}_1^*$ from this formula gives a generalization of Ramsey's formula including relaxation, first- and second-order Doppler effect as well as the recoil.²¹

In a second phase, in which the interaction takes place with two (−) field zones, we can independently describe the evolution of the spinors

$$\begin{pmatrix} \tilde{b}_1 \\ \tilde{a}_2 \end{pmatrix}$$

and

$$\begin{pmatrix} \tilde{b}_{-1} \\ \tilde{a}_0 \end{pmatrix}$$

from

$$\begin{pmatrix} \tilde{b}_1 \\ 0 \end{pmatrix} \text{ and } \begin{pmatrix} 0 \\ \tilde{a}_0 \end{pmatrix}.$$

The corresponding matrices are

$$\exp \left[\frac{i}{2} \Omega_0 (T + 2\tau) \right] \begin{pmatrix} A_4 & B_4 e^{-i\varphi_4^-} \\ C_4 e^{i\varphi_4^-} & D_4 \end{pmatrix} \begin{pmatrix} e^{i\Omega_3 T/2} & 0 \\ 0 & e^{-i\Omega_3 T/2} \end{pmatrix} \begin{pmatrix} A_3 & B_3 e^{-i\varphi_3^-} \\ C_3 e^{i\varphi_3^-} & D_3 \end{pmatrix}, \quad (15)$$

where

$$\Omega_3 = \Delta + kv_z - \delta + i(\gamma_b - \gamma_a)/2, \quad \Omega_0 = -\delta + kv_z + i\gamma_{ba}$$

for

$$\begin{pmatrix} \tilde{b}_{-1} \\ \tilde{a}_0 \end{pmatrix}$$

and

$$\Omega_3 = \Delta + kv_z + 3\delta + i(\gamma_b - \gamma_a)/2, \quad \Omega_0 = -5\delta - 3kv_z + i\gamma_{ba}$$

for

$$\begin{pmatrix} \tilde{b}_1 \\ \tilde{a}_2 \end{pmatrix}.$$

Combining the two sets of matrices we find

$$\begin{aligned} \tilde{b}_{-1}\tilde{b}_{-1}^* = & e^{-2\gamma_{ba}(T+2\tau)} e^{-\gamma_a T} \{ |B_1 C_2 B_3 A_4|^2 e^{-(\gamma_b - \gamma_a)T} + |B_1 C_2 D_3 B_4|^2 + |D_1 D_2 B_3 A_4|^2 + |D_1 D_2 D_3 B_4|^2 e^{(\gamma_b - \gamma_a)T} \\ & + [B_1 C_2 B_3 A_4 (D_1 D_2 D_3 B_4)^* e^{2i(\Delta - \delta)T} e^{i(\varphi_4^- - \varphi_3^- + \varphi_2^+ - \varphi_1^+)} + \text{c.c.}] \\ & + O(e^{\pm i k v_z T} \text{ or } e^{\pm 2i k v_z T}) \}, \end{aligned} \quad (16)$$

where the oscillating term $\exp[2i(\Delta - \delta)T]$ corresponds to the fringe pattern up shifted by the recoil shift δ , and

$$\begin{aligned} \tilde{b}_1\tilde{b}_1^* = & e^{-2\gamma_{ba}(T+2\tau)} e^{-\gamma_b T} \{ |B_1 A_2 A_3 A_4|^2 e^{-(\gamma_b - \gamma_a)T} + |B_1 A_2 C_3 B_4|^2 + |D_1 B_2 A_3 A_4|^2 + |D_1 B_2 C_3 B_4|^2 e^{(\gamma_b - \gamma_a)T} \\ & + [B_1 A_2 A_3 A_4 (D_1 B_2 C_3 B_4)^* e^{2i(\Delta + \delta)T} e^{i(\varphi_4^- - \varphi_3^- + \varphi_2^+ - \varphi_1^+)} + \text{c.c.}] + O(e^{\pm i k v_z T}, e^{\pm 2i k v_z T}) \}, \end{aligned} \quad (17)$$

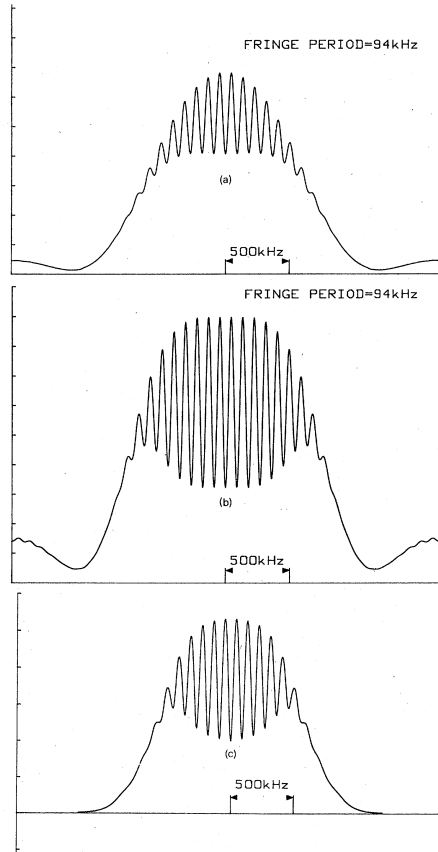


FIG. 4. Theoretical fringe patterns: (a) and (b) calculated from formulas (16) and (17) (without recoil and relaxation) with $T/\tau=5.25$ and $\Omega_1\tau=0.988$ for (a), $\Omega_1\tau=1.8$ for (b); (c) is calculated numerically for Gaussian laser profiles with a beam waist radius $w_0=0.5$ mm separated by 5 mm.

where the oscillating term $\exp 2i(\Delta + \delta)T$ corresponds to the fringe pattern down shifted by the recoil shift δ .

The signal is obtained by integration on v_z of the previous expressions. If the Doppler width is large compared to the fringe width, integrals of terms proportional to $e^{\pm i k v_z T}$ or $e^{\pm 2i k v_z T}$ are negligible; the v_z integrals of the terms in factor of $e^{2i(\Delta \pm \delta)T}$ give the fringe envelope and those of the remaining terms give the background broadened by the Doppler effect and by finite transit time. Figures 4(a) and 4(b) are typical graphs of the signal calculated from formulas (16) and (17) and numerically integrated over a flat velocity (v_z) distribution in the ideal case of four equal square pulses corresponding to $2\Omega_{ba}\tau=0.988$, for Fig. 4(a) and to the optimum value 1.8 for Fig. 4(b). (This optimum value is slightly higher than $\pi/2$ owing to the v_z dependence of Ω through Ω_3 .) In this last case the fringe contrast is close to $\frac{2}{3}$. A contrast of only 40% could be reached in the actual experiment [see Fig. 1(b)] for which the signal shape is better represented by Fig. 4(a). This can be explained by a number of practical reasons: distribution of Clebsch-Gordan coefficients; y distribution of the field, hyperfine structure of the lines, wave-front imperfections, Also to obtain the actual signal one should perform a final integration on the narrow (13%) distribution of velocity moduli which will reduce the size of side fringes.

The signal width has contributions from transit broadening and from Doppler broadening associated with the v_z distribution of molecules impinging on the bolometer. The corresponding parameters ($\tau, \Delta v_z$) are not very well defined in our experiment (because of light diffraction from the slits and of a lack of precise knowledge of the effective v_z distribution in the beam) and are coupled in a fit. To reproduce the signal background we have adjusted the time τ to an effective value $\frac{4}{5}$ of the value deduced from the slit width. Finally the overall shape of the signal is slightly better represented by a numerical calculation with Gaussian laser profiles as illustrated by Fig. 4(c) for the optimum value $2\Omega_{ba}w_0/v_x=1$.

V. PSEUDOSPIN REPRESENTATION

To understand in detail the origin of the fringes and their size, it is possible to follow the evolution of pseudospin vectors as illustrated in Fig. 3 in the ideal case of $\pi/2$

Appendix B

Mathematica model and Monte Carlo simulation

What follows is my Mathematica code to model a slightly simplified version of the Bordé process outlined in the 1984 Bordé et al publication in the previous Appendix section, applied to the specific geometry of the calcium Ramsey-Bordé thermal beam interferometer utilized in this dissertation. It begins by creating functions to calculate the excited state probability of each interferometer's isolated 2-level system, utilizing information about the laser intensity at each interaction, the longitudinal and transverse atomic velocity, and relativistic shifts. The velocities probability distributions are then modeled, with the longitudinal velocity by a Maxwell-Boltzmann distribution. Though not entirely accurate, the transverse velocity is modeled as a Gaussian distribution that is cut short at the calculated limits created by our apertures. However, more suitable approximations could be made.

