DEVELOPING A FREQUENCY CALIBRATION MODEL FOR TELLURIUM SPECTRA

By

Joshua Adam Salazar

A capstone project submitted for
Graduation with University Honors

June 9, 2017

University Honors
University of California, Riverside

APPROVED

_____
Dr. Harry W. K. Tom
Department of Physics and Astronomy

_____
Dr. Richard Cardullo, Howard H Hays Chair and Faculty Director, University Honors
Associate Vice Provost, Undergraduate Education

Abstract

The tellurium spectrometer, an optical setup exploiting saturation absorption spectroscopy (SAS) to measure Doppler-free spectra of $^{130}Te_2$ gas, is a preferred frequency reference in the exercise of laser spectroscopy. However, the particle-antiparticle research community has been seeking a resolution to the proton charge radius puzzle of quantum electrodynamic theory (QED). The community has thus been pushing for a positronium frequency 1^3S_1 - 2^3S_1 measurement precision greater than the MHz precision limit partly due to the limited reproducibility of tellurium spectra frequencies. This limited reproducibility is mostly attributed to design and temperature sensor discrepancies among manufactured tellurium ovens. A preliminary experiment was carried out to make headway towards resolving this tellurium spectra reproducibility issue. Using an Ultra-Low-Expansion (ULE) cavity as a frequency reference, frequency differences between individual 486 nm tellurium spectrum lines were measured for a set tellurium oven temperature to create the procedure groundwork of a frequency calibration model. Though the results of this practice experiment did not lead to the completion of this model nor demonstrated the model's effectiveness in reducing the MHz calibration uncertainty, the procedure executed in this experiment is deemed to be appropriate for the partial construction of the frequency calibration model.

Acknowledgements

I will be forever grateful to the University Honors program of University of California Riverside. The guidance University Honors provided me throughout the development of my project and the generous kindness they offered me after the recent passing of my mother, Desiree Salazar, was essential to the completion of this project. Being part of the University Honors program has surely gifted me a unique undergraduate education experience and skills that I will carry with me as I pursue my future professional career.

Thank you to the National Science Foundation for supporting my efforts to the learn research skills necessary for completing this project.

Thank you to my parents Gustavo and Desiree Salazar and my grandparents Richard and Lynn Jones. Their unconditional love, support and understanding of the many necessary hours that were spent in lab is what pressed me to persevere and complete my capstone project.

I cannot express enough thanks to my mentors Dr. Harry W. K. Tom, Harris Goldman and Yuanqian Liu. The completion of this project would not have been possible without their countless hours of aid and support. I sincerely appreciate all of the irreplaceable learning experiences that the Tom lab provided me. My time working with my mentors, my lab mates and friends has added to my education experience and has created memories that I will continue to cherish in the years to come.

Table of Contents

List of Tables and Figures

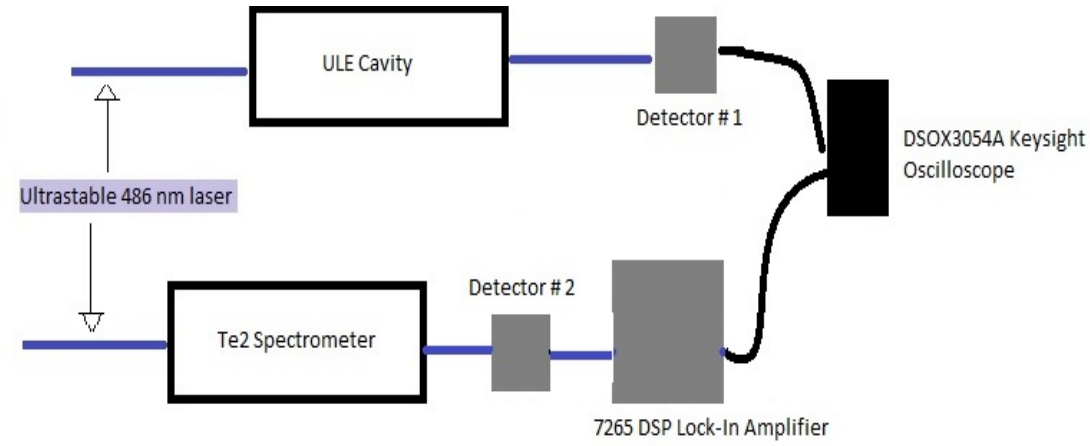

Figure 1: This schematic depicts the basic component setup of the experiment. Both the ULE cavity and the tellurium (Te_2) spectrometer received a portion of the laser light varying about the 486 nm wavelength. The ULE resonant peaks detected at Detector # 1 and the tellurium spectra detected at Detector # 2 were simultaneously measured through the Keysight oscilloscope.

Figure 2 – Tellurium Spectrometer Internal Components

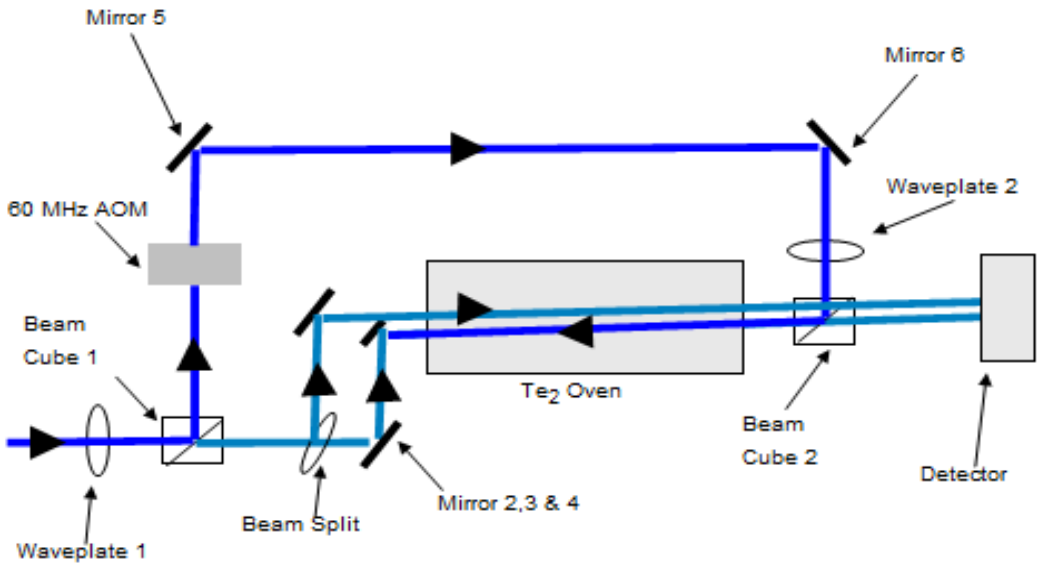

Figure 2: A simplified illustration of the optical setup making up the tellurium spectrometer system. The light blue lines represent the signal and reference probe beams, both of which were set to have a power of approximately 80 µW by using waveplate and polarization beam cube optics. The dark blue lines represent the pump beam which were, through similar methods, set to have a power of approximately 9 mW. The tellurium oven shown here is a containment and heating unit designed to isolate diatomic tellurium gas ($^{130}Te_2$) within a glass cell at a stable temperature (pressure) while allowing laser beams to probe the gas. A PID Omega temperature controller, two Variac variable transformers, and three solid-state relays were used to maintain the oven at a stable top stem temperature of approximately 535.00 ºC. The corresponding cold-point (base stem) temperature of the oven was measured to be 454.34 ± 0.64 ºC. To improve the heat insulation of the oven, its exterior was wrapped in fiber glass insulation. A majority of optics were mounted on inch diameter pedestal posts made of stainless steel.

