Measurement of hyperfine constants and the isotope shift of rubidium $5P_{1/2}$ excited-state using saturated absorption spectroscopy

P M Rupasinghe, Fiona Wee, Thomas Bullock and Jiaxing Liu

Department of Physics, State University of New York at Oswego, Oswego, NY 13126, United States of America

E-mail: priyanka.rupasinghe@oswego.edu

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Abstract

The Saturated Absorption Spectroscopy (SAS) was performed to measure the hyperfine constants of rubidium $5P_{1/2}$ excited state using an external-cavity diode laser (ECDL) operating at 795 nm. We report our measurements for the magnetic dipole coupling constants $120.79^{(29)}$ and $407.75^{(57)}$ for $^{85}$Rb and $^{87}$Rb respectively. Our measurement of the $^{85}$Rb hyperfine constant shows a good agreement with previous results and our measurement of the $^{87}$Rb hyperfine constant resolves the discrepancy of results that exists in the literature.

1. Introduction

Precise measurements of hyperfine structure and isotope shifts of multi-electron atomic systems provide valuable insight into the nuclear properties as well as our understanding of the electronic wavefunctions near the nucleus. Therefore, they play an important role in the development of new atomic theories that may rely on various approximation techniques. For example, two different theoretical methods estimate $^{85}$Rb magnetic-dipole coupling constants for the $5P_{1/2}$ state as 69.8 MHz and 120.4 MHz while the latter is close to the experimental results [1].

Rubidium can be considered as one of the well-studied alkali systems with a wide range of applications in frequency stabilization techniques, laser cooling and trapping, atomic clocks, development of atomic sensors, and quantum information processing [2–5]. A natural sample of rubidium contains two isotopes $^{85}$Rb and $^{87}$Rb with 72% and 28% abundances respectively. This system is an attractive candidate for experimental study using laser spectroscopy techniques primarily due to convenience in the accessibility of D1 ($5S_{1/2} \rightarrow 5P_{1/2}$) and D2 ($5S_{1/2} \rightarrow 5P_{3/2}$) excitation transitions using commonly available laser diodes operating at 795 nm and 780 nm as well as the availability of sufficient vapor pressure at or near the room temperature. For such an atomic system the energy of the hyperfine level measured from the center-of-mass energy can be written as,

$$W(F) = \frac{1}{2}AK + B\frac{(3/2)K(K + 1) - 2I(I + 1)J(J + 1)}{2I(I - 1)2J(J - 1)}$$

(1)

where $K = F(F + 1) - I(I + 1) - J(J + 1)$, $A$ and $B$ are the magnetic-dipole and electric-quadrupole hyperfine coupling constants, $I$ is the nuclear spin, $J$ is the total electronic angular momentum, and $F$ is the total angular momentum of the atom. The second term only contributes when $J > \frac{1}{2}$.

Hyperfine constants of rubidium $5P_{1/2}$ excited state have been measured previously with various techniques [6–11]. Here, we present another measurement of hyperfine coupling constants and the isotope shift of rubidium $5P_{1/2}$ state using a distinct approach from the previous techniques.

2. Experimental details

2.1. Optical system and experimental layout

We excited ground-state rubidium using 795 nm laser light according to the scheme shown in figure 1.
The external-cavity diode laser (ECDL) used in this experiment is a homemade system based on an existing design [12, 13] with a 10mW laser diode. An ultra-low expansion Fabry–Perot cavity was used to create a proper frequency axis for our spectra. As shown in figure 2 two low-power parallel beams with each having power of \(\sim 25\ \mu\text{W}\) transmitted through the Rb vapor cell held at 47 \(^\circ\text{C}\) are directed to a differential photodiode (Thorlabs 450A). While the laser is scanning through the relevant transition each beam produces an identical Doppler-broadened direct-absorption signal at individual photodiodes. This corresponds to a nearly zero signal at the detector output (differential signal). However, when a relatively strong beam (pump-beam) of power \(\sim 140\ \mu\text{W}\) is present, a large number of atoms absorb light from the strong beam. As a result, the direct-absorption signal corresponds to the relevant probe-beam shows absorption dips widely known as (hole burning) at certain frequencies.