While the model does not incorporate the Gaussian laser profile and phase distortions, it does allow for control over the phase seen in each zone. Furthermore, intermediate stages of the excited state probability for a single, 'most probable' atom are also given after each laser-atom interaction. The first reveals a Doppler profile with peak shifted from the bare resonance. The second creates oscillatory pattern over the earlier Doppler, scaled up to maximum probability of 1, as this is done for the most probable velocity, for which we tune the incident radiation to yield an ideal $\pi/2$ -pulse at each interaction. This gives conditions we associate with good fringe contrast. The third laser-atom interaction forces the two interferometers to be considered separately in their respective reference frames. The atomic signal is rather complex by the third and fourth interaction, but their

excitation probability is given. A zoomed-in view near the ensemble fringes is also given for the atom, showing how a single atomic state probability differs strongly from the ensemble average.

The last portion of the code utilizes a Monto Carlo simulation method to generate a large atomic ensemble where the entire possible atomic velocity space - both transverse and longitudinal - is propagated through the interferometer. A weighted average of the excited state probability as a function of the interrogation laser frequency is then given. Both fringe components were able to be resolved with fringe linewidths and shapes similar to what we actually find in our experiment. Though the model and my encoding of it is not perfect, it greatly improved my understanding of Bordé-Ramsey spectroscopy with a thermal beam.

Judith Olson - CU Boulder/NIST
 Ramsey-Bordé Atom Interferometer

Physical Constants

```
In[306]:= c = 299792458.; (*meters per second*)
pi = 3.141; (*dimensionless*)
hbar = 1.054572*10.^(-34.); (*Joule seconds*)
kb = 1.3806488*10.^(-23.); (*Boltzman's Constant, in meters^2 Kilograms per (seconds^2 Kelvin)*)
```

System Variables

```
In[295]:= λ = 657.2777*10.^(-9.); (*Transition wavelength*)
mass = 6.6551*10.^(-26.); (*Calcium Mass in kilograms*)
linewidth = 400.; (*Hz*)
wavelength = 657.*10.^(-9.); (*meters*)
ω0 = 2.*pi*c/wavelength; (*resonant transition frequency in radians per second*)
satintensitylab = 1330.*400./ (657.^3.);
satintensity = satintensitylab;
tau = .003/610.;
intensitylab = .019/ (.003*.01);
k = 2.*pi/λ; (*magnitude of wave vector*)
δ = hbar*k^2./ (2.*mass); (*Recoil Frequency*)
```

Rabi Oscillations

The Math

```
In[310]:= detuning[ω_] := ω - ω0
ΩR[intensity_] := Sqrt[intensity/satintensitylab]*linewidth/Sqrt[2]
ΩG[intensity_, ω_] := Sqrt[(ΩR[intensity])^2 + (detuning[ω])^2]

excited[intensity_, ω_] := (ΩR[intensity]^2)/(ΩG[intensity, ω]^2)* (Sin[tau*ΩG[intensity, ω]])^2

FindRoot[excited[intensity, ω0] == 0.5, {intensity, 0.00001}]
(*The "ideal" intensity to create a π/2 pulse is 598.0292965492904 Watts per meter squared*)

Out[314]:= {intensity -> 598.029}
```

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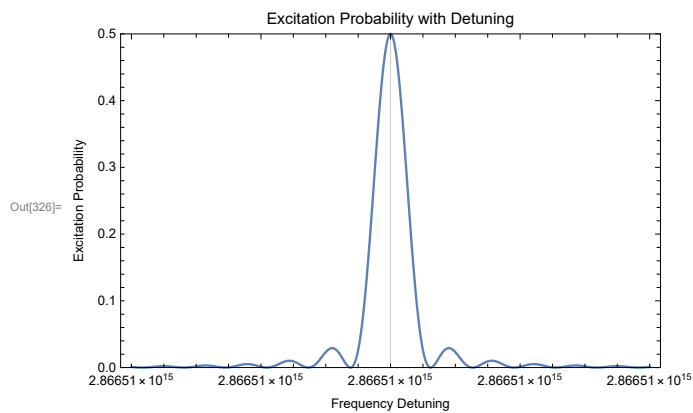
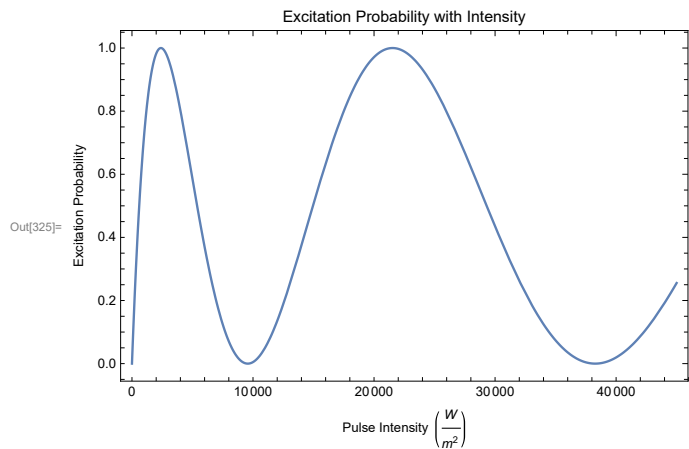
Rabi Oscillations Result

```

In[325]:= Plot[excited[intensity,  $\omega_0$ ], {intensity, 0, 45000}, PlotLabel → "Excitation Probability with Intensity",
  Frame → True, FrameLabel → {{ "Excitation Probability", None}, {"Pulse Intensity" Space[W / m^2], None}}]

(*{"Pulse Intensity", ""}, {"Excitation Probability", ""})*
Plot[excited[598.0292965492904,  $\omega$ ], { $\omega$ ,  $\omega_0 - 4 \times 10^6$ ,  $\omega_0 + 4 \times 10^6$ },
  PlotRange → {0, .5}, PlotLabel → "Excitation Probability with Detuning",
  Frame → True, FrameLabel → {{ "Excitation Probability", None}, {"Frequency Detuning", None}}, GridLines → {{ $\omega_0$ , None}}]

```



Rabi Oscillation Important Results

The ideal saturation intensity to achieve a $\pi/2$ pulse area is around 1900 Watts/m² when on resonance.

Characterizing the Experimental System

The atomic velocity distribution

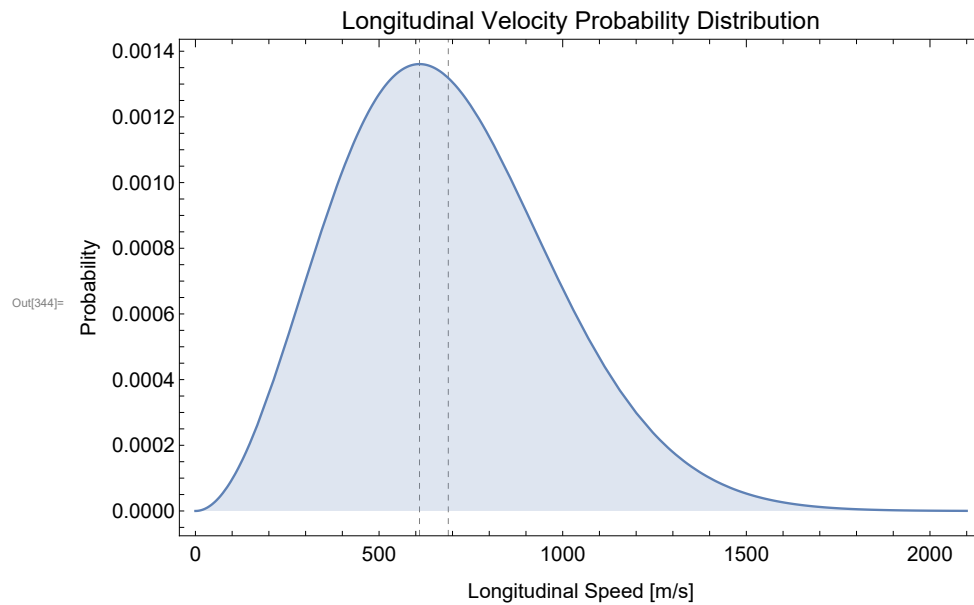
```
In[252]:= oventemp = 625.01 + 272.15; (*Oven temperature in degrees Kelvin,
determines the basic properties of the atomic velocity distribution*)
```