Figure 3 – Visual Data Sample

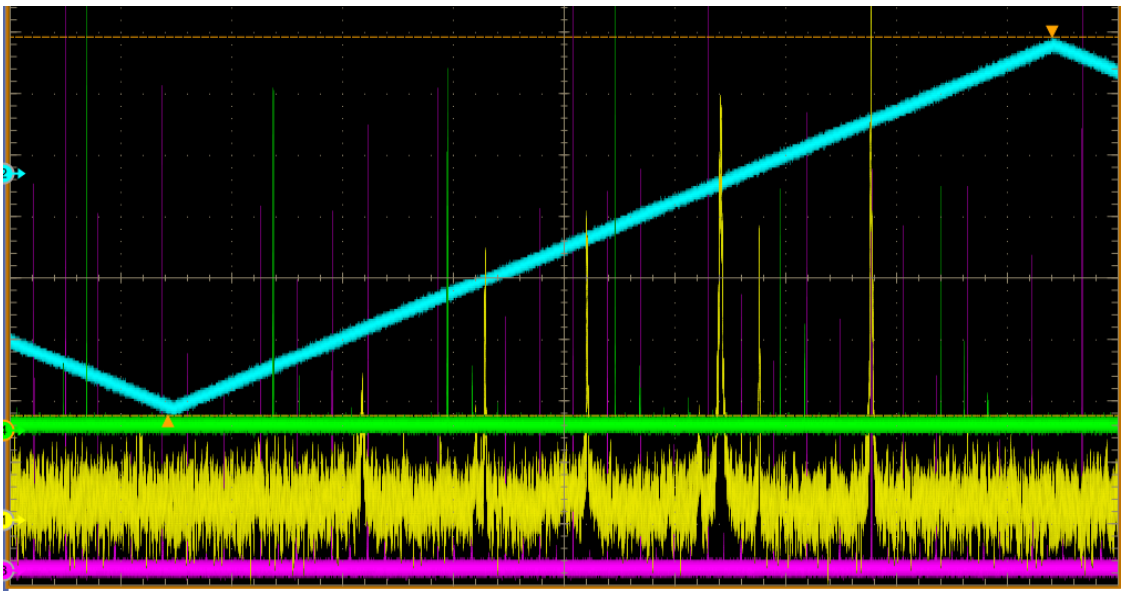

Figure 3: An oscilloscope screenshot of signals collected during this experiment. Though these signals in particular were not analyzed, they visually exemplify the actual signals taken for analysis. The ULE peaks (green), tellurium spectra (yellow), and the ramp signal (blue) displayed here all have the same shape as the analyzed signals. The resonance peaks in pink are from a lab made invar interferometer. The invar peaks were not analyzed during this preliminary experiment.

Figure 4 – Lorentzian Fits for Tellurium Spectra

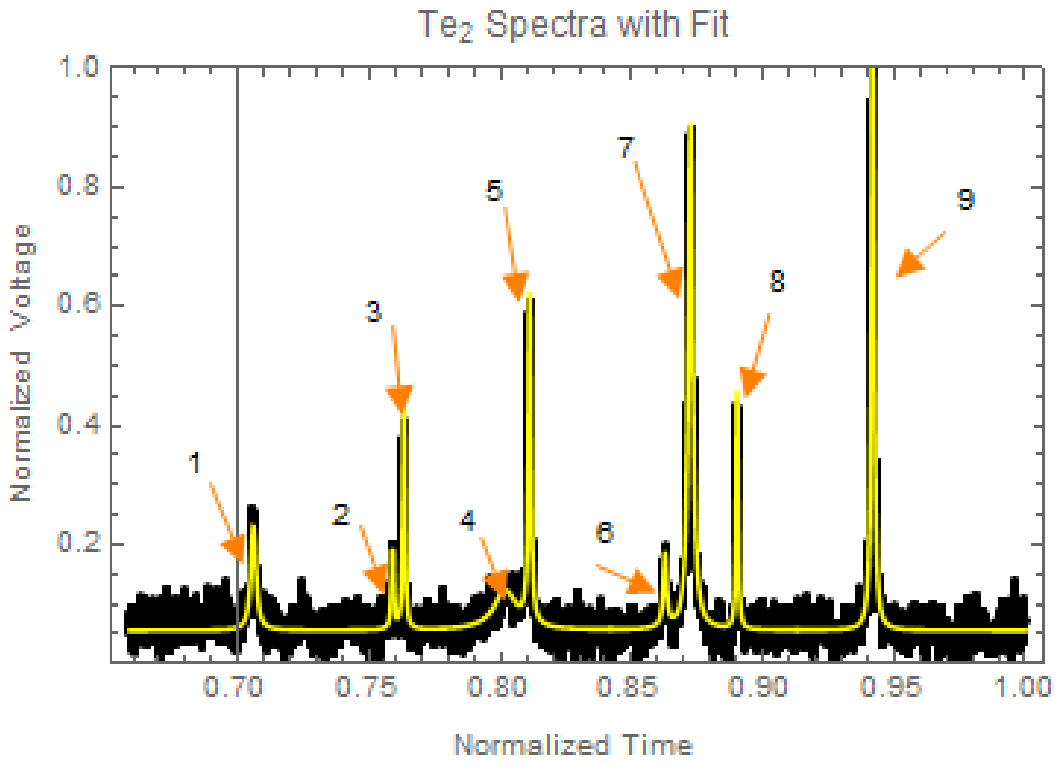

Figure 4: This displays the calculated Lorentzian fit (yellow) for the nine tellurium spectra sampled (black points) in the vicinity of the 486 nm absolute wavelength. Each of the peaks displayed here are labeled with an integer number 1 through 9 so that each peak remained identifiable throughout analysis.

