Alkali Linewidths under High Temperatures and Pressures of 3He

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Alkali Linewidths under High Temperatures and Pressures of $^3$He

A thesis submitted in partial fulfillment of the requirement for the degree of Bachelor of Science with Honors in Physics from the College of William and Mary in Virginia,

by

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Accepted for Honors

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Abstract

Current research at Thomas Jefferson National Accelerator Facility is being conducted to study the spin structure of the neutron through collisions with polarized $^3$He nuclei. The helium is contained in high pressure glass vessels (called cells) along with nitrogen, rubidium, and potassium. To deduce the spin structure from collisions, we need to know the precise number density of $^3$He in the cell. The process of polarizing $^3$He through spin-exchange optical pumping requires nitrogen and alkali metal. We can use the absorption linewidths of rubidium and potassium to more accurately determine the density of helium.

Throughout my research, I collected absorption measurements in order to explore how different laser powers, temperatures, and polarization angle affected the linewidths of the Rb D1, Rb D2, K D1, and K D2 transitions. I also plotted the predicted number densities using the empirical model derived in Kluttz et al. 2013 and tested the limitations of this model. My work showed support of this model in the case of both rubidium transitions, but showed that there is some undetermined effects impacting potassium as laser power is manipulated.
Acknowledgments

I would first like to thank Professor Todd Averett for all his help over the last year. His balance of instruction to independence let me discover for myself how fulfilling research can be, even when things break unexpectedly. For times when things did break, I’m also thankful for our machine shop director, Will Henninger, and glass blower at Princeton, Mike Souza. I would also like to thank the AMO group of Prof. Eugeniy Mikhailov, Prof. Irina Novikova, and Prof. Seth Aubin for all the expertise they offered me. Thank you to my friends and family for their support during the course of year, and most of all, thank you to my forever editor, Sida Wilson.
Chapter 1

Introduction

1.1 Goal

At Thomas Jefferson National Accelerator Laboratory, there is ongoing research that aims to understand how the orbital angular momentum of quarks and gluons sum to the total spin of the protons and neutrons. Because the neutron is neutral and free neutrons only have a half-life of about 15 minutes, the $^3\text{He}$ nuclei are used as a surrogate for neutrons. The $^3\text{He}$ nuclei have two protons with spins that are anti-parallel, so the neutron determines the total nuclear spin. Effectively, the $^3\text{He}$ nucleus will behave very similar to the neutron. These nuclei are polarized using SEOP in a cell (see Figure 1.1) [1]. Certain experiments at Jefferson Lab are contingent upon knowing the number density of helium to within 2%, but the current empirical model stated in equation 1.2 from Kluttz et al. fails to adequately predict the number density when using linewidths at higher temperatures [2]. Thus my goal is to validate the empirical model presented in Kluttz et al. or offer corrections that can improve our ability to calculate the number density of $^3\text{He}$ based on broadening effects on the alkali lineshapes. I am also seeking an explanation for any unexplained trends in the behavior of linewidths under variations in laser intensity or temperature.
1.2 Background and Theory

I will discuss two areas of background knowledge for my project. First, I will describe the apparatus and the purposes for which it is designed. Second, I will explain how I study the properties of the cell using optical properties of alkali atoms.

Starting with the apparatus of interest, we will discuss the cell shown in Figure 1.1. A cell is a sealed aluminosilicate glass vessel made for the purpose of studying $^3$He polarization. In general, a cell contains $^3$He, N$_2$, and either one or two alkali metals—either rubidium or a combination of rubidium and potassium is used. Looking at the cell pictured in Figure 1.2, we see that it has a spherical pumping chamber at the top which connects to the thin target chamber by way of two transfer tubes. Cells must be made of aluminosilicate glass to provide the conditions needed for spin-exchange optical pumping (SEOP) [3]. At room temperature, the cell is filled to about 10 atm of gas, and pumping is done at approximately 235°C [4].
SEOP, or hybrid SEOP in the case of two alkali metals, is the method of polarizing noble gas nuclei with light [5]. Circularly polarized photons in the near infrared transfer angular momentum to the rubidium atoms as they undergo D1 or D2 transitions. (The D1 transition is from the ground state $S_{1/2}$ to the $P_{1/2}$ state and the D2 transition is from the $S_{1/2}$ to the $P_{3/2}$ state [6, 7].) After the near IR light pumps the atoms, collisional mixing between alkali atoms and $^3\text{He}$ nuclei polarizes the $^3\text{He}$. The improvement of hybrid cells over standard Rb-only cells is demonstrated in the increased polarization percentage at lower laser power. Rubidium atoms still absorb circularly polarized photons, but, via collisions, they transfer their angular momentum
Figure 1.2: Hybrid cell containing $^3$He, N$_2$, Rb, and K, connected to thermocouple leads for measuring temperature, shown inside the experimental oven.