The Longitudinal Velocity Distribution:

```
In[340]:= (*Assuming a Longitudinal Maxwell-Boltzman Distribution*)
MBDist[v1_] := ProbabilityDistribution[
  Sqrt[(mass/(2*pi*kb*oventemp))^3]*4*pi*v1^2*Exp[-mass*v1^2/(2*kb*oventemp)], {v1, .001, 2000}]
Probv1[v1_] := Sqrt[(mass/(2*pi*kb*oventemp))^3]*4*pi*v1^2*Exp[-mass*v1^2/(2*kb*oventemp)]
v1mostprob = Sqrt[2*kb*oventemp/mass] (*most probably longitudinal velocity*)
v1avg = v1mostprob*2/Sqrt[3.141]
Plot[Probv1[v1], {v1, 0, 2100}, PlotLabel -> "Longitudinal Velocity Probability Distribution",
  Frame -> True, BaseStyle -> {FontSize -> 14}, Filling -> Axis, GridLines -> {{v1mostprob, v1avg}, {}},
  GridLinesStyle -> Directive[Gray, Dashed], FrameLabel -> {{{"Probability", None}, {"Longitudinal Speed [m/s]", None}}]
```

```
Out[342]= 610.119
```

```
Out[343]= 688.51
```



The Transverse Velocity Distribution:

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```

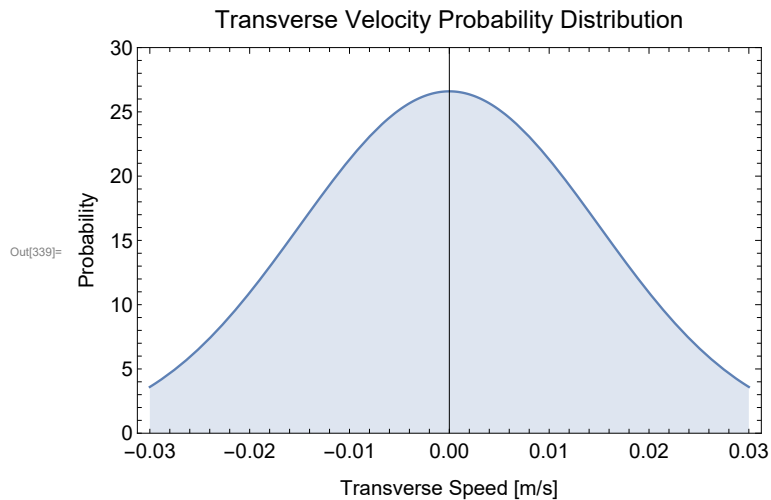
In[334]:= sigma = 1; (*Standard deviation*)
vzlim = 0.03;
Probtv[vz_] := PDF[NormalDistribution[0, .015], vz]
(*normconstant = Integrate[Probtv1[vz], {vz, -vzlim, +vzlim}]
Probtv[vz_] := Probtv1[vz] / normconstant*)
TransDist[vz_] := ProbabilityDistribution[Probtv[vz], {vz, -.03, .03}];
vzmostprob = 0;

Plot[Probtv[vz], {vz, -vzlim, vzlim}, PlotRange -> {0, 30},
PlotLabel -> "Transverse Velocity Probability Distribution", Frame -> True, BaseStyle -> {FontSize -> 14},
Filling -> Axis, GridLines -> {{vzmostprob, vzavg}, {}}, GridLinesStyle -> Directive[Gray, Dashed],
FrameLabel -> {{{"Probability", None}, {"Transverse Speed [m/s]", None}}]

(*a = .0005; (*microtubal diameter*)
L=0.005; (*microtubal length*)

Probtv[vz_] := (Abs[vz] * Exp[(-vz^2) / vzmostprob^2] * Gamma[-1/2, (vz^2 * L^2) / (vzmostprob^2 * a^2)]) /
Sqrt[8*Pi*vzmostprob^2 * Sqrt[1+a^2/L^2]]
Probtv[0] := 0.9783702;
Plot[Probtv[vz], {vz, -10, 10}, PlotRange -> Automatic
(*{0, 1}*)], PlotLabel -> "Transverse Velocity Probability Distribution",
Frame -> True, FrameLabel -> {{{"Probability", None}, {"Velocity [m/s^2]", None}}]
TransDist[vz_] := ProbabilityDistribution[Probtv[vz], {vz, -.01, .01}];
Probtv[vz_] := (Abs[vz] * Exp[-vz^2 / vzmostprob^2] * Gamma[-1/2, (vz^2 * L^2) / (vzmostprob^2 * a^2)]) /
Sqrt[8*Pi*vzmostprob^2 * Sqrt[1+a^2/L^2]]
vzmostprob =
0;*)

```



Velocity Dependent Variable Calculations

Special Relativistic Variables

```

γ[v1_] := 1/Sqrt[1 - v1^2/c^2]
γa = 0; (*level a's relaxation time, to be later incorporated*)
γb = 0; (*level b's relaxation time, to be later incorporated*)
γba = (γb + γa) / 2;

```

The Detuning Function

```

Δ[ω_, v1_] := γ[v1] * ω - ω0 (*The Detuning Calculation*)

```

The Various Rabi Frequencies - depend on the velocities AND some also depend on the individual beam intensities

```

Ωba = Sqrt[intensity/satintensity] * 400/Sqrt[2]; (*The Rabi frequency - Intensity dependent [[number]]*)

Ω0[vz_] := -δ - k*vz + I*γba; (*For beams 1 and 2, for all of the atoms*)
Ω0G[vz_] := -δ + k*vz + I*γba; (*For beams 3 and 4, with ground state atoms after 2*)
Ω0E[vz_] := -5*δ - 3*k*vz + I*γba; (*For beams 3 and 4, with excited state atoms after 2*)

Ω3[ω_, v1_, vz_] := Δ[ω, v1] - k*vz - δ + I*(γb - γa) / 2 (*For beams 1 and 2, where -kv_z is used*)
Ω3G[ω_, v1_, vz_] := Δ[ω, v1] + k*vz - δ + I*(γb - γa) / 2 (*For beams 3 and 4, with ground state atoms after 2*)
Ω3E[ω_, v1_, vz_] := Δ[ω, v1] + k*vz + 3*δ + I*(γb - γa) / 2
(*For beams 3 and 4, with excited state atoms after 2*)

Ω[ω_, v1_, vz_] := Sqrt[(Ω3[ω, v1, vz])^2 + 4*Ωba^2] (*Intensity dependent [[number]]*)
ΩG[ω_, v1_, vz_] := Sqrt[(Ω3G[ω, v1, vz])^2 + 4*Ωba^2]
ΩE[ω_, v1_, vz_] := Sqrt[(Ω3E[ω, v1, vz])^2 + 4*Ωba^2]

```

The red laser beam

```

Clear[power, beamarea, beamheight, beamwidth, intensity] (*Beam parameters will be listed as
{beam1, beam2, beam3, beam4} and can be individually referenced though [[location in vector]]*)
power = {17*10^(-3), 19*10^(-3), 19*10^(-3), 19*10^(-3)};
(*incident red power in Watts*)
beamheight = {.005, .007, .007, .007};
beamwidth = {.0015, .0015, .0015, .0015};
beamarea = beamwidth*beamheight; (*beam interaction area with the atoms in square meters*)
intensity = power/beamarea;

```

The Atom-Beam Interaction - For a Single Atom

```

phi = {0, 0, 0, 0};
T[v1_, vz_] := .07 / Abs[v1] (*Time between beams 1&2 and 3&4, independent of beam parameters*)
Tprime[v1_, vz_] := .005 / Abs[v1] (*Time between beams 2&3, has four values for the four beams [[number]]*)
ttau[v1_, vz_] := beamwidth / Abs[v1] (*Time in the four different beams [[number]]*)

