

Optical Pumping

Physics 111B: Advanced Experimentation Laboratory

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Abstract

In this experiment, we studied optical pumping in rubidium atoms to measure the resonant transitions in the Zeeman levels of the atomic electronic ground state. Using optically detected magnetic resonance (ODMR), we investigated the fine and hyperfine structures of the Rb-85 and Rb-87 isotopes and tested aspects of the Breit-Rabi equation. By applying a radio-frequency magnetic field and changing the external magnetic field with Helmholtz coils, we measured the resonance frequencies as a function of current and plotted the relationship. Our data exhibited the form of a linear model between resonance frequency and current, confirming theoretical predictions. From the slopes of the linear fits, we calculated the nuclear spins, and got the values of $185=2.509\pm0.0031$ for Rb85 and $187=1.509\pm0.0017$ for Rb87, closely matching both of the known values of 5/2 and 3/2. We also calculated the ambient magnetic field using two methods and concluded that the method based on linear fit intercepts provided more accurate and reliable results, yielding Bambient = 0.3165 ± 0.0054 for Rb85 and for Rb87 we got Bambient = 0.3137 ± 0.0044 Gauss.

Introduction

The main purpose of this experiment is to study optical pumping in rubidium (Rb) atoms and to measure the resonant transitions in the Zeeman levels of the atomic electronic ground state. Using the technique of optically detected magnetic resonance (ODMR), we will test certain aspects of the Breit-Rabi equation and examine the fine structure and the hyperfine structure of the Rb87 and Rb89 rubidium isotopes. Finally, we also want to measure the ambient magnetic field of our lab.

The goal of this experiment is to induce transitions between quantum states of rubidium and study these transitions using the methods of optical pumping and radio-frequency magnetic resonance. In our set up we perform spectroscopy and attempt to locate the resonant levels to see if it matches with theory.

The techniques that we used are:

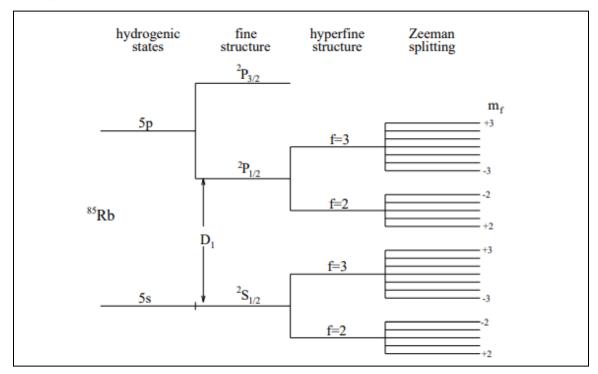
- Optical pumping for the selection of certain atomic polarization.
- Use of radio-frequency(rf) to jump between the different Zeeman sublevels.
- Lock-in detection of resonance frequencies, since detectors improve over time domain measurements.

The key concepts which we explore are:

- Hyperfine structure, which results from interactions between the nuclear and electron spins.
- Zeeman effect, where energy levels split in the presence of a magnetic field.
- Rabi oscillations, which occur during resonant transitions induced by the applied radio-frequency field.

This technique of optical pumping was invented by Alfred Kastler in the 1950s and won him the nobel prize in 1966. Many high level physics laboratory courses today require this lab technique to be taught because of the particularly interesting physics used which will be practiced during the procedure of it. This experiment we went through in particular combines the theoretical quantum mechanical predictions with experimental spectroscopy, providing an insight for us into the structure of atomic energy levels and magnetic interactions in the presence of external fields.

Background Research & Theory



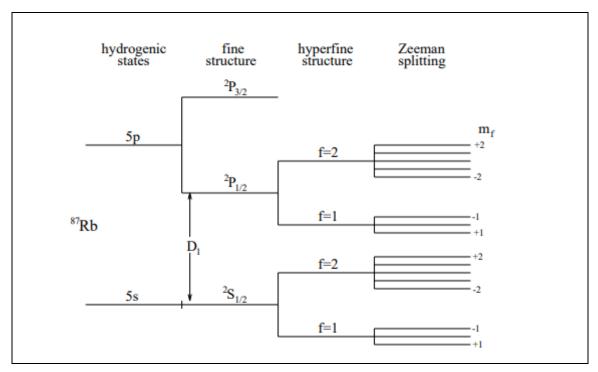


Figure 1 & 1.1 (Carnegie Mellon University 33.340 Modern Physics Laboratory)
*Note that the level of these splittings are not to scale

To fully understand the experiment, it is very important to understand the full atomic structure of Rubidium 85 & 87. In this part, I will now go over Figure 1 in detail, which summarizes it pretty well.