Figure 5 – Airy Distribution Fits for ULE Peaks

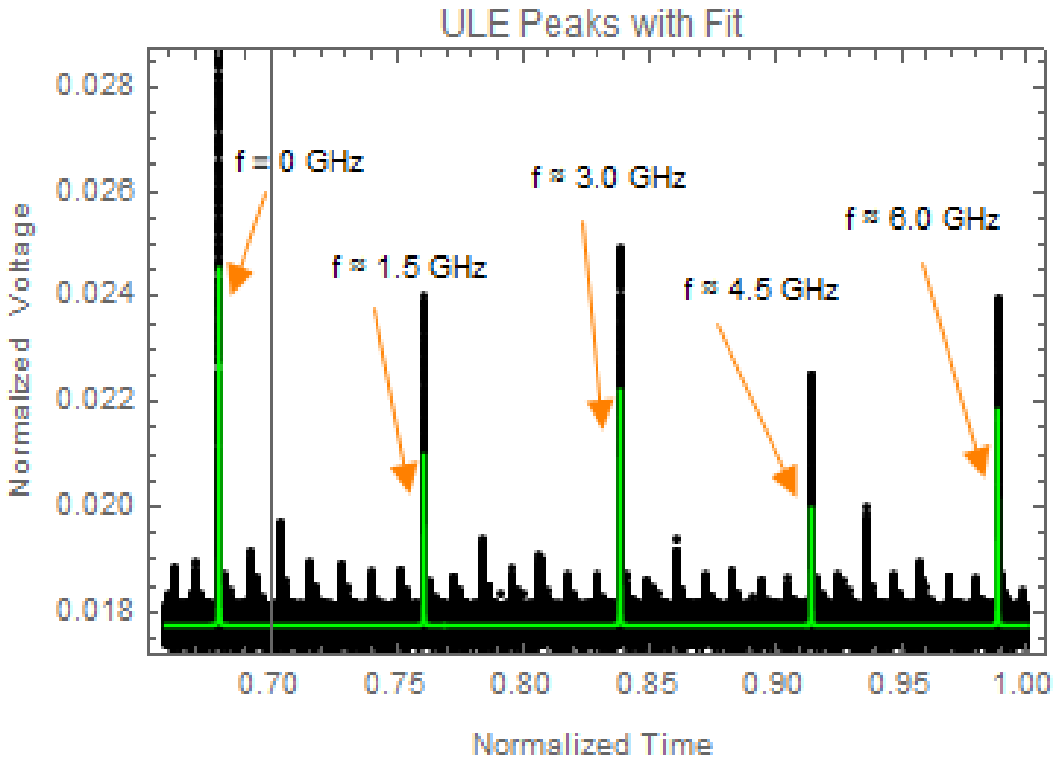

Figure 5: Displays the Airy distribution fit (green) for each of the ULE resonance peaks simultaneous with the nine sampled tellurium spectra shown in Figure 4. Here, each peak was assigned with their frequency relative to the first peak on the left. The true absolute frequency of this first peak is yet to be measured, but assigning it an absolute frequency of 0 GHz did not affect the calculations involved in the data analysis.

Figure 6 – Linear Fit for Ramp Signal

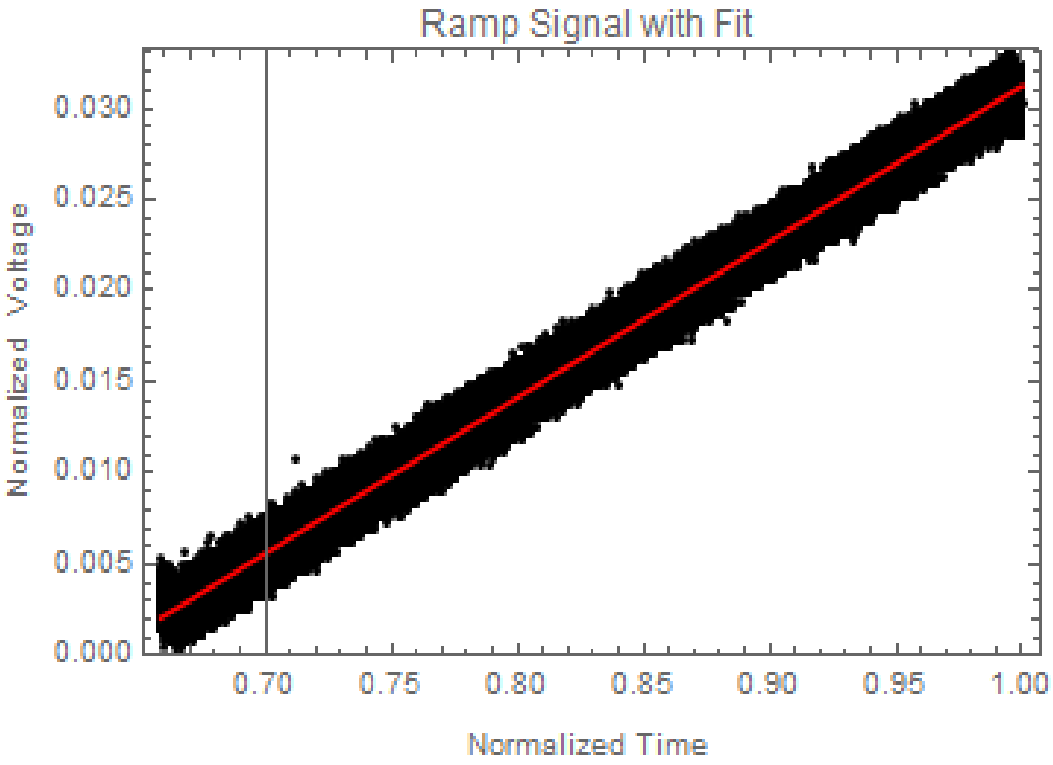

Figure 6: Displays the linear fit (red) of the triangle ramp signal half that produced the tellurium spectra and the ULE peaks of Figure 4 and Figure 5. The measured ramp signal (black) had a voltage bandwidth possibly due to impedance mismatching. This voltage bandwidth was considered in this experiment's error analysis.