(*PulseArea[number_, ω_, v1_, vz_] := Ω[ω, v1, vz] [[number]] * (ttau[v1, vz] [[number]]) / 2 (*[[number]]*)
PulseAreaG[number_, ω_, v1_, vz_] := ΩG[ω, v1, vz] [[number]] * (ttau[v1, vz] [[number]]) / 2 (*[[number]]*)
PulseAreaE[number_, ω_, v1_, vz_] := ΩE[ω, v1, vz] [[number]] * (ttau[v1, vz] [[number]]) / 2 (*[[number]]*)
*)
(*For forced pi/2 pulses:*)
PulseArea[number_, ω_, v1_, vz_] := pi / 4
PulseAreaG[number_, ω_, v1_, vz_] := pi / 4
PulseAreaE[number_, ω_, v1_, vz_] := pi / 4

origstate = {{0}, {1}};
(*There are four terms in each interaction zone matrix that determine
the interaction itself. These four terms are represented here by AA, BB, CC, DD*)
Clear[AA, BB, CC, DD, beam]
AA[number_, t_, ω_, v1_, vz_] :=
Cos[PulseArea[number, ω, v1, vz]] + I * Ω3[ω, v1, vz] / Ω[ω, v1, vz] [[number]] * Sin[PulseArea[number, ω, v1, vz]]
BB[number_, t_, ω_, v1_, vz_] := 2 * I * Ωba[[number]] / ΩG[ω, v1, vz] [[number]] * Sin[PulseArea[number, ω, v1, vz]]
CC[number_, t_, ω_, v1_, vz_] := BB[number, t, ω, v1, vz]
DD[number_, t_, ω_, v1_, vz_] :=
Cos[PulseArea[number, ω, v1, vz]] - I * Ω3[ω, v1, vz] / Ω[ω, v1, vz] [[number]] * Sin[PulseArea[number, ω, v1, vz]]

AG[number_, t_, ω_, v1_, vz_] :=
Cos[PulseAreaG[number, ω, v1, vz]] + I * Ω3G[ω, v1, vz] / ΩG[ω, v1, vz] [[number]] * Sin[PulseAreaG[number, ω, v1, vz]]
BG[number_, t_, ω_, v1_, vz_] := 2 * I * Ωba[[number]] / ΩG[ω, v1, vz] [[number]] * Sin[PulseAreaG[number, ω, v1, vz]]
CG[number_, t_, ω_, v1_, vz_] := BB[number, t, ω, v1, vz]
DG[number_, t_, ω_, v1_, vz_] :=
Cos[PulseAreaG[number, ω, v1, vz]] - I * Ω3G[ω, v1, vz] / ΩG[ω, v1, vz] [[number]] * Sin[PulseAreaG[number, ω, v1, vz]]

AE[number_, t_, ω_, v1_, vz_] :=
Cos[PulseAreaE[number, ω, v1, vz]] + I * Ω3E[ω, v1, vz] / ΩE[ω, v1, vz] [[number]] * Sin[PulseAreaE[number, ω, v1, vz]]
BE[number_, t_, ω_, v1_, vz_] := 2 * I * Ωba[[number]] / ΩE[ω, v1, vz] [[number]] * Sin[PulseAreaE[number, ω, v1, vz]]
CE[number_, t_, ω_, v1_, vz_] := BB[number, t, ω, v1, vz]
DE[number_, t_, ω_, v1_, vz_] :=
Cos[PulseAreaE[number, ω, v1, vz]] - I * Ω3E[ω, v1, vz] / ΩE[ω, v1, vz] [[number]] * Sin[PulseAreaE[number, ω, v1, vz]]

(*number indicates the beam number to which is being referenced in the calculation*)
beam[number_, t_, ω_, v1_, vz_] := {{AA[number, t, ω, v1, vz], BB[number, t, ω, v1, vz] * Exp[-I * phi[[number]]]},
{CC[number, t, ω, v1, vz] * Exp[I * phi[[number]]], DD[number, t, ω, v1, vz]}};
beamG[number_, t_, ω_, v1_, vz_] := {{AG[number, t, ω, v1, vz], BG[number, t, ω, v1, vz] * Exp[-I * phi[[number]]]},
{CG[number, t, ω, v1, vz] * Exp[I * phi[[number]]], DG[number, t, ω, v1, vz]}};
beamE[number_, t_, ω_, v1_, vz_] := {{AE[number, t, ω, v1, vz], BE[number, t, ω, v1, vz] * Exp[-I * phi[[number]]]},
{CE[number, t, ω, v1, vz] * Exp[I * phi[[number]]], DE[number, t, ω, v1, vz]}};

```

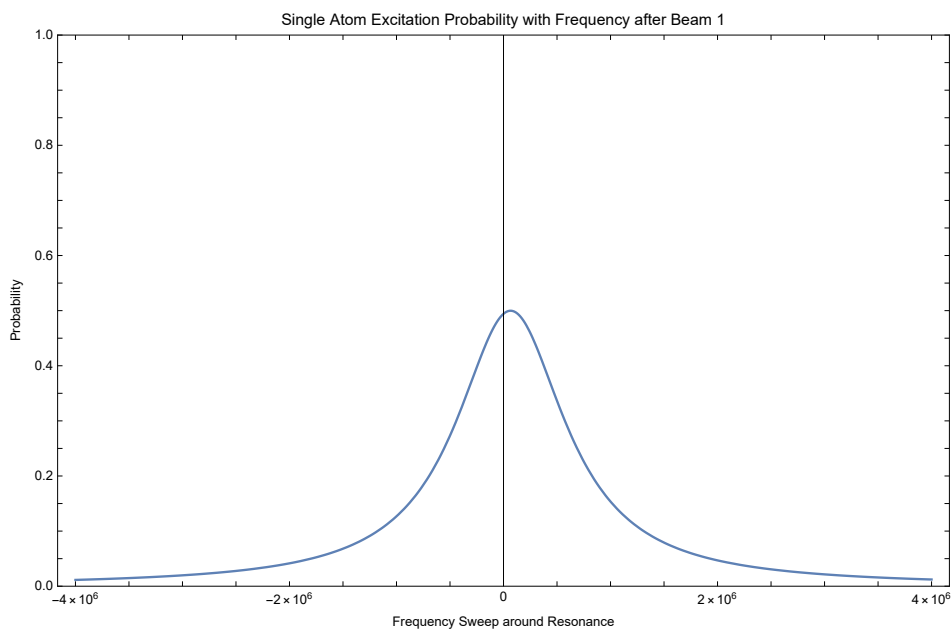
Beam 1

```

(*with t=0 being the moment the atoms enter the beam 1 interaction zone*)
Clear[t]
dark1[ω_, v1_, vz_] := {{Exp[I * Ω3[ω, v1, vz] * (T[v1, vz]) / 2], 0}, {0, Exp[-I * Ω3[ω, v1, vz] * (T[v1, vz]) / 2]}}
phase1[t_, v1_, vz_] := Exp[I / 2 * Ω0[vz] * (T[v1, vz] + tttau[v1, vz] [[1]] + t)]
Stateafter1[t_, ω_, v1_, vz_] := phase1[t, v1, vz] * dark1[ω, v1, vz].beam[1, t, ω, v1, vz].origstate
Excitedafter1[t_, ω_, v1_, vz_] := Stateafter1[t, ω, v1, vz] [[1]]
Signalafter1[t_, ω_, v1_, vz_] := Excitedafter1[t, ω, v1, vz] * Excitedafter1[t, ω, v1, vz]*

```