to a greater number of potassium atoms. These potassium atoms are more efficient at polarizing the helium [4]. The mass ratio of rubidium to potassium is chosen such that the number densities will be the ‘right’ proportion at operating temperatures. This right proportion is the best ratio for SEOP to occur and is a significantly higher number density of potassium than rubidium in the vapor phase. I will be using the uncommon unit “amagat” for number density here, and it is defined such that 1 amg is the number density of an ideal gas at standard temperature and pressure (0°C and 1 atm). Now, in order to prevent fluorescence from equilibrating the helium out of its hyperpolarized state, nitrogen molecules are added to quench this re-emitted light [5]. Lastly, the glass itself is made to have very few paramagnetic impurities that
could cause depolarization when atoms of helium collide with the inner surface. The target cell I worked on, named Florence, is pictured in Figure 1.2. The cell is made of aluminosilicate glass and contains both K and Rb.

SEOP relies on the fact that the polarized alkali atoms can collisionally mix with, and thus transfer their angular momentum to, helium. The alkali metal atoms undergo D1 and D2 transitions, shown in Figure 1.3. Heating the cell to high temperatures vaporizes the alkali metal, and a near infrared laser is used to access these transitions. The resonances of the D1 and D2 transitions are 769.90 nm and 766.49 nm for K and 794.76 nm and 780.03 nm for Rb in air [8, 9]. The vapor pressures of rubidium and potassium increase rapidly with temperature, and their number densities are on the order of $10^{15}$ cm$^{-3}$ at the operating temperatures of cells [4].

Before I move onto data collection, I will provide some background on absorption spectroscopy. We will hypothesize the characteristics affecting absorption: imagine a single atom sitting perfectly still in free space. Hypothetically, this atom could only absorb the exact frequencies of light which would excite it to a higher energy
level. However, this ‘perfectly still’ atom is not physical and we must consider the
Heisenberg uncertainty principle in its energy-time form:

\[ \Delta E \Delta t \geq \frac{\hbar}{2} \]  

[10]. The \( \Delta E \) here represents the uncertainty in magnitude of energy for any of the
excited states and \( \Delta t \) refers to the lifetime of said excited state. This variation in
absorption is known as natural broadening. Next, we consider temperature effects.
The atom must have nonzero temperature, so at any instant it is moving stochastically
at velocities described by the Maxwell-Boltzmann distribution. If the particle is
moving toward the photon, the light will be blueshifted in the frame of atom and
appear to be at a higher frequency. The same argument applies if the atom was
traveling directly away from the photon, causing it to be redshifted. When using
the Maxwell-Boltzmann distribution to account for the variation in velocities due to
temperature, we expect the absorption resonance lineshape to be Gaussian [11]. This
is known as Doppler broadening. Lastly, if we consider not just one atom, but a dense
population of atoms, there is another mechanism for broadening the lineshape. When
other atoms are present, the proximity of their outer shells can lead to shifts in the
energy levels due to electrostatic repulsion. This also occurs due to collisions, which
is distinguished as collisional broadening. However, for the sake of simplicity we will
group collisional broadening into the category of general pressure broadening. The
effect on lineshape due to pressure broadening is approximately Lorentzian, and at
the pressures in our system, this effect dominates the Gaussian shape from Doppler
broadening [11].

In the recent literature, Kluttz, Averett, and Wolin [2] determined an empirical
formula relating the helium number density (\( \rho \)), temperature (\( T \)), and linewidth (\( \gamma \))
Table 1.1: This table shows the empirical values presented directly from Kluttz et al. 2013.