Figure 7 – Ramp Voltage and Frequency Relation

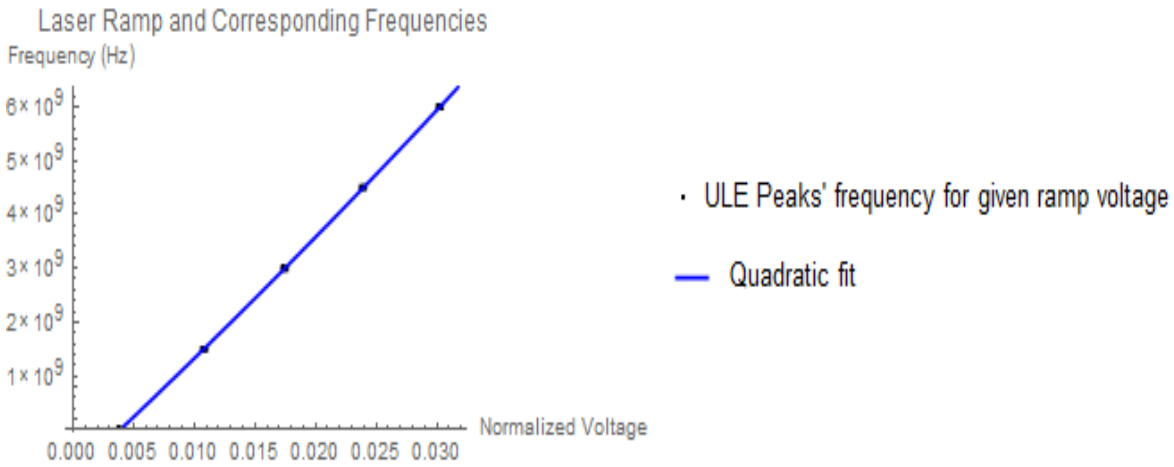

Figure 7: The ULE peak center frequencies each plotted with their corresponding ramp signal voltage. This quadratic fit appears linear. However, a quadratic fit modeled the ramp voltage to frequency relation better than a linear fit, so the quadratic fit was used for later calculations. The small error bars reflect contributions from oscilloscope resolution uncertainty, the ramp voltage bandwidth, and the errors associated with the fit parameters of the tellurium and ULE fits shown in Figures 4 and 5. The error bars are on the scale of MHz.

Figure 8 – Frequency Differences between Tellurium Spectra

Abs Frequency Differences	Calculated Difference (GHz)	Estimated Error (GHz)
-f1 + f2	0.989	0.034
-f1 + f3	1.070	0.034
-f1 + f4	1.806	0.034
-f1 + f5	1.986	0.035
-f1 + f6	2.990	0.036
-f1 + f7	3.185	0.036
-f1 + f8	3.537	0.036
-f1 + f9	4.574	0.037
-f2 + f3	0.081	0.035
-f2 + f4	0.817	0.035
-f2 + f5	0.997	0.035
-f2 + f6	2.000	0.036
-f2 + f7	2.195	0.037
-f2 + f8	2.548	0.037
-f2 + f9	3.585	0.038
-f3 + f4	0.737	0.035
-f3 + f5	0.916	0.035
-f3 + f6	1.920	0.036
-f3 + f7	2.115	0.037
-f3 + f8	2.467	0.037
-f3 + f9	3.504	0.038
-f4 + f5	0.180	0.036
-f4 + f6	1.183	0.037
-f4 + f7	1.378	0.037
-f4 + f8	1.730	0.038
-f4 + f9	2.768	0.039
-f5 + f6	1.004	0.037
-f5 + f7	1.198	0.037
-f5 + f8	1.551	0.038
-f5 + f9	2.588	0.039
-f6 + f7	0.195	0.038
-f6 + f8	0.547	0.039
-f6 + f9	1.585	0.040
-f7 + f8	0.352	0.039
-f7 + f9	1.390	0.040
-f8 + f9	1.038	0.040

Figure 8: This table displays the absolute value of the frequency differences between each of the nine tellurium peaks labeled in Figure 4. The estimated error includes parameter fit errors, oscilloscope resolution error (a max of ~ 13 mv) and scanning ramp width error (~8 mV).

Introduction

Spectroscopy is currently the key to expanding and consolidating the knowledge platform of the atomic physics community. The advancement of spectroscopy technologies and techniques has enabled atomic physicists to probe deeper within the atom's electron and nucleon constituents. More specifically, spectroscopy has enabled atomic physicists to carry out quality assurance of Quantum Electrodynamic Theory (QED) throughout its ongoing development.

The significance of spectroscopy and the collaborative QED quality assurance effort is depicted well though atomic physicists' current efforts to measure energy level transitions of hydrogen-like atoms, atoms with similar single proton-electron structure as a hydrogen atom. There is quite a selection of hydrogen-like atoms, all of which simplify the complexities of measurement and data analysis in unique ways. One popular hydrogen-like atom is the positronium atom, a hydrogen-like atom composed of an electron and its anti-particle known as a positron. At the moment, the Dr. Mill lab and the Dr. Tom lab, located at University of California Riverside (UCR), are currently building an apparatus for the eventual measurement of the 1^3S_1 - 2^3S_1 energy level emission frequency of positronium. This particular measurement has already been repeated a number of times before in previous experiments [1, 2]. However, the most recent of these experiments yielded a measurement precision of ± 3.2 MHz [1], whereas the completed UCR positronium experiment will yield a measurement precision on the kHz scale. The increased precision of this measurement could enable atomic physics theorists to resolve conceptual enigmas of QED theory. For instance, facing theorists at this time is a QED enigma commonly referred to as the proton-charge radius puzzle. The essence of this

puzzle lies in that there is yet no agreeable explanation for the significant 0.0366 fm discrepancy between the proton charge radius, the effective radius of the proton, measured from energy transitions of the hydrogen atom and the muonic hydrogen atom, a hydrogen-like atom with a heavy muon particle instead of a much lighter electron [3,4,5,6,7]. The kHz precision positronium energy measurement to come can help find an agreeable explanation and, possibly, reveal new QED physics. Because of the higher measurement precision required by the process of QED quality assurance, high precision for energy transition measurements are undoubtingly important for experiments such as the UCR positronium experiment.

There are many factors of a spectroscopy setup that influence the measurement precision outcome, one of the important factors being the measurement precision of a frequency reference, an alternate atomic spectrum line of known absolute frequency that is used to calculate the frequency of another atomic spectrum line. During the initial stages of the UCR positronium experiment, a particular spectrum line generated from a tellurium (Te_2) spectrometer will be used as a frequency reference. The tellurium spectrometer, and its variations, has been atomic physicists preferred frequency reference source due to the numerous spectrum lines it yields and the wide frequency range at which these spectrum lines exist. Unfortunately, experiments have found that there needs to be a “standard procedure” taking into account manufacturing discrepancies among tellurium spectrometers before any tellurium spectra can be a frequency reference reproducible beyond MHz precision [8, 9]. Based on such findings and additional complications caused by the short life-time of the positronium atom, the tellurium spectrometer integrated in the UCR positronium experiment will eventually be replaced

by a laser frequency comb, a novel system that can serve as a frequency reference source with a precision approximately 1000 times greater than the highest precision tellurium spectrometers yet [10, 11].