```
Plot[Evaluate[Signalafter1[1,  $\omega + \omega_0$ , vlmostprob, 0]], { $\omega$ ,  $-4 \times 10^6$ ,  $4 \times 10^6$ }, PlotRange -> {0, 1},
PlotPoints -> 10, PlotLabel -> "Single Atom Excitation Probability with Frequency after Beam 1",
Frame -> True, FrameLabel -> {{{"Probability", None}}, {"Frequency Sweep around Resonance", None}},
GridLines -> {{ $\omega_0$ }, None} (*Puts vertical line on resonance*)]
```



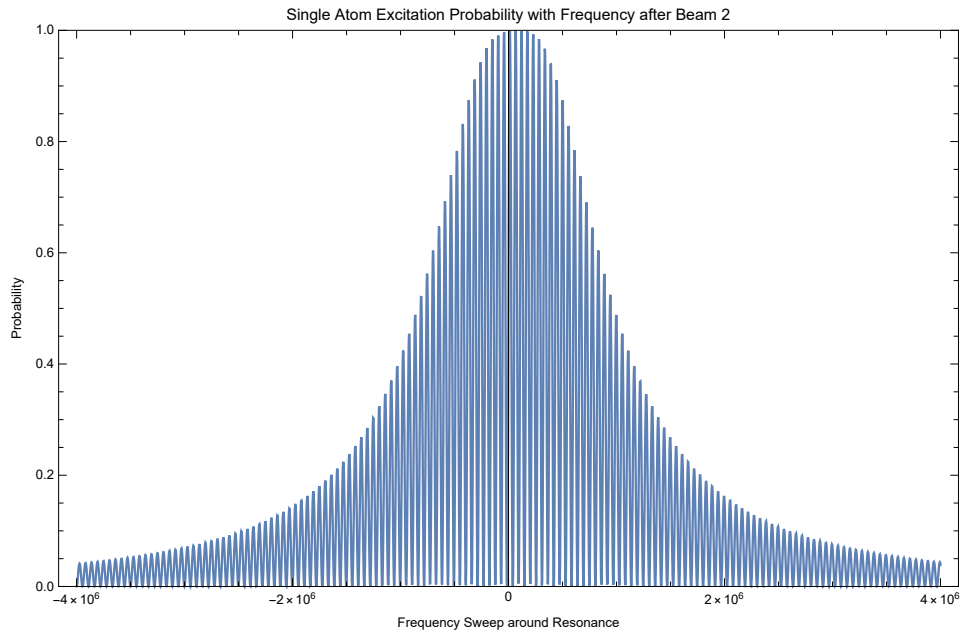
Beam 2

```
dark2[ $\omega$ _, v1_, vz_] :=
  {{Exp[I *  $\Omega$ 3[ $\omega$ , v1, vz] * (Tprime[v1, vz]) / 2], 0}, {0, Exp[-I *  $\Omega$ 3[ $\omega$ , v1, vz] * (Tprime[v1, vz]) / 2]}}
phase2[t_,  $\omega$ _, v1_, vz_] := Exp[I / 2 *  $\Omega$ 0[vz] * (Tprime[v1, vz] + ttau[v1, vz] [[2]])]
Stateafter2[t_,  $\omega$ _, v1_, vz_] := phase2[t, v1, vz] * dark2[ $\omega$ , v1, vz] . beam[2, t,  $\omega$ , v1, vz] . Stateafter1[t,  $\omega$ , v1, vz]
Excitedafter2[t_,  $\omega$ _, v1_, vz_] := Stateafter2[t,  $\omega$ , v1, vz] [[1]]
Signalafter2[t_,  $\omega$ _, v1_, vz_] := Excitedafter2[t,  $\omega$ , v1, vz] * Excitedafter2[t,  $\omega$ , v1, vz]*

Stateafter2G[t_,  $\omega$ _, v1_, vz_] := {{0}, Stateafter2[t,  $\omega$ , v1, vz] [[2]]}
Stateafter2E[t_,  $\omega$ _, v1_, vz_] := {Stateafter2[t,  $\omega$ , v1, vz] [[1]], {0}}
```



```
Plot[Evaluate[Signalafter2[1,  $\omega + \omega_0$ , v1mostprob, v2mostprob]], { $\omega$ ,  $-4 \times 10^6$ ,  $4 \times 10^6$ }, PlotRange -> {0, 1},
PlotPoints -> 50, PlotLabel -> "Single Atom Excitation Probability with Frequency after Beam 2",
Frame -> True, FrameLabel -> {{{"Probability", None}}, {"Frequency Sweep around Resonance", None}},
GridLines -> {{ $\omega_0$ }, None} (*Puts vertical line on resonance*)]
```



Beam 3 - Now have two different atomic groups for the two different interferometer paths

(*The higher frequency fringe comes from the atoms in the excited state between beams 2&3, while the lower frequency fringe comes from the atoms in the ground state between 2& 3. These will be described by G and E suffixes in the variables*)

```
dark3G[ $\omega_{-}$ , v1_, vz_] := {{Exp[I* $\Omega$ 3G[ $\omega$ , v1, vz] * (T[v1, vz]) / 2], 0}, {0, Exp[-I* $\Omega$ 3G[ $\omega$ , v1, vz] * (T[v1, vz]) / 2]}}
phase3G[t_, v1_, vz_] := Exp[I/2* $\Omega$ 0G[vz] * (T[v1, vz] + ttau[v1, vz][3])]]

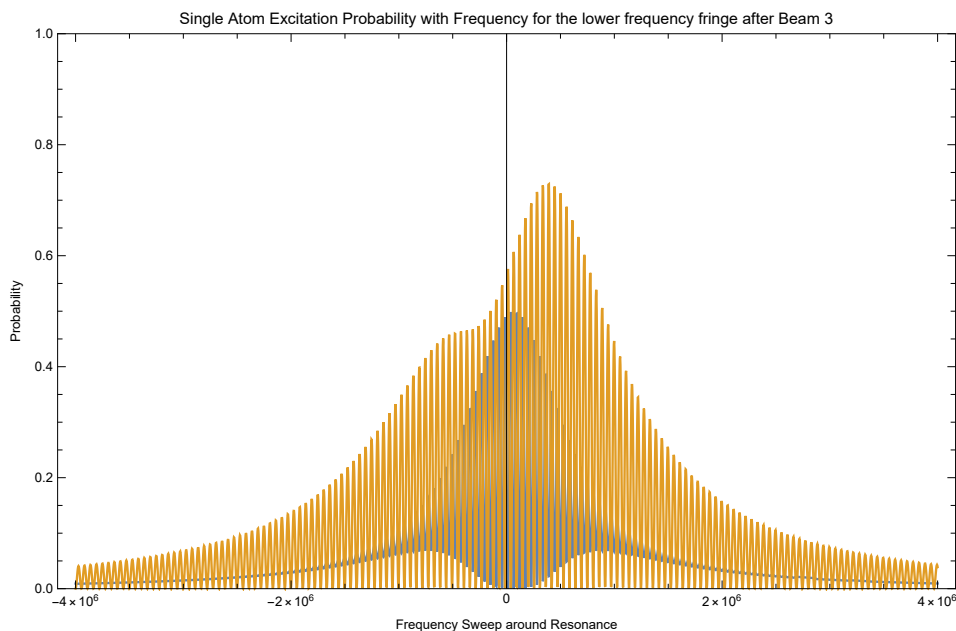
dark3E[ $\omega_{-}$ , v1_, vz_] := {{Exp[I* $\Omega$ 3E[ $\omega$ , v1, vz] * (T[v1, vz]) / 2], 0}, {0, Exp[-I* $\Omega$ 3E[ $\omega$ , v1, vz] * (T[v1, vz]) / 2]}}
phase3E[t_, v1_, vz_] := Exp[I/2* $\Omega$ 0E[vz] * (T[v1, vz] + ttau[v1, vz][3])]]

Stateafter3G[t_,  $\omega_{-}$ , v1_, vz_] :=
  phase3G[t, v1, vz] * dark3G[ $\omega$ , v1, vz].beamG[3, t,  $\omega$ , v1, vz].Stateafter2G[t,  $\omega$ , v1, vz]
Stateafter3E[t_,  $\omega_{-}$ , v1_, vz_] :=
  phase3E[t, v1, vz] * dark3E[ $\omega$ , v1, vz].beamE[3, t,  $\omega$ , v1, vz].Stateafter2E[t,  $\omega$ , v1, vz]

Excitedafter3G[t_,  $\omega_{-}$ , v1_, vz_] := Stateafter3G[t,  $\omega$ , v1, vz][[1]]
Excitedafter3E[t_,  $\omega_{-}$ , v1_, vz_] := Stateafter3E[t,  $\omega$ , v1, vz][[1]]

Signalafter3G[t_,  $\omega_{-}$ , v1_, vz_] := Excitedafter3G[t,  $\omega$ , v1, vz] * Excitedafter3G[t,  $\omega$ , v1, vz]*
Signalafter3E[t_,  $\omega_{-}$ , v1_, vz_] := Excitedafter3E[t,  $\omega$ , v1, vz] * Excitedafter3E[t,  $\omega$ , v1, vz]*
```

```
Plot[Evaluate[{Signalafter3G[1,  $\omega + \omega_0$ , vlmostprob, vzmostprob], Signalafter3E[1,  $\omega + \omega_0$ , vlmostprob, vzmostprob]}],
{ $\omega$ ,  $-4 \times 10^6$ ,  $4 \times 10^6$ }, PlotRange  $\rightarrow$  {0, 1}, PlotPoints  $\rightarrow$  50,
PlotLabel  $\rightarrow$  "Single Atom Excitation Probability with Frequency for the lower frequency fringe after Beam 3",
Frame  $\rightarrow$  True, FrameLabel  $\rightarrow$  {"Probability", None}, {"Frequency Sweep around Resonance", None}},
GridLines  $\rightarrow$  {{ $\omega_0$ }, None} (*Puts vertical line on resonance*)]
```