A. Basic Atomic Structure

Rubidium is an alkali metal located in the first group of the periodic table, characterized by having a single unpaired, optically-active valence electron. When this electron is excited to its first excited state, it behaves similarly to a hydrogen atom and we will refer to this as the "hydrogenic state." This state occurs when the electron transitions from the 5s orbital to the 5p orbital in Rubidium. It is important to note that since there is one valence electron its spin will be precisely ½ and thus the spin of Rb85 is 5/2 and Rb87 is 3/2.

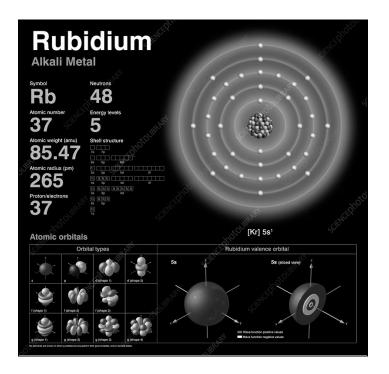


Figure 2 (CARLOS CLARIVAN / SCIENCE PHOTO LIBRARY) https://www.sciencephoto.com/media/553899/view/rubidium-atomic-structure

B. First order correction

The first-order correction to Rubidium's atomic structure arises from fine energy splitting, which is caused by relativistic effects that couple the electron's orbital angular momentum with its spin. It is important to note that these are two separate phenomena that are considered in the correction. This fine energy splitting is relatively minor compared to the Bohr energy levels, smaller by a factor of α^2 , where $\alpha \approx 1/137.036$.

The hamiltonian including the fine structure correction will take on the form

$$H \propto S \rightarrow L$$

Note that in this form, The hamiltonian will no longer commute with **L** and **S**, and thus the spin and angular momentum are no longer conserved separately (as we learned in quantum mechanics). However the hamiltonian will commute with L^2 , S^2 and the total angular momentum J where J = L + S.

To transform the Hamiltonian we will consider

$$J^2 = (L + S) \cdot (L + S)$$

and thus the result is:

$$H \propto L \cdot S = \frac{1}{2} (J^2 - L^2 - S^2)$$

The resulting eigenvalues after this transformation are in the form:

$$\frac{\hbar^2}{2} \left[j \left(j+1 \right) - \ell \left(\ell+1 \right) - s \left(s+1 \right) \right]$$

C. Second order correction

The second order correction is called the *hyperfine structure* correction as we see in the f orbitals of <u>Figure 1</u>, which results from electromagnetic multipole interaction between the nucleus and electron. In this case, the electronic angular momentum and the nuclear spin couple together once again to cause another splitting which is also on the order of about α^2

D. Zeeman Splitting

The zeeman effect is caused by the presence of an external magnetic field and will result in the splitting of spectral lines which would otherwise be degenerate (again we can see this in Figure 1). In the alkali-atom ground level the energies of the Zeeman effect can be calculated using the *Breit-Rabi* formula which in the low-field case will be:

$$\frac{\nu}{B_{\text{ext}}} = \frac{2.799}{2I + 1} \frac{\text{MHz}}{\text{G}}$$

A derivation of the Breit-Rabi formula can be found at 'Introduction to Elementary Particles' by Griffiths in section 5.5, we won't go over it here because it isn't necessary while being mathematically intensive

E. Optical pumping

This section is also really important to fully understand because it is the basis of this experiment. Again, our goal here is that we want to see the transition energies/lines in the electronic ground states of rubidium atoms.

However, we first have to get an initial spin polarized state such that there are a lot more atoms in the initial state compared to the final state. We can use a technique called optical pumping to generate this spin polarization and in this experiment we do so by driving all atoms through the D1 transition using infrared light. If we send the light in through the z direction then the transition will cause the electron to excite increasing the magnetic quantum number, mf by one. When this excited state decays the electron will go through spontaneous emission and mf

can change by +1.0, or -1. However after many cycles we see that most atoms will end up 'pumped' to the highest mf value.