Although the laser frequency comb seems to make the tellurium spectrometer obsolete as a frequency reference source, the novelty and complexity of the relatively expensive laser frequency comb can make it an unfamiliar tool and, thus, a learning hurdle to even the most experienced spectroscopy experimentalists. Furthermore, though high measurement precision is typically the goal for experiments such as the forthcoming UCR positronium experiment, it is not necessarily the goal during an experiment's developmental and initialization stages. The goal during such stages is to efficiently ensure that a valid and reliable frequency measurement can be taken (quality assurance of the experimental setup). Therefore, having a versatile and dependable frequency reference system that is relatively simple to deploy is not just significant to the development of the UCR positronium experiment and similar high precision spectroscopy experiments, but also to any spectroscopy experiment not in need of the high precision offered by a frequency comb. Such spectroscopy experiments include but are not limited to the ongoing barium ion precision experiments and experiments involving active laser-locking techniques [12, 13].

Given the tellurium spectrometer's practical utility as a frequency reference source, a sort of frequency calibration model can thus be a desirable solution to this MHz reproducibility limit. This model will rely on the unique frequency-pressure shifts of individual tellurium spectrum lines to reduce, if not avert, the MHz reproducibility limit. By relying on these unique frequency-pressure shits, the temperature measurement

method and tellurium cell characteristics of a given laboratory's tellurium spectrometer, the current contributors of the MHz calibration uncertainty, become moot in the calibration process. The complete frequency calibration model will entail a standardized calibration procedure relying on two fit tables. The first fit table (fit-table-1) will contain an assortment of frequency differences between tellurium spectra peaks, each frequency difference as a function of the tellurium spectrometer system's cold-point temperature. The second fit table (fit-table-2) will contain the absolute frequencies of each tellurium peak, each individual peak frequency mapped to the same cold-point temperature.

This experiment executes the opening procedure stages for constructing fit-table-1. The relative frequency spacings among nine Doppler-free tellurium spectra at approximately 486 nm, a few of which having been frequency references in past positronium frequency measurements, were calculated at one tellurium spectrometer cold-point temperature. The results from this preliminary calculation and relevant literature findings will be compared to determine the accuracy of this experiment's procedure and thus whether this experiment's procedure has an appropriate part in the construction of fit-table-1.

Methodology

Figure 1 depicts a very basic schematic setup for this preliminary experiment. Both an Ultra-Low-Expansion (ULE) Fabry-Pérot interferometer and the tellurium spectrometer were receiving 486 nm laser light from a ultrastable Toptica laser system. The internal cavity of the Toptica laser system was varied through an electronic triangle ramp signal such that the laser frequency was scanned (modulated) about 486 nm within a maximum range of approximately 6 GHz. The laser frequency scan allowed the ULE to

output five TEM_{00} mode laser pulses with a free spectral range (FSR) of approximately 1.5 GHz. The five ULE resonant pulses were detected through a high gain photodetector (Detector # 1) and displayed on a DSOX3054A Keysight Oscilloscope (8-bit resolution and a maximum 4 Gsa/sec sampling rate) as depicted in Figure 1.

Meanwhile, a portion of the Toptica laser propagated through the tellurium spectrometer. The optical setup of the tellurium spectrometer is presented with more detail in Figure 2. This version of the tellurium spectrometer resembles the tellurium spectrometer used in the experiment of Fee et al. [1]. This experiment's tellurium oven and cell were of the same design and manufacture as the one in the Fee experiment. Also like the Fee experiment, this experiment's tellurium spectrometer employed the spectroscopy technique of Saturation Absorption Spectroscopy (SAS). Overall, the technique of SAS, the scanning of the Toptica laser, and the 60 MHz upshifting of the pump beam through an Acoustic Optic Modulator (AOM) produced hyperfine Tellurium absorption spectra in the vicinity of the positronium 1^3S_1 - 2^3S_1 transition frequency.

These hyperfine tellurium spectra were initially received as time variations of laser intensity by an 1837 GHz Nirvana auto-balancing photodiode (Detector # 2 in Figure 1 and the detector in Figure 2.) The Nirvana detector used a portion of the incoming laser light (reference beam) to subtract the fine structure tellurium spectra and extraneous noise from the signal beam (overlapped by pump beam inside the tellurium oven). The Nirvana detector sent the signal to a 7265 DSP Lock-In amplifier. The Lock-In amplifier transmitted to the Keysight Oscilloscope (Figure 1) the Nirvana signal that was in phase with the AOM modulation frequency of 100 kHz.

The resulting tellurium spectra and ULE signal were thus displayed together on the Keysight Oscilloscope with the laser ramp signal as displayed in Figure 3. The three signals were taken from the Keysight Oscilloscope in csv file format for analysis. The tellurium spectra, ULE resonance peaks, and ramp signal data points were normalized such that each point's measured amplitude (voltage units) and corresponding time (units of seconds) were scaled to be a decimal within the range of zero and one. The tellurium spectra, ULE resonance peaks, and ramp signal data were then respectively fitted to a superposition of Lorentzian functions (nine Lorentzian functions in total), a superposition of generic Airy distributions (five Airy distributions in total), and a linear function. These results are displayed in Figure 4, 5, and 6.

The tellurium spectra fit (Figure 4) generated fit parameter estimates of the normalized time positions of each hyperfine peak's center. The ULE fit (Figure 5) was similarly used to acquire the normalized time positions of each ULE resonant peak's center. In addition, the first ULE peak center was arbitrarily assigned a frequency of 0 GHz while the remaining ULE peaks were assigned a frequency according to the ULE's approximate FSR. The linear ramp fit (Figure 6) was utilized to acquire the ramp voltage as a function of time.

The ramp voltage and time relation from the linear ramp fit was then used to acquire the ramp voltages corresponding to the time positions of each ULE peak center. To take into account any possible non-linearities of the ramp signal, a quadratic rather than a linear model was used to fit these ULE ramp voltages to their corresponding ULE peak center frequencies. The resulting quadratic fit and the ULE peak frequencies with their ramp voltages are shown in Figure 7. The linear ramp fit and the quadratic fit in

combination enabled the conversion of the tellurium peak center times into frequencies. The frequency differences between each of the tellurium peaks were then calculated, thus effectively completing one entry for fit-table-1.

Literature Review

Though tellurium gas is a versatile frequency reference, its molecular structures and dependencies are considered quite complex. Therefore, most of the research community's empirical understanding of tellurium frequency-pressure shifts, frequency shifts of tellurium spectra lines due to shifts in tellurium (Te_2) vapor pressure (temperature), was developed through a few significant experiments. Furthermore, there seems to be no direct presentation of calibration procedures for tellurium spectrometers in the literature. Therefore, the tellurium spectrometer calibration standards, seemingly mutual among spectrometer users, were likewise developed and solidified through the same experiments. In considering these important experiments, the MHz reproducibility limit problem and a solution to it emerges.