```
(*Plot[Signalafter3E[1,  $\omega$ , 610, vzmostprob], { $\omega$ ,  $\omega_0 - \omega_0/100000000$ ,  $\omega_0 + \omega_0/100000000$ }, PlotRange  $\rightarrow$  {0, 1}, PlotPoints  $\rightarrow$  10,
PlotLabel  $\rightarrow$  "Single Atom Excitation Probability with Frequency for the higher frequency after Beam 3",
Frame  $\rightarrow$  True, FrameLabel  $\rightarrow$  {"Probability", None}, {"Frequency Sweep around Resonance", None}},
GridLines  $\rightarrow$  {{ $\omega_0$ }, None} (*Puts vertical line on resonance*)] *)
```

Beam 4

```
phase4G[t_, vl_, vz_] := Exp[I/2 *  $\Omega_0$ G[vz] * (ttau[vl, vz] [[4]])]
phase4E[t_, vl_, vz_] := Exp[I/2 *  $\Omega_0$ E[vz] * (ttau[vl, vz] [[4]])]

Stateafter4G[t_,  $\omega$ _, vl_, vz_] := phase4G[t, vl, vz] * beamG[4, t,  $\omega$ , vl, vz].Stateafter3G[t,  $\omega$ , vl, vz]
Stateafter4E[t_,  $\omega$ _, vl_, vz_] := phase4E[t, vl, vz] * beamE[4, t,  $\omega$ , vl, vz].Stateafter3E[t,  $\omega$ , vl, vz]

Excitedafter4G[t_,  $\omega$ _, vl_, vz_] := Stateafter4G[t,  $\omega$ , vl, vz] [[1]]
Excitedafter4E[t_,  $\omega$ _, vl_, vz_] := Stateafter4E[t,  $\omega$ , vl, vz] [[1]]

Signalafter4G[t_,  $\omega$ _, vl_, vz_] := Re[Excitedafter4G[t,  $\omega$ , vl, vz] * Excitedafter4G[t,  $\omega$ , vl, vz]*]
Signalafter4E[t_,  $\omega$ _, vl_, vz_] := Re[Excitedafter4E[t,  $\omega$ , vl, vz] * Excitedafter4E[t,  $\omega$ , vl, vz]*]

Signalafter4G[2,  $\omega_0$ , 610, 0]
```

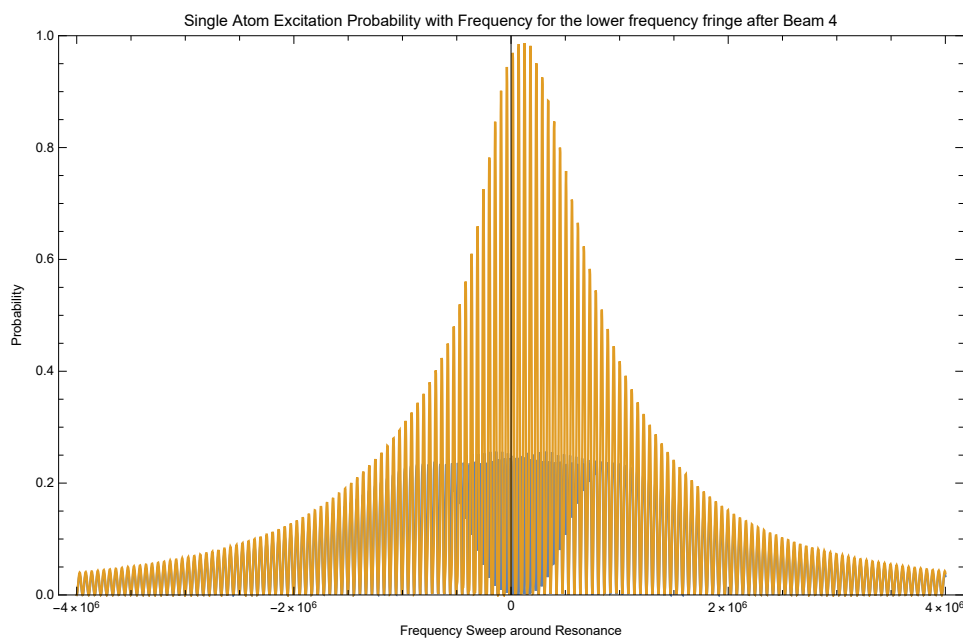
```
{0.246916}
```

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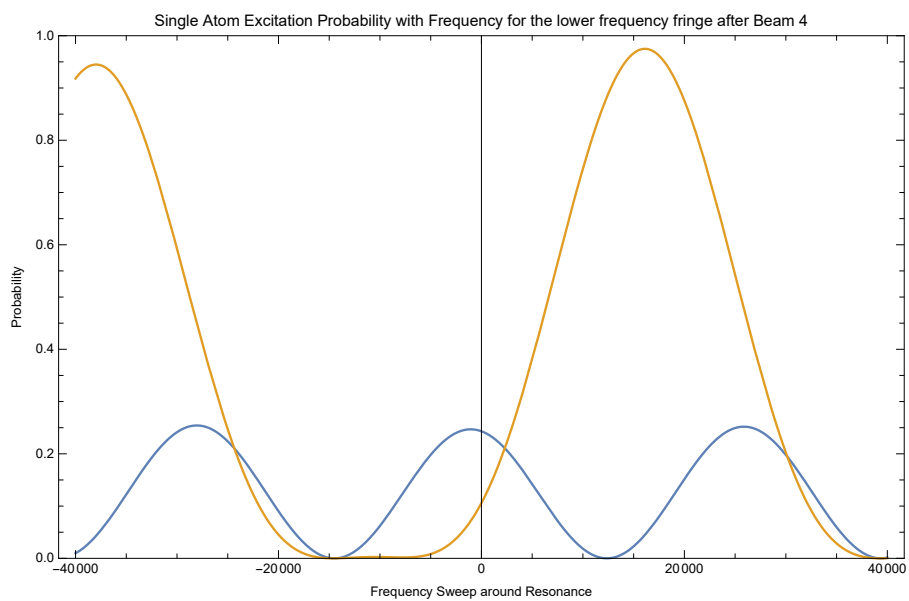
```

Plot[Evaluate[{Signalafter4G[1,  $\omega + \omega_0$ , vImostprob, vzmostprob], Signalafter4E[1,  $\omega + \omega_0$ , vImostprob, vzmostprob]}],
{ $\omega$ ,  $-4 \times 10^6$ ,  $4 \times 10^6$ }, PlotRange  $\rightarrow$  {0, 1}, PlotPoints  $\rightarrow$  50,
PlotLabel  $\rightarrow$  "Single Atom Excitation Probability with Frequency for the lower frequency fringe after Beam 4",
Frame  $\rightarrow$  True, FrameLabel  $\rightarrow$  {"Probability", None}, {"Frequency Sweep around Resonance", None}},
GridLines  $\rightarrow$  {{ $\omega_0$ }, None} (*Puts vertical line on resonance*)]

```

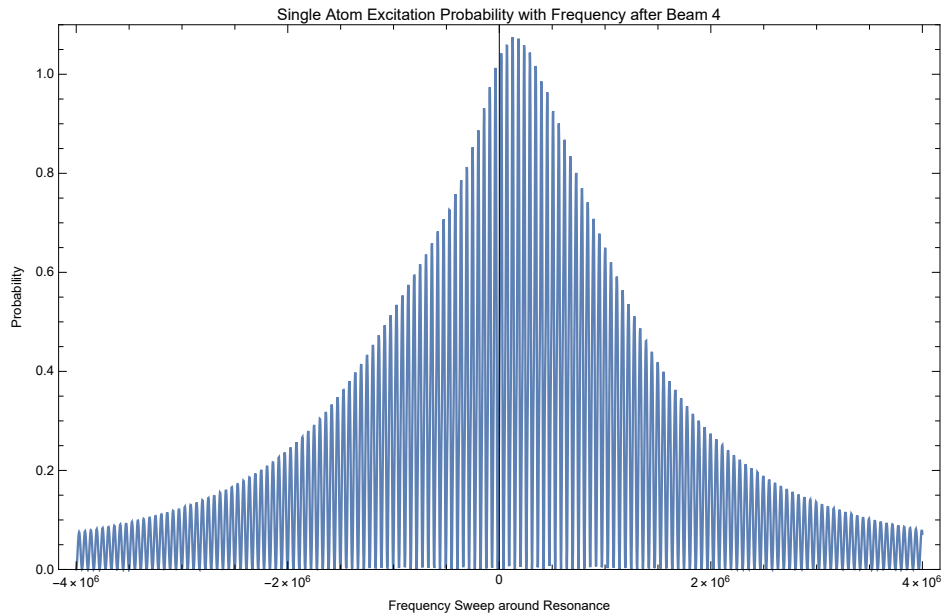