<table>
<thead>
<tr>
<th></th>
<th>α (GHz/amg)</th>
<th>n</th>
<th>β (GHz)</th>
<th>$\chi^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rb $D_1$</td>
<td>18.31 ± 0.07</td>
<td>0.26 ± 0.04</td>
<td>−0.19 ± 0.13</td>
<td>1.1</td>
</tr>
<tr>
<td>Rb $D_2$</td>
<td>20.51 ± 0.08</td>
<td>0.39 ± 0.04</td>
<td>−0.35 ± 0.15</td>
<td>1.0</td>
</tr>
<tr>
<td>K $D_1$</td>
<td>14.26 ± 0.09</td>
<td>0.44 ± 0.06</td>
<td>0.04 ± 0.11</td>
<td>1.9</td>
</tr>
<tr>
<td>K $D_2$</td>
<td>19.59 ± 0.10</td>
<td>0.39 ± 0.05</td>
<td>0.11 ± 0.13</td>
<td>1.5</td>
</tr>
</tbody>
</table>

for Rb $D_1$, Rb $D_2$, K $D_1$, and K $D_2$ transitions according to

$$\gamma(\rho, T) = \alpha \rho \left(\frac{T}{T_0}\right)^n + \beta$$  \hspace{1cm} (1.2)

working over the 340-400 K range (~70°C - 130°C). $T_0$ is the reference temperature, set to 353 K. $\alpha$, $\beta$, and $n$ are empirically determined constants that were found per metal and per transition, which results in four parameter sets in total (see Table 1.2). This functional dependence was determined using a cell that could be evacuated and filled with different densities of $^3$He and N$_2$. Unfortunately, the filling process for the target cells does not allow for as an easy assessment of initial fill pressure as the adjustable cell. Thus, we use simpler cell as calibration for the permanently sealed target cell. This temperature range appears to not be representative of the $\gamma$ versus temperature trend at temperatures greater than 400 K.

The working temperatures for the present cell are greater than 430 K (157°C), so any model used to predict number density of helium must at least be consistent for temperatures up to ~500 K (227°C).
Chapter 2

Experimental Technique and Methods

Given that the current model begins to diverge from the experimental measurements at temperatures higher than 130°C, first I collected data on temperature trends (of all four transitions) up to approximately 150°C. Because the reason for these deviations in predicted density is still unknown, I also searched for any laser power dependence across a variety of temperatures for the Rb D1 absorption at 127, 137, 150°C. Data collection was done using the setup shown in Figure 2.1. The near-infrared laser scans through a range of wavelengths, approximately 5-20 nm depending on the transition(s) being studied. The path is described in Figure 2.1 and a photograph of the components is shown in Figure 2.2.
Figure 2.1: Schematic of near infrared absorption setup. The beam is directed with two mirrors through an iris, where it then is split into a portion which goes to the wavemeter. The majority of the beam intensity continues forward through a linear polarizer and then through another beam splitter. One portion goes to the reference photodiode while the main portion passes through another iris before being directed into the cell. After passing through the cell and oven, the beam is captured by the transmitted photodiode.

Figure 2.2: Photograph of actual components in optical setup.
From Ref. [2], the equation relating the log of the ratio of intensities to the linewidth is derived to be:

\[
\ln \left( \frac{S_t}{S_r} \right) = \left( \frac{-\gamma[A] \sigma_0 L}{2\pi} \right) \left( 1 + 0.6642 \times 2\pi \Delta t_d \right) + \ln \left( \frac{G_r}{G_t} \right)
\]  
(2.1)

where \( S_t \) and \( S_r \) are the measured transmitted and reference signal, respectively. These signals are the product of the associated intensity and gain of the photodiodes (\( G_t \) or \( G_r \), respectively). Furthermore, \( L \) is the path length through the cell, \( [A] \) is the alkali metal density, \( \gamma \) is the full width at half maximum (FWHM), \( \sigma_0 \) is the total optical absorption cross section over all frequencies, \( \Delta \) is really \( \Delta(\nu) = \nu - \nu_0 - \delta \) (\( \nu_0 \) is the natural resonant frequency and \( \delta \) is the collisional frequency shift) and \( t_d \) is the collision duration.