An important point is that once the gas reaches this pumped state, the atoms no longer absorb light, allowing it to pass through the gas without resistance and reach the photodetector. This state is referred to as the "dark state." This concept is crucial because we will use it to detect the frequency of magnetic transitions in the vapor. If the atoms are not exposed to a resonant radio-frequency magnetic field, the vapor remains in its dark state, and the photodetector reads a high light level. Otherwise, a low light level is recorded. This technique is known as optically detected magnetic resonance (ODMR).

Experimental design

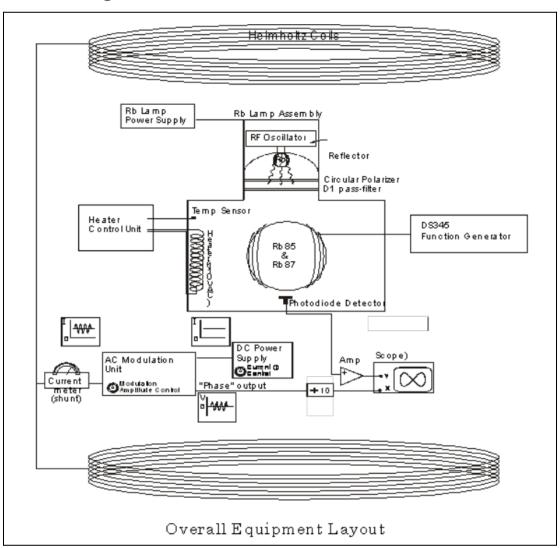


Figure 3 (Lab manual)

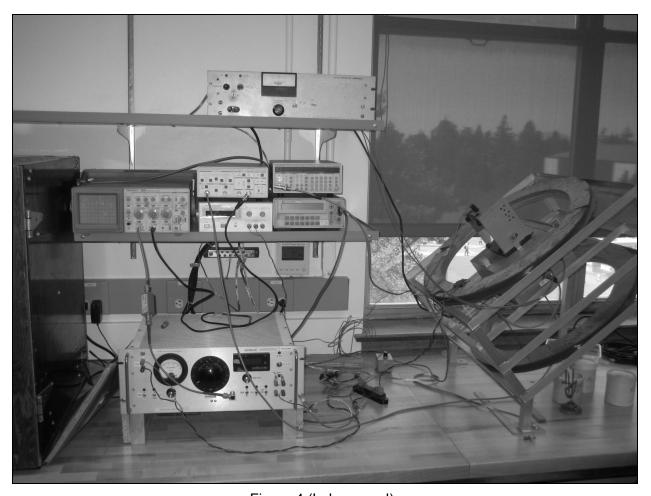


Figure 4 (Lab manual)

I will now go over figure 4 because it is crucial for us to understand the set up for us to understand the experiment:

- 1. **Heater Control Unit:** This device regulates the temperature of the bulb. It is necessary because, at room temperature, the rubidium in the atomic vapor cell (a spherical glass bulb) remains on the surface. By heating the bulb, some of the rubidium is released into the buffer gas. Importantly, the higher the temperature, the denser the rubidium vapor cloud becomes. It's also worth noting that when the heater is active, it generates its own magnetic field, which can interfere with other magnetic fields. Therefore, no measurements should be taken while the heater is on.
- 2. **Rubidium lamp:** The rubidium lamp will serve as the optical pumping light in this experiment, which is appropriate because the energy required to excite the rubidium vapor matches the energy emitted by the lamp. It's important to note that the light from the lamp will be circularly polarized and passed through a band-pass filter, allowing only D1 transitions to occur.

- 3. **Radio-frequency coil:** The RF frequency can be modulated using these coils, which are connected in series and positioned around the vapor cell. A function generator drives the modulation, producing a radio-frequency magnetic field that is perpendicular to both the optical axis and the direction of a strong external magnetic field.
- 4. **Light-proof box:** All of these components are housed in a lightproof (and also thermally insulated) box because the optical signal from the rubidium lamp is relatively weak, and we want to prevent any interference from ambient light in the room.
- 5. **Helmholtz coils:** These coils produce a near-constant axial magnetic field, given by the formula:

$$0.9 \times 10^{-2} \, \frac{Ni}{a} \times \frac{\text{G m}}{\text{A}}$$

With N being the number of turns, *i* being the current and *a* being the radius.