```
Plot[Evaluate[{Signalafter4G[1,  $\omega + \omega_0$ , vImostprob, vZmostprob], Signalafter4E[1,  $\omega + \omega_0$ , vImostprob, vZmostprob]}],
{ $\omega$ , -40000, 40000}, PlotRange -> {0, 1}, PlotPoints -> 50,
PlotLabel -> "Single Atom Excitation Probability with Frequency for the lower frequency fringe after Beam 4",
Frame -> True, FrameLabel -> {"Probability", None}, {"Frequency Sweep around Resonance", None}},
GridLines -> {{ $\omega_0$ }, None} (*Puts vertical line on resonance*)]
```

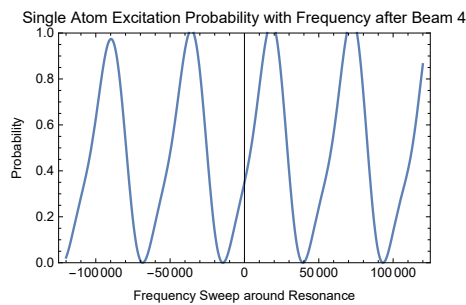


The Final Result:

```
FinalExcitedPopulation[t_,  $\omega$ _, v1_, vz_] := (Signalafter4G[t,  $\omega$ , v1, vz] + Signalafter4E[t,  $\omega$ , v1, vz])
Plot[Evaluate[FinalExcitedPopulation[1,  $\omega$  +  $\omega_0$ , v1mostprob, vzmostprob]], { $\omega$ ,  $-4 \times 10^6$ ,  $4 \times 10^6$ }, PlotRange -> {0, 1.1},
PlotPoints -> 50, PlotLabel -> "Single Atom Excitation Probability with Frequency after Beam 4",
Frame -> True, FrameLabel -> {{{"Probability", None}}, {"Frequency Sweep around Resonance", None}},
GridLines -> {{ $\omega_0$ }, None} (*Puts vertical line on resonance*)]
```

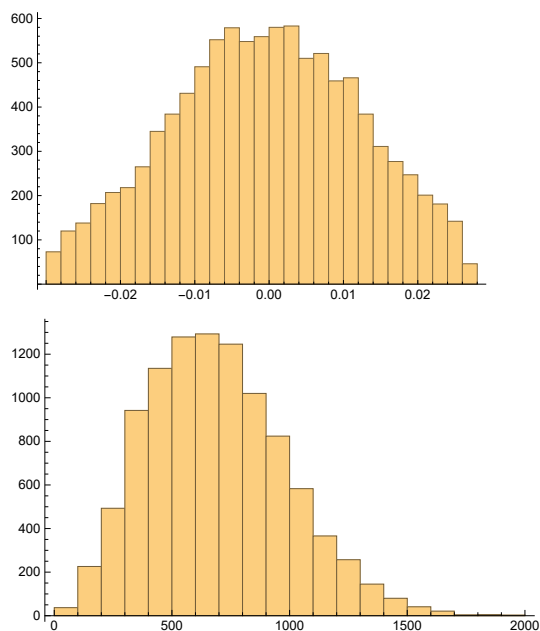


```
Plot[Evaluate[FinalExcitedPopulation[1,  $\omega$  +  $\omega_0$ , v1mostprob, vzmostprob]], { $\omega$ , -120000, 120000}, PlotRange -> {0, 1},
PlotPoints -> 50, PlotLabel -> "Single Atom Excitation Probability with Frequency after Beam 4",
Frame -> True, FrameLabel -> {{{"Probability", None}}, {"Frequency Sweep around Resonance", None}},
GridLines -> {{ $\omega_0$ }, None} (*Puts vertical line on resonance*)]
```



Including the Velocity Distributions - The Many Atom Case

```
num = 10000; (*number of atoms to include*)
vldata = RandomVariate[MBDist[v1], num];
vzdata = RandomVariate[TransDist[vz], num];
Histogram[vzdata]
Histogram[vldata]
```



```
Clear[FinalG, FinalE]
FinalExcitedPopulationSum[ $\omega$ _] := Sum[FinalExcitedPopulation[1,  $\omega$ , vldata[[n]], vzdata[[n]], {n, 1, num}]/num
(*FinalExcitedPopulationSum[ $\omega$ _] := Sum[FinalExcitedPopulation[1,  $\omega$ , 610, vzdata[[n]], {n, 1, num}]/num*)
FinalExcitedPopulationSum[ $\omega$ 0]
FinalG[ $\omega$ _] := Sum[SignalAfter4G[1,  $\omega$ , vldata[[n]], vzdata[[n]], {n, 1, num}]/num
FinalE[ $\omega$ _] := Sum[SignalAfter4E[1,  $\omega$ , vldata[[n]], vzdata[[n]], {n, 1, num}]/num
FinalG[ $\omega$ 0]
FinalE[ $\omega$ 0]
{0.508621}

{0.250486}

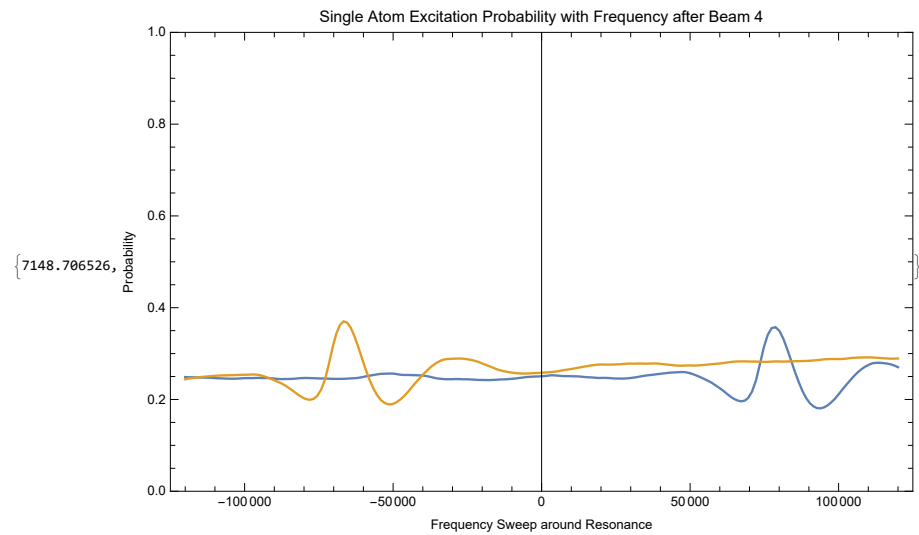
{0.258136}
```

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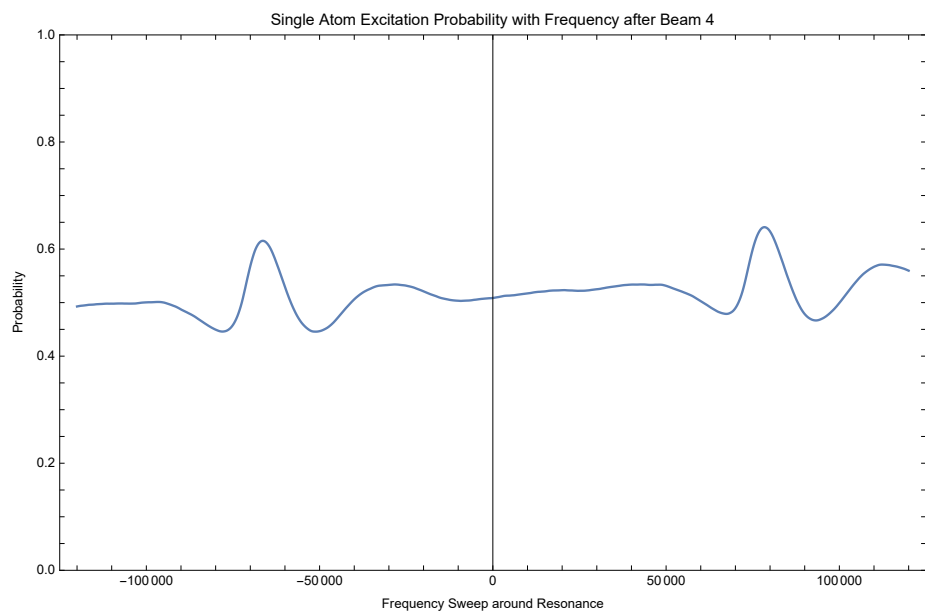
```