Using \( \ln(\frac{S_t}{S_r}) \), where \( S_t \) and \( S_r \) are the signals measured at the transmitted and reference photodiodes, respectively, and the frequency of light, one can create a Lorentzian plot describing the absorption. (Often these are simply written as \( T \) and \( R \)) I determined the linewidth, \( \gamma \), by fitting the plots of \( \ln(\frac{T}{R}) = y(\nu) \) to the modified Lorentzian in Eqn. 2.2,

\[
y(\nu) = M \frac{\gamma[1 + t(\nu - \nu_c)]}{(\nu - \nu_c)^2 + (\gamma/2)^2} + a\nu + b
\]  
(2.2)

which was determined in Kluttz, 2013 [2]. In Eqn. 2.2, \( \nu_c \) is the resonant frequency of the transition, \( M \), \( a \), and \( b \) are constants for scaling the modified Lorentzian. For clarity, note that the \( a \) and \( b \) shown in Eqn. 2.2 have no relationship to the \( a \) and \( b \) in the linear regressions later. Here \( a \) and \( b \) parameterize the background. Lastly, the fact it is a modified Lorentzian is from the \( t(\nu - \nu_c) \) term.
Figure 2.3: Shown here is an example fit for the Rb D1 transition. The parameters in the textbox align with Eqn. 2.2.

In Figure 2.3, the parameters are listed with their standard deviations. The standard deviation on the linewidth $\gamma$ is used later for the error bars on the plots of $\gamma$ vs. temperature and $\gamma$ vs. power.

Fitting any function to the data is most accurate when the plot has very little noise. To minimize obfuscation of the absorption lineshape, laser power is increased to raise the signal to noise ratio. See Figure 2.4 for absorption data taken with a good signal to noise ratio, and see Figure 2.5 for absorption data with a low signal to noise ratio. The four graphs in each figure show signal measured at the reference and transmitted photodiodes (PD) in the bottom left and bottom right, respectively. The upper left plot shows the ratio of transmitted and reference signals; the upper right shows the pertinent natural log of the ratio of the transmitted and reference signals. These data sets are all plotted with respect to wavelength.

Another possible issue with fitting, besides the signal to noise ratio, comes at low laser intensity. When the laser power is so low that the transmitted signal is measured
to be zero, then the plot of $\ln(T/R)$ “bottoms out” and no longer takes the shape of a Lorentzian. When the beam is being fully absorbed by the vapor, it is difficult to determine the FWHM for the absorption lineshape using Eqn. 1.2.

Besides full absorption occurring at high laser intensity and the signal to noise ratio, we must consider optical saturation. Saturation is based on the proportion of atoms in their excited state relative to the incident photons. Unlike full absorption, there is a continuum between unsaturated, saturating, and saturated, but I will only be describing the two extremes here. If laser intensity is low enough that small increases in laser intensity have little effect on the transmitted signal, the medium (alkali vapor in this case) is unsaturated. If the laser intensity is so high that atoms do not have enough time to decay from the excited state before another photon passes them, then we consider the medium to be saturated. In the saturated case, linear increases in laser intensity yield linear increases in transmitted signal. It was not obvious when this project began, but data should be collected in the unsaturated range to prevent power dependencies from skewing the linewidths.
Figure 2.4: Here is an example of an absorption spectrum collected at a high enough laser power such that noise is minimized.

Figure 2.5: Example data collected at lower laser power (\( \sim 0.1 \text{ mW} \)) so the signal to noise ratio is low.
To examine the power dependence for any single (or double, in the case of potassium) resonance, I adjusted the angle of the linear polarizer (see Figure 2.1) to limit the intensity downstream. The light coming out of the laser is polarized, so rotating a polarizer allows laser power on the cell to be decreased without varying the frequency or anything about the laser itself. Using a power meter, I recorded the measured power immediately after the polarizer and before any other optical components. I did this at the same wavelength for each plot—either at the initial or final wavelength of the scan. However, because power was recorded at different wavelengths between groups of trials, it is only valid to compare relative power trends. The reason for the power variation with frequency originates at the tunable diode laser. The nature of the laser system causes the power output to change as it changes frequencies; this is why the power ranges for Rb transitions can extend to well over double the laser power used during the for K transitions.