Raw Data

Voltage across the shunt resistor: 10.1950 mV

For I = 1A

Initial Frequency peak from sweep for Rb85 : 2.2Mhz Initial Frequency peak from sweep for Rb87 : 3.3Mhz

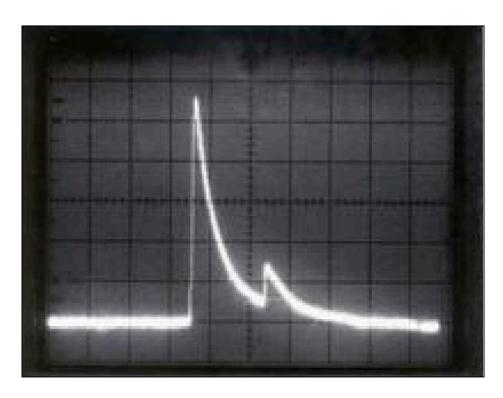


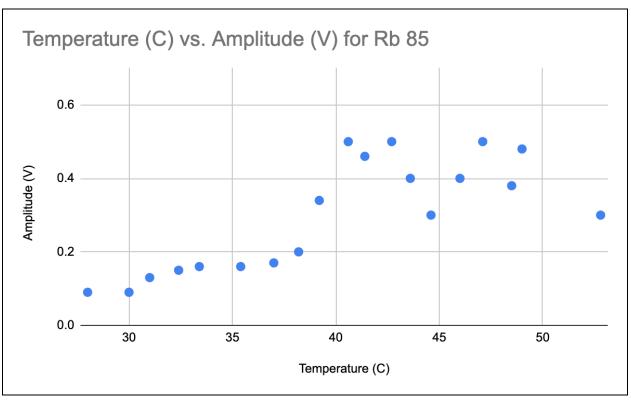
Figure 5: Amplified photodiode signal with large span frequency modulation from lab manual *Note* that this is not a picture we took but we had a similar result

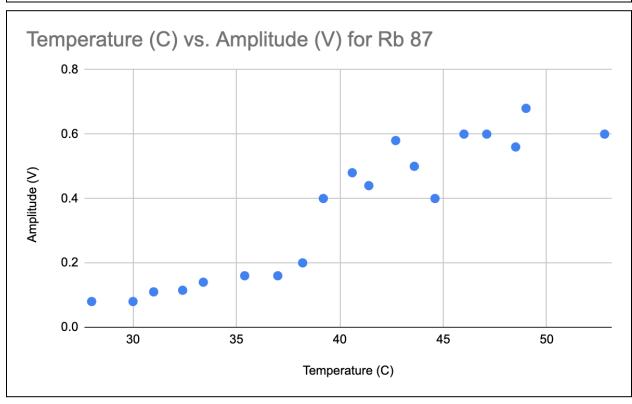
Temperature (C)	Unit/div	Rb- 85 (#div)	Rb-87 (#div)
28	0.05	1.8	1.6
30	0.05	1.8	1.6
31	0.05	2.6	2.2
32.4	0.05	3	2.3
33.4	0.05	3.2	2.8
35.4	0.05	3.2	3.2
37	0.05	3.4	3.2
38.2	0.05	4	4
39.2	0.1	3.4	4
40.6	0.1	5	4.8
41.4	0.1	4.6	4.4
42.7	0.1	5	5.8
43.6	0.1	4	5
44.6	0.1	3	4
46	0.1	4	6
47.1	0.1	5	6
48.5	0.1	3.8	5.6
49	0.1	4.8	6.8
52.8	0.1	3	6

Amplitude here is calculated by multiplying units per div with # of div

Amplitude 85 (V)	Amplitude 87 (V)	Uncertainty (V)
0.09	0.08	0.02
0.09	0.08	0.02
0.13	0.11	0.02
0.15	0.115	0.02

0.16	0.14	0.02
0.16	0.16	0.02
0.17	0.16	0.02
0.2	0.2	0.02
0.34	0.4	0.04
0.5	0.48	0.04
0.46	0.44	0.04
0.5	0.58	0.04
0.4	0.5	0.04
0.3	0.4	0.04
0.4	0.6	0.04
0.5	0.6	0.04
0.38	0.56	0.04
0.48	0.68	0.04
0.3	0.6	0.04





Resonance frequency vs. Current data scan for each measurement the uncertainty is +/- 10kHz