Perhaps the experiments that enabled tellurium spectrometers to become widely used were those conducted by physicists of the Aimé Cotton Laboratory (LAC) at the National Center of Scientific Research (CNRS) located in France. Their experiments led to the construction of the most well-known reference table of Doppler-limited tellurium spectra frequencies ranging approximately from 300 to 900 nm [14]. This reference table is commonly referred to as the Tellurium Atlas.

The Tellurium Atlas has enabled atomic physicists to study specific tellurium spectra collectively and with greater depth. In particular, the Tellurium Atlas had enabled Barr and his collaborators to study two tellurium spectra with frequencies near that of

hydrogen-deuterium isotope shifts [15]. For the purpose of later enabling higher precision measurements of Balmer transitions, Barr et al. conducted an experiment aiming to measure the absolute frequencies of two tellurium spectrum lines, which they labelled as the b_1 and b_2 lines, to a precision higher than any of the measurements provided by the Tellurium Atlas. Barr observed the b_1 and b_2 lines by employing SAS, using a 486 nm continuous wave (cw) dye laser (pumped by a krypton-ion laser) as the laser source. The measurements of the two lines' absolute frequencies were obtained by using a confocal Fabry-Pérot interferometer to compare the 486 nm dye laser outputted at the b_1 and b_2 transitions to a 633 nm He-Ne laser. The He-Ne laser itself was mode-locked to a manufactured iodine reference laser to ensure its stability. During these measurements, the tellurium oven within their tellurium spectrometer system was maintained at a cold-point temperature of 625 ± 6 °C. This temperature was deduced from chromel-alumel paired thermocouples located at the wall of the cell body rather than by a more modern sidearm apparatus. In addition to reporting their cold-point temperature measurement method, Barr et al. also reported the heating method of their tellurium oven to be a 10 Ω nichrome heater wire.

Barr et al. were able to measure the absolute frequencies of both lines to an approximately 0.25 MHz precision uncertainty. In their calculation of this precision uncertainty, Barr et al. concluded that a considerable portion of their precision uncertainty comprised of frequency-pressure shift uncertainty, an uncertainty corresponding to their cold-point measurement uncertainty and their frequency-pressure shift (gradient) measurements of the b_1 line (0.041 ± 0.005 MHz/torr).

Barr et al. thus conducted one of the first experiments illustrating the large effect

frequency-pressure shifts can have on the precision measurements of tellurium spectra lines. They also demonstrated frequency-pressure shift uncertainty to be rooted in a lab's measurement of cold-point temperature. However, this experiment, which will now be referred to as the Barr experiment, did not explore calibration procedures of tellurium spectrometers. Nonetheless, the tellurium spectrometer design featured in the Barr experiment has been used as basis for proceeding experiments.

Shortly after the Barr experiment, McIntyre and his fellow researchers conducted an experiment similar to the Barr experiment [16]. However, unlike the Barr experiment, the goal of McIntyre et al. was to measure the absolute frequency of a tellurium line (termed as the e_3) that had been used as a frequency reference for positronium in previous experiments. One of the main differences between the two experiment setups was that this experiment compared the spectra of two tellurium spectrometers (both employing SAS) rather than comparing the spectra of a tellurium spectrometer to a He-Ne laser. By systematically scanning the laser frequency, the first tellurium spectrometer produced the b_2 line while the second tellurium spectrometer produced the e_3 line. In order to use the b_2 absolute frequency measurement of the Barr experiment, McIntyre et al. reported efforts to replicate the tellurium spectrometer settings of the Barr experiment. McIntyre claimed their b_2 line to replicate the Barr experiment b_2 line on the grounds that their tellurium cell dimensions and the tellurium oven manufacture were similar to that of the Barr experiment. McIntyre further emphasized the validity of their claim on the basis that their cold-point temperature for both of their tellurium ovens was set (513 ± 5 °C) such that their linear absorption measurements were also similar to that of the Barr experiment. Despite the tellurium spectrometer similarities, the tellurium ovens used by McIntyre

were of clamshell design, thus employing a heating method different from the method used in the Barr experiment.

McIntyre measured the e_3 line absolute frequency to a precision uncertainty of 0.71 MHz, attributing most of this uncertainty to their etalon calibration process. McIntyre also measured the pressure shifts for the e_3 and b_2 lines to be -1.05 ± 0.32 MHz/torr and -1.06 ± 0.6 MHz/torr, respectively. The frequency-pressure shift uncertainty factored into their overall uncertainty was approximately the same magnitude as was reported in the Barr experiment, mainly because both of their cold-point temperature uncertainties were essentially equivalent.

Since the two frequency-pressure shift measurements were within error, this experiment did not succeed in demonstrating the unique frequency-pressure shifts of tellurium spectra lines. Nonetheless, a worthy note is that the resulting cold-point temperature discrepancy between the tellurium ovens of McIntyre's experiment and the tellurium oven of Barr's experiment was significant (approximately a 110 °C difference). McIntyre did not remark on this significant cold-point temperature discrepancy, thus failing to offer insight as to its cause or its implications. McIntyre also did not investigate the possible implications of their few tellurium oven design discrepancies. Regardless of these shortcomings, the overall experiment well demonstrated the typical calibration standards deemed acceptable for replicating tellurium spectrometer setups: matching tellurium oven manufacturer, matching tellurium cell size dimensions, and matching linear absorption measurements.

Despite their carefully calibrated measurement of the e_3 line, McIntyre and others later remeasured the b_1, b_2, e_3 line and another popular tellurium frequency reference line

labeled as i_2 [8]. This was motivated by the 20 MHz and 1 MHz discrepancies found between experiments' positronium and hydrogen transition measurements, respectively. McIntyre noted that these discrepancies occurred despite the fact that the compared experiments utilized "nominally identical Te_2 cells". McIntyre et al. thus sought to remeasure the aforementioned absolute frequencies through an approach slightly different from their earlier experiment. The absolute frequency measurements in this experiment were obtained by comparing the spectra generated from a tellurium spectrometer system (one from the earlier experiment of McIntyre et al. [16]) to the frequency standard spectra generated from an iodine spectrometer. Like the tellurium spectrometer of the earlier experiment, the iodine spectrometer utilized SAS and an oven system but, instead of Te_2 gas, the cell unit within the oven contained diatomic, isotopic iodine gas ($^{129}I_2$).

Along with their absolute frequency estimates to a total uncertainty of approximately 0.61 MHz, McIntyre et al. also measured the frequency-pressure shifts of the four tellurium lines. While the measured frequency-pressure shifts for the b_2 and e_3 lines were equivalent to the shifts measured in their earlier experiment, the frequency-pressure shifts of the b_1 and i_2 lines (-1.45 ± 0.25 and -1.10 ± 0.10 MHz/torr) were measurably different relative to each other and to the b_2 and e_3 lines. Furthermore, because the significant lack of agreement between the b_1 absolute frequency and b_1 frequency-pressure shift measurements of the Barr experiment, McIntyre concluded that their tellurium spectrometer conditions were probably, after all, not identical to tellurium spectrometer conditions used in the Barr experiment.