A tunable diode laser was used for these measurements because it can lase over a wide range of wavelengths by manipulating the angle of an internal grating. Without any modifications the laser will produce a different intensity/power at different output wavelengths. When the laser is fixed at one wavelength or when scans are over a small wavelength range (less than 1 nm), the variation in power is minimal (±1 mW). However, any time the potassium resonances are examined, scans must be taken from 765 to 772 nm. This 7 nm range means the power will increase significantly (approximately a factor of two) between the initial and final wavelength. The two potassium resonances must be measured and fit together because with high broadening there is no longer significant spacing between the wings of the two Lorentzians.

After collecting my first data set using the standard target cell pictured in Figure 1.1, I filled a new, simpler cell with a valve attached directly to the stem. This cell is not meant to function as target in the future, so it only needs the pumping
Figure 2.6: Pictured here is the lower pressure adjustable cell. It is not intended to be used as a target, it consists of only a pumping chamber.
chamber side of the standard set up in Figure 1.1. With the intent of refilling the cell to incrementally higher pressures and noting different behavior, this second cell was initially filled with approximately 2.5 amg of \(^{3}\text{He}\). Like the first cell, this one contains both potassium and rubidium. However, this new cell contains only helium but no nitrogen. The typically small partial pressures of nitrogen likely have a minimal effect on the pressure broadening.

Even though the initial plan was to collect the following data sets over many resonances and at multiple pressures, they were only measured for potassium resonances due to time constraints. A few of the technical problems include: filling pre-existing adjustable cell that all the alkali had already oxidized in, breaking the glass of another cell, breaking the turbo vacuum pump which was being used in the high-vacuum filling setup. This last malfunction required completely abandoning the newly built ‘permanent’ filling setup.

To study the effect of polarization on absorption, a half-wave plate (HWP) was placed just after the linear polarizer in Figure 2.1. The angle of the HWP was adjusted to change the polarization of incident light on the cell.

Following this, the temperature trend was extended to 176 \(^{\circ}\text{C}\) and the linewidths/predicted densities were plotted based on potassium resonances. After realigning the optical setup, a power output of approximately 36 mW at 772 nm could be reached. This allowed me to search for similar or different power dependence trends between the higher pressure and lower pressure cells.
Chapter 3

Results

It might be stating the obvious, but before examining these plots of predicted density, some simple predictions should be noted. Density should be constant for a given cell and calculations of density via different resonances should yield equivalent densities.

Figures 3.1-3.4 show measurements that made at William & Mary in black, measurements made by a collaborator (Christopher Jantzi) from the University of Virginia in blue, and the predicted $\gamma$ values using Eqn. 1.2 in red. The line in red came from inputting a predicted density of 7 amg and finding the expected linewidths at a given temperature. The estimated density of 7 amg was given based by my advisor based on estimations from the initial cell filling.

Any parameters $a$ and $b$ shown with linear regressions are describing lines of the form $y = a + bx$. These parameters are presented next to their associated line of best fit (strictly for the WM data in black or UVA data in blue since the red line is not linear). The error bars on the data in black are plus or minus one standard deviation of the initially calculated linewidths for each set of absorption data. All plotted error bars for $\gamma$ or densities are statistical and based on how well the absorption data could be fit. For the uncertainty in density, I treated the equation for density (Eqn. 1.2) as perfectly accurate and converted the standard deviation of the linewidth (in GHz) to amagats. The data sent from UVA did not come with uncertainties, but each plotted
Figure 3.1: Rb D1 γ vs. temperature, using $\rho = 7 \text{ amg}$ and the empirical model from Eqn. 1.2 and laser power at 41.0 mW.

value is the average of five measurements taken at that temperature. Even though these data sets are measured on the same cell, there is a clear difference in both absolute predicted densities and trend with temperature between UVA and W&M data. The calculated predictions are in agreement with the W&M data for rubidium, but the potassium trends in Figures 3.3 and 3.4 show three distinct behaviors. This is the reason for only examining potassium when time constraints limited data collection on the new cell.