Current (A)	Rb 85 (Khz)	Rb 87 (Khz)
-2	3950	5920
-1.9	3750	5620
-1.8	3540	5300
-1.7	3340	5010
-1.6	3130	4690
-1.5	2930	4390
-1.4	2730	4090
-1.3	2520	3770
-1.2	2330	3490
-1.1	2120	3170
-1	1910	2870
-0.9	1710	2570
-0.8	1510	2250
-0.7	1320	1970
-0.6	1100	1650
-0.5	890	1340
-0.4	690	1030
-0.3	500	750
-0.2	310	460
0.05	300	410
0.1	370	550
0.15	480	710
0.2	570	860
0.25	670	1000
0.3	780	1170
0.4	990	1470
0.5	1180	1770
0.6	1390	2080

0.7	1580	2380
0.8	1790	2680
0.9	2000	2980
1	2220	3300
1.1	2400	3610
1.2	2610	3910
1.3	2820	4230
1.4	3020	4530
1.5	3230	4840
1.6	3430	5140
1.7	3630	5440
1.8	3850	5760
1.9	4050	6070
2	4250	6360

Zero current resonance frequency for 85	212kHz
Zero current resonance frequency for 87	363kHz

Total pumping time measured: 60 ms 1/e characteristic time => 22.07 ms

Analysis

1.

In our raw data, the plots resemble absolute value signs since frequency cannot be negative. However, we found it useful to extend the positive values into the negative range when plotting the line of best fit for our data. It's important to note that while the Helmholtz coil generates a magnetic field along a single axis, the ambient magnetic field may be oriented slightly differently. We can account for this by checking for systematic errors in our experiment. If the ambient magnetic field is significantly misaligned, we would observe a consistent shift in frequency, either upward or downward, across all data points.

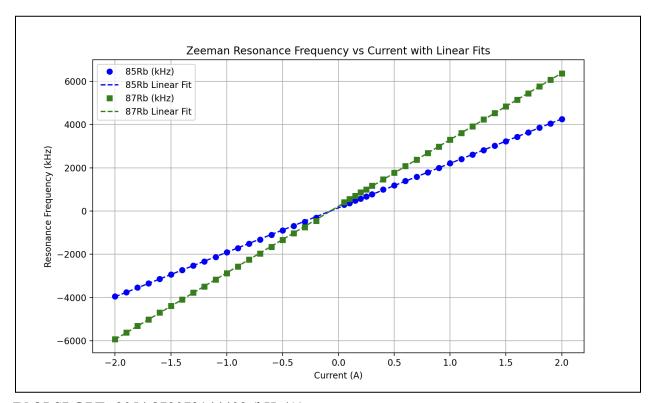
The trendline follows the form (from lab manual):

$$v = \left(\frac{2.79}{2I+1} \frac{0.9N}{a}\right) \cdot i + \left(\frac{2.799}{2I+1}\right) B_e$$

Where v = frequency, I = nuclear spin, N = Number of turns, a = radius of Helmholtz coil, i = current and Be = Ambient field

I took our data and coded it up in python to get the figures and values, the link to find the raw code is:

https://docs.google.com/document/d/10jxBpFVwFQZM-pcfTGomM1g4v_soQ1E8b0xSJtmwUsQ/edit?usp=sharing



Rb85 SLOPE: 2054.879073144409 (kHz/A) **Rb85 INTERCEPT:** 147.61467880406136 (kHz) **Rb87 SLOPE:** 3078.0966565544736 (kHz/A) **Rb87 INTERCEPT:** 219.4534961641677 (kHz) **Rb85: R^2** = 0.99996 || **Rb87: R^2** = 0.99997

Slope Error 85: 2.13711876860446 (kHz/A), **Slope Error 87:** 2.6607042276688255 (kHz/A) **Intercept Error 85:** 2.5000744872964 (kHz), **Intercept Error 87:** 3.11258263020185 (kHz)