Ultimately, the results of this experiment not only offered evidence of unique frequency-pressure shifts among tellurium spectra, but also established that the tellurium

spectrometer calibration standards could not accurately reproduce another lab's tellurium lines to a precision greater than the MHz scale. However, while McIntyre commented on the large discrepancy between the b_1 frequency-pressure shifts measured by them and the Barr experiment, their experiment did not produce results that explained the discrepancy. Furthermore, though McIntyre identified a MHz calibration uncertainty unreducible by the current tellurium calibration standards, McIntyre could not quantify this MHz limit nor identify its source beyond the vague association to tellurium oven manufacturing.

An experiment conducted by Barwood and his fellow colleagues fortified the findings of unique tellurium frequency-pressure shifts and further explored the MHz calibration uncertainty [9]. Like McIntyre in their previously described experiment [8], Barwood et al. noted the hydrogen frequency measurement discrepancies among experimenters, experimenters who used seemingly identical tellurium spectrometers as frequency reference sources. Barwood also commented on the same MHz reproducibility limit of tellurium spectrometers found in the aftermath of the 2nd McIntyre experiment [8], indirectly labeling this reproducibility limit as a "cell-to-cell tellurium frequency" uncertainty. The experiment of Barwood thus aimed to assess systematic errors associated with the tellurium spectrometer by remeasuring five pervious absolute frequency measurements of tellurium spectrum lines (d_4, b_2, b_1, e_3, i_3) in six different tellurium cells, doing so through the interferometric comparison method used in the Barr experiment. Unlike the previously described experiments [8,15,16], Barwood obtained the Doppler-free tellurium spectra by constructing two tellurium spectrometers setups that exploited frequency modulation (FM) techniques. Each of the two corresponding tellurium ovens had a sidearm and a R-type thermocouple for direct measurement of the

cold-point temperature. Mica insulation surrounded the cells when inside an oven to improve cold-point temperature stability. To ensure additional temperature stability, each one of the ovens' heaters were controlled by a temperature controller located outside of the ovens. Despite these tellurium spectrometer design changes, their two tellurium spectrometers were calibrated to the tellurium spectrometer of the 2nd McIntyre experiment according to the calibration standards established in the 1st McIntyre experiment [16]. As a result of their calibration, they carried out their absolute frequency measurements at a cold-point temperature of 520 ± 2 °C.

Barwood et al. measured the five tellurium spectra lines to total uncertainty of 0.47 MHz. While frequency-pressure shifting was among the smallest uncertainty contributors, the estimated cell-to-cell uncertainty (approximately 0.37 MHz) was one of the largest uncertainty contributors. Based on the linear absorption measurements spanning a cold-point temperature range of approximately 160 °C, Barwood deemed this cell-to-cell uncertainty to be due to the variations of tellurium gas quantity within each cell. Also based on the linear absorption measurements, Barwood emphasized that the Barr and the McIntyre experiments' differing cold-point temperature measurement methodology led those experimenters to report faulty cold-point temperatures, which created discrepancies among their absolute frequency results. In assessing these absolute frequency discrepancies, Barwood found that those faulty cold-point measurements effectively contributed a calibration uncertainty to their own absolute frequency measurements on the scale of 0.2 MHz. Barwood also calculated the frequency-pressure shifts for the five tellurium lines to an improved precision than the 2nd McIntyre experiment (d_4, b_2, b_1, e_3, i_3 with -1.21, -1.35, -1.51, -1.35, -1.17 MHz/torr, respectively,

and each with an uncertainty of 0.04 MHz/torr). Overall, Barwood et al. reasoned that their absolute frequency measurements were within 1 MHz uncertainty of the previously discussed experiments.

The increased precision of the frequency-pressure shift measurements and the additional measurement of the d_4 line confirms that tellurium lines do indeed have relative but unique frequency-pressure shifts. However, Barwood et al. strangely did not offer an explanation for the discrepancy between their and the Barr experiment's b_1 frequency-pressure shift measurement. Though the case, Barwood's observation of critical cold-point temperatures (pressures) for each cell, temperatures at which linear absorption and frequency-pressure shift trends drastically deviate, can be used to deduce an explanation for this discrepancy, an explanation not relevant to goals of this literature review. In addition to this experiment's exploration of tellurium frequency-pressure shifts, this experiment highlighted Te_2 quantity variations among cells and cold-point temperature measurement methods to contribute to the MHz reproducibility limit of tellurium spectrometers.

If using the estimates of the Barwood experiment, these two tellurium spectrometer design factors contribute an approximate 0.42 MHz uncertainty to absolute frequency measurements. These two factors are large systematic errors not yet accounted for by the current tellurium calibration standards expressed through the 1st McIntyre experiment. However, the unique frequency-pressure shifts of tellurium lines are calculable to an uncertainty minor in comparison to this 0.42 MHz uncertainty. Thus, incorporating the unique frequency-pressure shifts into the calibration standards can reduce the systematic calibration error associated with Te_2 quantities and cold-point

temperature measurement methods.

Analytical Discussion

The frequency differences between all the nine Tellurium spectra lines (Figure 4) are displayed in the table of Figure 8. A couple frequency differences found in the literature are within error of the frequency differences calculated here. Based on the absolute frequencies reported by Barwood et al. [9], they measured a frequency difference between the i_3 and e_3 peak of $2.5898 \pm 7.0 \times 10^{-4}$ GHz. The frequency difference of the corresponding peaks as measured by this experiment ("$f_9 - f_5$" in Figure 8) was 2.588 ± 0.039 GHz. Furthermore, Fee et al. [1] reported a frequency difference of approximately 175 MHz between the e_3 peak and a broad peak Barwood labeled as d_3 [9]. The frequency difference of the corresponding peaks as measured by this experiment was 180 ± 36 MHz ("$f_5 - f_4$" in Figure 8). The imprecise results of this experiment are within error of the results found in the literature, thus indicating that the methodology carried out in this experiment can serve as the procedural basis for constructing fit-table-1.

As briefly described in Figure 8, the error estimated for the measured frequency differences includes the oscilloscope resolution uncertainty and the ramp signal's voltage bandwidth. Though not formerly considered within the calculations leading up to Figure 8, The uncertainty contributions due to the temperature uncertainty of the cold-point temperature were estimated though standard calculations [17] to be approximately $\pm$ 25 kHz, an insignificant uncertainty contribution in relation to oscilloscope resolution and ramp width. Furthermore, though the manufacture specifications are yet to be confirmed, the ULE cavity was built to ensure high temperature stability and vibration impedance such that its frequency drift is less than 5 kHz/ day [18].