For the power dependencies, I first measured all four transitions at 137°C, and, once again, there was a clear difference between the potassium and rubidium transition linewidths with increasing power. The K D1 and K D2 linewidths decreased, with slopes of $-1.221 \pm 0.298$ and $-2.839 \pm 0.409$, respectively. On the other hand, neither rubidium transition showed significant variation with temperature. I collected data for the Rb D1 transition at a lower temperature of 126°C (Figure 3.5) and at
Figure 3.2: Rb D2 $\gamma$ vs. temperature, using $\rho = 7$ amg and the empirical model from Eqn. 1.2 and laser power at 20.0 mW.

Figure 3.3: K D1 $\gamma$ vs. temperature, using $\rho = 7$ amg and the empirical model from Eqn. 1.2 and laser power at 14.1 mW.
Figure 3.4: K D2 $\gamma$ vs. temperature, using $\rho = 7$ amg and the empirical model from Eqn. 1.2 and laser power at 14.1 mW.

a higher temperature of 150°C (Figure 3.7). Again, neither temperature showed a significant change to the linewidth-power relationship. My thought process here was that perhaps the number density of rubidium was too low at 137°C to see peculiar behavior, so I wanted to compare the trend at a lower and higher temperature. Based on data in Ref. [12], there is approximately a 10 times greater number density of rubidium and potassium at 150°C compared with 120°C.

Figures 3.11–3.15 show the data collected on the lower pressure cell, Namor. In Figures 3.13 and 3.13, the plots are using the calculated density based on Eqn. 1.2, the temperature of the cell, and the fit parameters from Kluttz et al. 2013, which are in Table 1.2 [2].

Data collected on the new cell (2.5 ± 0.1 amg of $^3$He) began with simple measurements of laser polarization dependence. We hypothesized there would be no dependence. As before, the $y$-axis has the linewidth measured in units of GHz, and angle of the half-wave plate is tracked in degrees. This data was taken with the
Figure 3.5: Rb D1 $\gamma$ vs. initial laser power at 126°C.

Figure 3.6: Rb D1 $\gamma$ vs. initial laser power at 137°C.
Figure 3.7: Rb D1 $\gamma$ vs. initial laser power at 150°C.

Figure 3.8: Rb D2 $\gamma$ vs. initial laser power at 137°C.
Figure 3.9: K D1 $\gamma$ vs. initial laser power.

Figure 3.10: K D2 $\gamma$ vs. initial laser power.
cell at a temperature of 169°C. The slope of linear fit is $0.0285 \pm 0.0277$ for the K D2 resonance and $-0.0156 \pm 0.0225$ for the K D1 resonance, so within one standard deviation both are virtually zero.

Potassium resonances exhibited unexplained power dependencies shown in Figures 3.9 and 3.10. Even if uncertainties on the linewidths are being underestimated, these two graphs show a significant difference between the trend of measured linewidths (of Florence) across temperatures. These trends are supported in later data collected on the Namor cell. This is presented in Figure 3.12.

The plots in Figures 3.12, 3.13, show both the raw data and calculated densities, as well as their lines of best fit. There is no specific reason that we expect to see a linear trend, but linear regressions allow for easy comparisons (especially when error bars are this large). Drawing attention to Figure 3.12, the slopes are $-0.33 \pm 0.10$ and $-0.51 \pm 0.20$. Just as variation in power on the target cell, Florence, showed variation, we again see decreasing slopes. After calculating the densities from the raw data, shown in Figure 3.13, the trend is significant enough to be concerning. The
Figure 3.12: K D1 and K D2 Linewidth vs. Power on Namor.

The slope is $-0.021 \pm 0.008$ for the K D1 resonance and $-0.024 \pm 0.009$ for the K D2 resonance.

Viewing Figure 3.13 again, the difference in vertical shifts is unexplained at this point; note the $y$-intercepts of $2.6 \pm 0.2$ and $1.9 \pm 0.1$. Whether or not a mechanism explaining this behavior exists, the unknown causes should be contained in the empirically derived constants that give a density prediction, assuming the model encompasses all necessary parameters for relating number density of $^3$He and alkali linewidth. Thus, the difference in vertical shift of predicted number density of He shows room for re-calibration of the $\alpha$, $\beta$, and $n$ parameters for the potassium resonances.