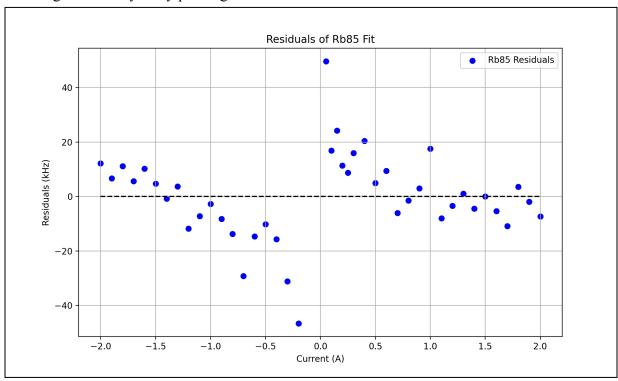
2. *Interpretation of graph:*

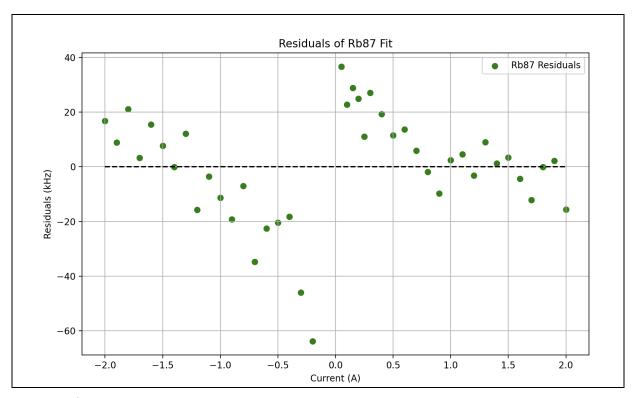
Now from the plot above, it looks like the data is a perfect fit to a linear function but how would we address the goodness of the fit? One method is using the chi-squared test $\chi 2$. Why would we use this method rather than just using the R^2 values we calculated and saying it's just an almost

perfect fit with (**Rb85**: $\mathbf{R}^2 = 0.99996 \parallel \mathbf{Rb87}$: $\mathbf{R}^2 = 0.99997$)? In short there are two main reasons why I think it would be a better choice:

- 1. Chi-squared incorporates measurement uncertainties, allowing us to weigh data points appropriately.
- 2. R^2 does not account for uncertainties and can be misleading, especially when data points have different errors

Let's begin our analysis by plotting the residuals:





Interpretation:

Now instead of a perfect linear fit, we can see from these residual plots that as we get closer and closer to 0 current our error grows. Why is this the case in this experiment? It probably is due to the fact that a low current with a misaligned ambient vector field has a greater effect on the end, resulting in bigger errors.

*Note that now that we have errors, our linear fit should account for those errors as well. There are two ways to do this, one being that all data points have the same weight and our linear regression formula is already calculated above. Another potential way is that the size of the error will determine the weight of the data point, meaning some outlier with a big error should not sway the fit so that it's just inaccurate.

Here in python I will compute a weighted and compare it to unweighted fit to see if they both agree:

** The code is in the same link above in part 1 **

Weighted Fit Rb85 Slope: $2054.8790731391173 \pm 1.3191016814870824 \text{ kHz/A}$ Vs. Unweighted $2054.879073144409 \pm 2.13711876860446 \text{ (kHz/A)}$

Weighted Fit Rb85 Intercept: $147.61467885319027 \pm 1.5431301628018486$ kHz Vs. Unweighted $147.61467880406136 \pm 2.5000744872964$ (kHz)

Weighted Fit Rb87 Slope: $3078.0966565556773 \pm 1.3191016921702219 \text{ kHz/A}$ Vs. Unweighted $3078.0966565544736 \pm 2.6607042276688255 \text{ (kHz)}$

Weighted Fit Rb87 Intercept: $219.45349615768316 \pm 1.5431302204262596$ kHz Vs. Unweighted $219.4534961641677 \pm 3.11258263020185$ (kHz)

Reduced Chi-squared Rb85: 2.6248274729907126 Reduced Chi-squared Rb87: 4.068521782991443

It appears that our fits both agree with each other very well (virtually identical).

However there is one big glaring problem here and it's that our chi-squared values are both much greater than 1. The interpretation I think is correct here is that we underestimated our uncertainties by a lot. In the lab to measure it we skewed the figure 8 until it was slightly off and that's why our data points were always $\pm 10k$ hz for the frequency. However, I think if I were to go back I would skew it much more than I did and repeat the same process for each one, ending up with a different range of frequency errors.

3.

Knowing the formula for our trendline we can take the ratios of our slopes to get the ratio of spins between Rb85 and Rb87:

$$v = \left(\frac{2.79}{2I+1} \frac{0.9N}{a}\right) \cdot i + \left(\frac{2.799}{2I+1}\right) B_e$$

Ratio = $3078.1 / 2054.9 = 1.497944 \pm 0.000709$ (Calculated from Error propagation) which is just about 1.5 (what we expect). Also note that with the knowledge that these two isotopes should have half integer spin it is not enough information to figure out the isotopes: there are infinitely many combinations of odd numbers we could use to get us a number close to the slope ratio we observed (a spin 1111/2 and 739/2 will get roughly 1.5 when divided). However, these spins in nature are impossible, so it is natural to deduce that the most likely spins of these two isotopes are 5/2 for Rb85 and 3/2 for Rb87.