Plot[{FinalG[ $\omega + \omega_0$ ], FinalE[ $\omega + \omega_0$ ]}, { $\omega$ , -120000, 120000}, PlotRange -> {0, 1},
PlotPoints -> Automatic, PlotLabel -> "Single Atom Excitation Probability with Frequency after Beam 4",
Frame -> True, FrameLabel -> {{{"Probability", None}}, {"Frequency Sweep around Resonance", None}},
GridLines -> {{ $\omega_0$ }, None} (*Puts vertical line on resonance*), PerformanceGoal -> "Speed"] // AbsoluteTiming

```



```
Plot[Evaluate[FinalExcitedPopulationSum[ $\omega + \omega_0$ ]], { $\omega$ , -120000, 120000}, PlotRange -> {0, 1},
PlotPoints -> 100, PlotLabel -> "Single Atom Excitation Probability with Frequency after Beam 4",
Frame -> True, FrameLabel -> {{{"Probability", None}, {"Frequency Sweep around Resonance", None}},
GridLines -> {{ $\omega_0$ }, None} (*Puts vertical line on resonance*), PerformanceGoal -> "Speed"] // AbsoluteTiming
{301.569154,
```

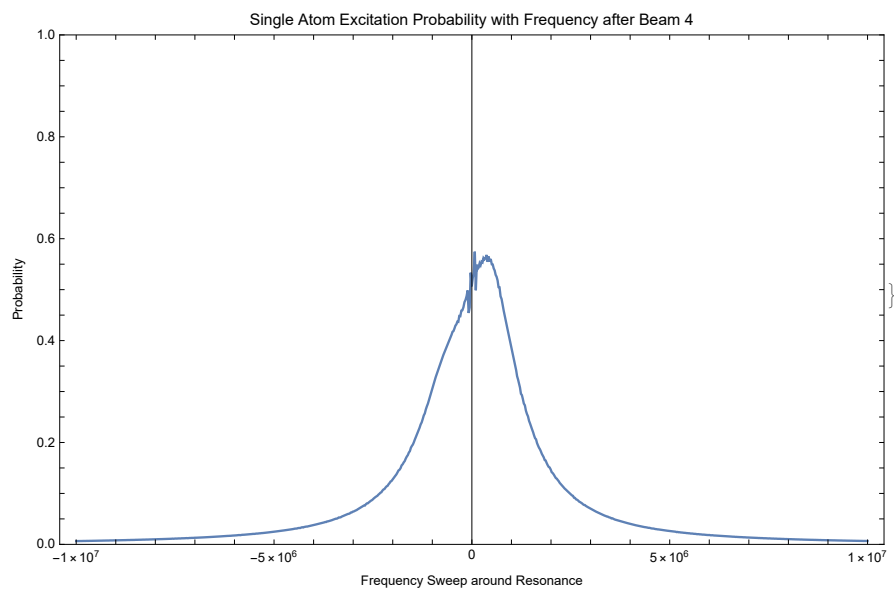


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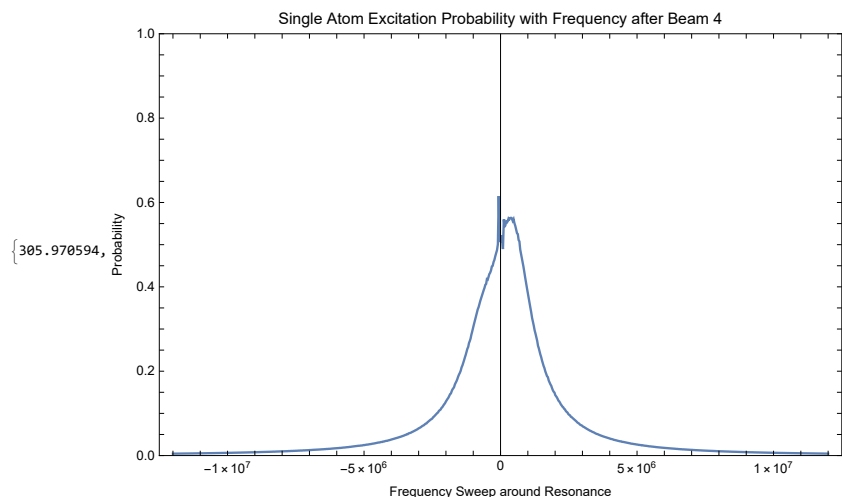
```

Plot[Evaluate[FinalExcitedPopulationSum[ $\omega + \omega_0$ ]], { $\omega$ , -10000000, 10000000}, PlotRange -> {0, 1},
PlotPoints -> 100, PlotLabel -> "Single Atom Excitation Probability with Frequency after Beam 4",
Frame -> True, FrameLabel -> {{{"Probability", None}, {"Frequency Sweep around Resonance", None}},
GridLines -> {{ $\omega_0$ }, None} (*Puts vertical line on resonance*), PerformanceGoal -> "Speed"] // AbsoluteTiming
{300.909088,

```



```
ClearSystemCache[];
AbsoluteTiming[Plot[Evaluate[FinalExcitedPopulationSum[ $\omega + \omega_0$ ]], { $\omega$ , -12000000, 12000000}, PlotRange -> {0, 1},
  PlotPoints -> 100, PlotLabel -> "Single Atom Excitation Probability with Frequency after Beam 4",
  Frame -> True, FrameLabel -> {{{"Probability", None}, {"Frequency Sweep around Resonance", None}},
  GridLines -> {{ $\omega_0$ }, None}, PerformanceGoal -> "Speed"]]
```



```
(*Clear[FinalG,FinalE]
FinalExcitedPopulationSum[ $\omega$ _.]:=Sum[FinalExcitedPopulation[1, $\omega$ ,vldata[[n]],vzdata[[n]],{n,1,num}]/num
FinalG[ $\omega$ _.]:=Sum[Signalafter4G[1, $\omega$ ,vldata[[n]],vzdata[[n]],{n,1,num}]/num
FinalE[ $\omega$ _.]:=Sum[Signalafter4E[1, $\omega$ ,vldata[[n]],vzdata[[n]],{n,1,num}]/num*)

(*Clear[plotpoints]
plotpoints = Table[{ $\omega - \omega_0$ , FinalExcitedPopulationSum[ $\omega$ ][[1]]}, { $\omega$ ,  $\omega_0 - 240000$ ,  $\omega_0 + 240000$ , 100}]; *)

(*plotpoints;
ListLinePlot[plotpoints]*)

(*ListLinePlot[plotpoints]*)

(*Plot[{FinalG[ $\omega + \omega_0$ ], FinalE[ $\omega + \omega_0$ ]}, { $\omega$ , -240000, 240000}, PlotRange -> {0, 1}, PlotPoints -> 200, PlotLabel -> "Single Atom Excitation Probability with
Frequency after Beam 4", Frame -> True, FrameLabel -> {{{"Probability", None}, {"Frequency Sweep around Resonance", None}}, GridLines ->
{{ $\omega_0$ }, None} (*Puts vertical line on resonance*)]*)

(*Plot[FinalExcitedPopulationSum[ $\omega + \omega_0$ ], { $\omega$ , -4*10^6, 4*10^6}, PlotRange -> {0, 1}, PlotPoints -> 10, PlotLabel -> "Single Atom Excitation Probability with
Frequency after Beam 4", Frame -> True, FrameLabel -> {{{"Probability", None}, {"Frequency Sweep around Resonance", None}}, GridLines ->
{{ $\omega_0$ }, None} (*Puts vertical line on resonance*)*)
```