To reiterate, we should not expect to see a trend in the predicted densities with laser power because nothing is entering or leaving the cell while it is being measured. Because the linewidth varies with laser power, there is possibly an effect of laser power which went unaccounted in the initial empirical model—since that only used
Figure 3.13: Predicted densities based on K D1 and K D2 resonance linewidths at a variety of laser powers on Namor.

temperature and the type of linewidth to relate number density to $\gamma$. 
Figure 3.14: K D1 Linewidth vs. Temperature on Namor.

Figure 3.15: K D2 Linewidth vs. Temperature on Namor.
Figure 3.16: Predicted densities based on K D1 and K D2 linewidths vs. temperature on Namor.
Chapter 4

Conclusions

Based on the results in Figures 3.1–3.4, we note similar trends between the changes (really, the lack thereof) in linewidths of the Rb D1 and Rb D2 transitions with increasing temperature. Both of these trends are different from the experimental observations made at UVA on the same cell (shown in blue). These points were not given to me with any associated uncertainties, but each point is the average of five measurements at that temperature. The red line on the graphs comes from using Eqn. 1.2 with fixed the values of $\alpha$, $\beta$, and $n$ using the empirically determined values from Ref. [2]. Because the density is required to predict the linewidth, $\gamma$, I used the estimate of 7 amg to predict the observed linewidths. The K D1 and K D2 transitions in Figures 3.3 and 3.4 show very similar relations of linewidth increasing with temperature following a concave-up curve. Rb D1 and Rb D2 do not show this trend. In Figures 3.1 and 3.2 linewidth is relatively constant with increasing temperature, which is in disagreement with data collected from UVA on the exact same cell. So in every case of temperature dependency, the data from UVA disagrees with data I collected, and for both potassium transitions, my data did not validate the data from Eqn. 1.2 or the UVA data.

These initial discrepancies were what set this project in motion. The “smoking gun” for something unexplained was in Figures 3.9 and 3.10. When this project
began, I had no concept of “saturation limit” for absorption, so I began extending these ranges in the hopes of measuring trends over the widest ranges of laser power. After discussing the trends seen here with the AMO physicists at W&M, it remains unexplained, but better characterization of this trend will come from decreasing the uncertainties in measurements, especially at lower laser power.

In the spring semester, I planned to examine multiple pressures of $^3$He in a new cell, but due to time constraints and supply chain issues, I could only examine one pressure. Because the rubidium behavior was well explained by the empirical model, I also used my time more efficiently by only examining the less predictable potassium resonances.

The trends of K D1 and K D2 densities versus power (Figure 3.13) are both decreasing in a statistically significant trend. Another strange behavior is that the predicted densities are displaced from each other such that the D2 resonance (always being scanned at higher powers) is greater than the predicted density based on the D1 linewidth. If both linewidths are taken at the same temperature, we should at least hope for self-consistent calculated densities. Once again, regardless of the mechanism for these trends, the empirical $\alpha$, $\beta$ and $n$ parameters in Kluttz et al. (2013) should be corrected to account for these differences. If the model is not able to consistently calculate density using three parameters in this form, then perhaps an equation of a different form entirely should be tested and calibrated with old and new linewidths. I was unsure if certain implicit assumptions were made in using this equation, but no mention of a minimum or maximum laser power are mention in Ref. [2]. In certain instances, such as Figure 3.16, trends may or may not be statistically significant. In the future, parsing the differences in predicted densities (and with the raw data as well) will require improving the scanning precision and recording $\chi^2$ values to keep track of what trends are the most trustworthy. It might be most useful to first work
toward understanding what systematically causes the wild outliers that are nowhere near the neighboring data before describing the mechanism for a general trend.

Because the uncertainty in linewidth stems from how the absorption is fit to a Lorentzian, the parameters that lead to narrower lines often lead to inconsistent linewidths. A scan takes measurements every 0.035 nm, and if the linewidth is so narrow that the scan misses many of the points in the trough, there will be a greater uncertainty in the $\gamma$ value. This was only apparent in the low temperature measurements of the second cell (Namor) because it had less overall $^3$He and thus less pressure broadening at any given temperature. In the first cell, power dependence data was collected at $\sim 137^\circ$C. However, for the second cell, I collected power dependencies at 153-155$^\circ$C, and the polarization data was collected at 169$^\circ$C to make up for the pressure broadening being decreased in Namor.