The next method is to determine the nuclear spins directly by doing a bit of math, using the Breit-Rabi formula along with our slope value we got we can plug in the numbers and do the calculation directly:

$$\frac{\nu}{B_{\text{ext}}} = \frac{2.799}{2I + 1} \frac{\text{MHz}}{\text{G}}$$

So first we calculate B ext/I given the Helmholtz coil parameters:

$$B_{ext} = (0.009N/a) I =$$
 note (135 coils and coil radius is 27.5 cm)

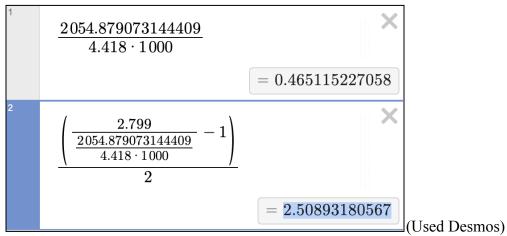
$$B_{ext}/I = 0.009 \times 135/0.275 \approx 4.418 \text{ G/A}$$

Our slope is v/I then knowing this we have:

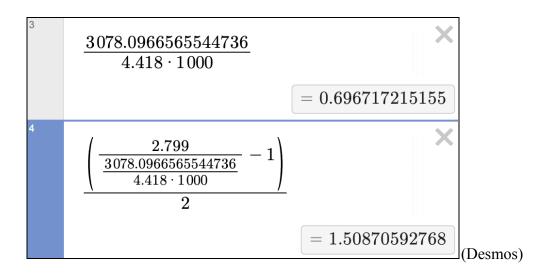
$$v/B_{ext} = slope/B_{ext}/I$$

Which is the left half of the Breit-Rabi formula, plugging our slopes in and solving for I:

For Rubidium 85 we get:



For Rubidium 87 we get:



Finally to get the uncertainties I will propagate the errors from the slope (I just plugged it into wolfram alpha):

Which means in our second method we get:

$$I_{85} = 2.509 \pm 0.0031$$

 $I_{87} = 1.509 \pm 0.0017$

The experimental values we got are very close to the true values of the spins, although the errors after propagation do not fit this might be due to the statistical/systematic errors we do not account for: Statistically, the uncertainties in linear fits of the data may not fully capture random errors in the measurements with the equipment we have. The systematic errors here can arise from misalignment of the magnetic field, inaccuracies in coil parameters, and unaccounted residual magnetic fields. Thus, with all this in mind it is very well possible we underestimated our errors here. Despite this I think these values are pretty accurate and consistent given the circumstances since within less than 1% of the known nuclear spins for both Rb85 and Rb87.

4.

Again equation 6 is:

$$\frac{\nu}{B_{\text{ext}}} = \frac{2.799}{2I + 1} \frac{\text{MHz}}{\text{G}}$$

So solving for B ext we get:

$$B_{ext} = v \frac{2I+1}{2.799} Gauss$$

Lets pick positive I coil = 1.0 A and negative I coil = -1.0 A

Now we can solve for B ext for Rb85 for both positive and negative current:

Plugging it all in for Rb85:

$$B_{ext}^{+} = 4.721 \, Gauss$$

$$B_{ext}^{-} = -4.089 \, Gauss$$

(We can do the same for Rb87 and it gives similar values)

Now we also know from before that:

$$\pm B_{ext} = (0.009N/a) I => (135 \text{ coils and coil radius is } 27.5 \text{ cm})$$

Which gives:

$$B_{ext} = 4.4182 \, Gauss$$

Now comparing the two:

We have

$$B_{ext}^{+} = 4.721 \, Gauss$$

$$B_{ext}^{-} = -4.089 \, Gauss$$

VS.

$$B_{ext} = 4.4182 \, Gauss$$

Which is pretty solid considering the coil calculator to be directly in the middle of the positive and negative coil calculations. The accuracy of this can be improved by considering the errors I discussed in the earlier question, along with the accuracy of our knowledge of the Bohr magneton μB , possible errors from neglecting the nuclear Zeeman energy, the accuracy of describing the Helmholtz coils as coils of precisely defined and known radius and position, the accuracy of the shunt resistor and other parts of the measurement apparatus (Lab manual).