Conclusions

The consistency between the tellurium spectra frequency differences calculated from this experiment and the literature indicates that this experiment's procedure can serve as the foundation for constructing fit-table-1 of the frequency calibration model. However, the complete construction of fit-table-1 would thus require repeating this experiment's procedure for different cold-point-temperatures and discerning which peak pairs display a measurable, high precision frequency shift over changes in temperature.

To maximize the precision of fit-table-1, the resolution and systematic errors noted within this experiment must be reduced if not eliminated. The oscilloscope used in this experiment was not the most appropriate for recording the high finesse (small frequency bandwidth) ULE resonant peaks. Thus, an oscilloscope that balances a higher bit resolution and sampling rate is desired. While the resolution uncertainty is straightforward to address, the ramp bandwidth noise is not, at least momentarily. Currently, the cause of the ramp bandwidth noise is unknown, though the cause is speculated to be related to some impedance mismatching. Furthermore, to ensure precision of the information presented in fit-table-1, the temperature uncertainty of the measured cold-point-temperature must be minimized. The largest proponent of temperature uncertainty within this experimental setup was statistical, uncertainty due to the thermal interactions between the tellurium oven and the outside lab environment. Though the fiber glass insulation applied to the tellurium oven observably reduced the statistical uncertainty, plans have been made to further insulate the tellurium oven with thermally insulating millboard material.

In addition to the high precision, fit-table-1 needs to be reliable to other

laboratories. As a start to ensuring reliability, the cold-point temperature measurements required throughout the construction of fit-table-1 need to be correct, meaning the cold-point temperature measurements need to be calibrated such that the coldest point inside the tellurium oven is actually measured. Such a cold-point temperature calibration was not done prior to this experiment. For the experiments to come, the cold-point temperature calibration of a tellurium spectrometer conducted by Barwood can be emulated [9]. In addition, the reliability of fit-table-1 can also be ensured by avoiding the critical temperatures associated with individual tellurium cells. Based on the observations of Barwood, these critical temperatures can be avoided by ensuring the Te_2 quantity of each cell is above a particular threshold [9].

Bibliography

[1] Fee, M. S., S. Chu, A. P. Mills Jr., R. J. Chichester, and D. M. Zuckerman. "Measurement of the positronium 1^3S_1-2^3S_1 interval by continuous-wave two-photon excitation." *Physical Review A,* vol. 48, no. 1, July 1993, pp. 192-220.

[2] Chu, Steven, Allen P. Mills Jr., and John L. Hall. "Measurement of the Positronium 1^3S_1-2^3S_1 Interval by Doppler-Free Two-Photon Spectroscopy." *Physical Review Letters*, vol. 52, no. 19, 7 May 1984, pp. 1689-92.

[3] Antognini, Aldo, et al. "Proton structure from the measurement of 2S-2P transition frequencies of muonic hydrogen." *Science*, vol. 339, no. 6118, 2013, pp. 417-20., doi:10.1126/science.1230016.

[4] Mohr, Peter J., et al. "CODATA Recommended Values of the Fundamental Physical Constants: 2010." *Reviews of Modern Physics*, vol. 84, no. 4, 2012, pp. 1527–1605., doi:10.1103/revmodphys.84.1527.

[5] Hill, Richard J., and Gil Paz. "Model-Independent Extraction of the Proton Charge Radius from Electron Scattering." *Physical Review D*, vol. 82, no. 11, Mar. 2010, doi:10.1103/physrevd.82.113005.

[6] Gabrielse, G., et al. "New determination of the fine structure constant from the electron g value and QED." *Physical Review Letters*, vol. 97, 2006, pp. 030802-1--4, doi:10.1103/PhysRevLett.97.030802.

[7] Odom, B., D. Hanneke, B. D'Urso, and G. Gabrielse. "New measurement of the electron magnetic moment using a one-electron quantum cyclotron." *Physical*

Review Letters, vol. 97, 2006, pp. 030801-1--4, doi:10.1103/PhysRevLett.97.030801. Accessed 5 June 2017.

[8] McIntyre, D. H., et al. "Interferometric frequency measurement of $^{130}Te_2$ reference transitions at 486 nm." *Physical Review A*, vol. 41, no. 9, 1 May 1990, pp. 4632-35.

[9] Barwood, G. P., et al. "Interferometric measurements of $^{130}Te_2$ reference frequencies for 1S-2S transition in hydrogenlike atoms." *Physical Review A*, vol. 43, no. 9, 1 May 1991, pp. 4783-90.

[10] Scholl, T. J., Steven J. Rehse, R. A. Holt, and S. D. Rosner. "Absolute wave-number measurements in $^{130}Te_2$ reference lines spanning the 420.9-464.6-nm region." *Journal of the Optical Society of America B: Optical Physics*, vol. 22, no. 5, May 2005, pp. 1128-33. *Scholarship at UWindsor*, scholar.uwindsor.ca/physicspub/15.

[11] Bradler, Maximilian, and Marc Fischer. "7th optical frequency comb seminar." MenloSystems, 18 Jan. 2016, Martinsried, Germany. Power Point Presentation.

[12] Dutta, T., D. De Munshi, and M. Mukherjee. "Absolute Te_2 reference for barium ion at 455.4 nm." *Journal of the Optical Society of America B: Optical Physics*, vol. 33, no. 6, June 2016, pp. 1177-81, https://doi.org/10.1364/JOSAB.33.001177.

[13] Burns, I. S., J. Hult, and C. F. Kaminski. "Use of $^{130}Te_2$ for frequency referencing and active stabilisation of a violet extended cavity diode laser." *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, vol. 63, no. 5, Apr. 2006, pp. 905-09. *ScienceDirect*, https://doi.org/10.1016/j.saa.2005.10.053.

[14] J. Cariou, P. Luc, “Atlas du spectre d’absorption de la molecule de tellure, Laboratoire Aimé-Cotton, CNRS II”, Orsay, France (1980).

[15] Barr, J.R.M., et al. "Interferometric frequency measurements of $^{130}Te_2$ transitions at 486 nm." *Optics Communications*, vol. 54, no. 4, 15 June 1985, pp. 217-21.

[16] McIntyre, D. H., and T. W. Hansch. "Absolute calibration of the $^{130}Te_2$ reference line for positronium 1^3S_1-2^3S_1 spectroscopy." *Physical Review A*, vol. 34, no. 5, Nov. 1986, pp. 4504-07.

[17] Machol, Robert E., and Edgar F. Westrum Jr. "Vapor Pressure of Liquid Tellurium." *Journal of the American Chemical Society*, vol. 80, no. 12, June 1958, pp. 2950-52.

[18] Stable Laser Systems, Boulder, CO 80301 USA.