In conclusion, the empirical model based on Eqn. 1.2 and Table 1.2 was validated for rubidium resonances across power and temperature variations, but potassium resonance linewidths show unexplained decreasing with laser power, and thus predicted densities are also varying. The predicted densities from K D1 and K D2 linewidths are both not self consistent, and—further distinguishing them from Rb resonances—also increase with temperature. The data presented in Figure 3.16 is still too obfuscated with uncertainty to determine if the rates of change are equal, but to the naked eye, there is some direct relationship between predicted density and temperature.
Appendix A

For Future Experimenters

It seems that even when all measurable parameters are held constant, such as the composition of the cell, laser power, and temperature, there is enormous inconsistency in repeat measurements. Only toward the end of my project did I learn some important information on fitting criteria. In talking with Christopher Jantzi about how he collected his data, extremely low laser powers (on the scale of tens of $\mu W$) were used. This means they were most likely well under the saturation intensity of rubidium or potassium. I suspect that further measurements done on the Namor would show that calculated densities from measurements at low powers and calculated densities from measurements at higher powers are not equivalent. The work done by Kluttz et al. most likely assumed a certain range of power would exclusively be used, and working in a range above or below that may ruin the validity of the model.

A few characteristics that have yet to be controlled/examined include 1. alkali-alkali interactions, 2. ‘scale limited’ power dependence, 3. beam intensity, and 4. cell surface inhomogeneities.

1. As the number density of alkali vapor increases, there is a temperature range at which Rb and K form diatomic molecules in the vapor phase. This will slow the normally exponential increase in number density of alkali vapor. Speaking
with Prof. Irina Novikova, an AMO physicist at W&M, this seemed to be insignificant within the operating temperatures.

2. I use the phrase “scale limited power dependence” to describe the idea that there might be a power dependence only near the saturation intensity of potassium. The saturation intensity of potassium vapor is 1.75 mW/cm$^2$ and this might lead to different absorption behavior when measuring above or below this limit [13]. Speaking again with Prof. Novikova, the saturation limit of rubidium is $\sim 6$ mW/cm$^2$, so these hypothetical “scale limited trends” should be apparent on the Rb linewidth vs. laser power plots if they exist. They are not, so it is likely some other difference between Rb and K is causing this.

3. I have been using measured power as a proxy for intensity, but the distinction might be important. The cross section of the beam is small, but nonzero and has not been recorded across trials. Using long focal length lenses would let us increase or decrease the intensity of photons on the cell.

4. The laser is passing through a curved surface, and even on a small scale, the surface is not perfectly smooth. Minor glass deformations from the hand blown glassware could be causing unpredictable changes to the linewidth, and thus affecting the density calculations. These should be considered when determining the uncertainties in future measurements.

As potassium and rubidium are both Group 1 elements, so we should expect very similar behavior for both of them. The average atomic masses of K and Rb are 39.0983(1) amu and 85.4678(3) amu, respectively, so one hypothesis I had is that the different velocities of nuclei could be impacting the temperature dependence [14].

I hope this section can serve to assist whomever takes up the mantle of this work in a year or more from now. Even though the model that Kelly Kluttz created was based
on a cell filled to similar pressures as the target cells being used at Jefferson Lab, the temperature range is not comparable. Kluttz’s measurements were taken at less than 150°C. I extended the range to higher temperatures because the predicted density seemed to vary, but it is not imperative that model is correct at all temperatures. If the density can be calculated accurately using a temperature range that isn’t near the working temperature of the target cell (≈ 220°C) then it should be noted that the model fails to predict outside of a certain range. By analogy, if a telescope lets you see objects in the distance, but it only works in the daytime and with clear weather, it does not mean the telescope is broken. The user simply knows its limitations, and recognizes that the objects on the horizon are the same whether or not the telescope is usable at that moment.

This project has been exploratory, and even though the conclusions show there are some properties being unaccounted for, there are two paths to take from here. It would be enlightening to continue examining what properties of potassium vapor lead to this strange power dependence, (without considering predicted densities of the $^3$He) but it might be more useful to re-calibrate the initial model and determine where it is able to predict valid densities.
References