5.

There are two ways to calculate the ambient field, let's begin by going through the first way: Equation 6 is again, :

$$\frac{\nu}{B_{\text{ext}}} = \frac{2.799}{2I + 1} \frac{\text{MHz}}{\text{G}}$$

Here we can plug in the known nuclear spins into the formula and plug in the intercept frequency (where current is 0) to isolate the ambient field B_ext here.

Using those values in part 1 we get:

B Field Rb85 = 0.3165 ± 0.0054 Gauss B Field Rb87 = 0.3137 ± 0.0044 Gauss

Which both match up pretty well!

Another way is to take the zero current resonance frequency and solve for the field since it should be a somewhat direct measurement of the ambient field. However the closest we got to zero was at I = 0.05 A which is not exactly zero but we can still see what it yields us.

Plugging in the frequencies at current = 0.05 A, and solving for B ext into the formula below:

$$\frac{\nu}{B_{\text{ext}}} = \frac{2.799}{2I + 1} \frac{\text{MHz}}{\text{G}}$$

We get:

B Field Rb85 = 0.643 ± 0.0214 Gauss B Field Rb87 = 0.3651 ± 0.0143 Gauss

Which do not agree with the other method or even each other. Note that we weren't able to get a zero current measurement because the figures were too blurry to make out, which is why we took it at 0.05 which was the closest to zero we could possibly get here. Also note that earlier I discussed the underestimation of our errors in the intercept when calculating the chi-squared, those underestimations would definitely come up here as well in the errors since I got those values by propagating the $\pm 10k$ frequency range.

Why are the values different and which values should we trust here from method 1 vs method 2? The reason why the answer here is method 1 is because of a few big reasons:

- 1. Statistical Reliability Through Linear Fit: Here we use the predicted intercept on multiple points rather than a single zero field measurement which is known to not be as accurate based on the residual plot. The linear fit averages out random errors and minimizes the impact of any single anomalous measurement which is why it should be more accurate.
- 2. Smaller and Better-Characterized Uncertainties: The uncertainties in the intercepts obtained from method 1 are smaller due to the averaging effect of multiple data points and these uncertainties are better characterized because they are derived from the fit, which accounts for the spread and variance in the data.
- 3. Consistency between isotopes: The values of B_ambient obtained from method 1 for both Rb-85 and Rb-87 are in close agreement, which reinforces the reliability of the measurements and suggests that systematic errors are minimal.

Which is why here we end up deciding with B Field Rb85 = 0.3165 ± 0.0054 Gauss and B Field Rb87 = 0.3137 ± 0.0044 Gauss as our values

6.

There is a resonance at zero field due to the coil not being perfectly aligned with the ambient magnetic field. While our data shows that the Helmholtz coil is very closely aligned, it's not perfect, leaving a small residual field. As a result, a resonance can still be detected even at zero field

7.

From stat mech we know that the characteristic pumping time goes by a factor 1/e: This is because as more and more atoms become pumped, the population of un-pumped atoms will drop, lower and lower, meaning it will take more time to get the few left over. This fact remains the same for de-pumping the atoms. We also need to know that compared to pumping, de-pumping as a process will take much longer and be much slower. Pumping is an active process in which we actively pump the atoms into the dark state while pumping depends on the 'slow' relaxation time for the gas.

Conclusion

We were able to successfully utilize optical pumping and rf magnetic resonance techniques to induce and study transitions between quantum states of rubidium atoms. Our measurements confirmed the linear dependence of the Zeeman resonance frequencies on the external magnetic field, as predicted by the Breit-Rabi equation. The nuclear spins determined from our experimental data were $185 = 2.509 \pm 0.0031$ and $187 = 1.509 \pm 0.0017$, which again, are in agreement with the established values of 5/2 and 3/2.

When determining the ambient magnetic field, we compared two possible methods: one using the intercepts from linear fits of our resonance frequency data, and another using direct measurements at a current at zero. The first method provided consistent results with smaller uncertainties, we got: B Field Rb85 = 0.3165 ± 0.0054 Gauss and B Field Rb87 = 0.3137 ± 0.0044 Gauss. The second method resulted in values that completely failed to agree with each other or the known value of Earth's field at that point (between 0.2 and 0.4 Gauss per GSI)

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- 1. http://experimentationlab.berkeley.edu/sites/default/files/writeups/OPT.pdf
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