The hole burning can occur at hyperfine transition frequencies as well as exactly in the middle of the hyperfine transition frequencies when the Doppler shift can compensate for the energy splitting between the two levels (cross-over resonances). These cross-over absorption features show up due to the fact that there are two velocity classes of atoms exist in the cell such that the pump beam is in resonance with one hyperfine transition, and the probe beam is in resonance with the other [14].

When the pump (strong beam) is present the output signal (differential signal) of the differential photodetector shows sharp peaks only corresponding to hyperfine as well as cross-over resonances as shown in figures 3 and 6.

2.2. Data acquisition system
After tuning the laser to near transition frequency we scan the laser by applying a triangular voltage signal such that we collect up-scans and down-scans alternatively. During each scan period our Labview program samples about 1000 data points per channel in order to create spectra using National Instruments PCI-6221 Multifunction I/O data acquisition card with BNC2110-DAQ device. We record hundreds of spectra under various experimental conditions (laser scan direction, speed, excitation transition, etc) in fully automated
fashion with 20–30 scans in each run. Specifically, for each scan we collect the laser scan voltage ramp (up and down), corresponding Fabry–Perot signal, and the vapor cell signal (the saturated absorption spectrum).

3. Data and analysis

3.1. Creating a linearized frequency axis

The data analysis procedure starts with Fabry–Perot spectrum. We fit Fabry-Pérot signal to modified Airy function as a function of normalized data point \( x \). The normalization of data point numbers is created using the conversion \( x = \frac{X - N/2}{N/2} \), where \( X \) is the data point \( #(0, 1, 2, \ldots, 999) \) and \( N \) is the highest data point (999). Specifically, we fit our Fabry–Perot spectra to the model,

\[
\mathcal{A}(x) = \frac{b_0 + b_1 x}{1 + F \sin^2 \left[ \frac{\pi}{\Delta \nu} f(x) \right]}
\]

where, \( b_0 \) and \( b_1 \) are constants, \( F \) is the coefficient of finesse, \( \Delta \nu \) is the free-spectral range (FSR) in MHz (500 MHz in this case), and \( f(x) = a_0 + a_1 x + a_2 x^2 + \ldots \) is a polynomial. This fit model allows us not only to transform data point numbers to a frequency axis but also to remove any nonlinearities associated with the laser scan due to the hysteresis of the piezo-electric device (PZT) that used to change the angle of the diffraction grating of the ECDL. Such a fit is shown in figure 3, notice the slightly increasing separations between peaks in the spectrum which is attributed to a slight nonlinearity of the laser scan.

During the fit, all the parameters are floated except the free-spectral range which is kept fixed at its value (500 MHz in our case). Initial conditions for parameters \( a_0 \) and \( a_1 \) were set by mapping peak positions \( x_i \) where, \( i \) is the peak number, to relative frequency values \( f(x_i) \) separated by FSR and fitting \( f(x) \) versus \( x_i \) to a linear function \( a_0 + a_1 x \). Higher order coefficients \( a_2, a_3, \ldots \) were set to zero. After the first fit, all the initial conditions have been updated with new fit parameters and refit for improved results (Bootstrapping). The final fit parameters \( a_0, a_1, \ldots \) defines our new frequency axis \( f(x) \). Figure 4 shows Fabry–Perot signal plotted against new frequency axis \( f(x) \) with a 4th order polynomial. Unlike the figure 3, in figure 4 equal spacing between peaks indicates that the non-linearity associated with laser scans has been removed. There was no statistically significant change in results was observed by using higher order polynomials which further assures that the 4th order polynomial used in this analysis was sufficient to completely remove the nonlinearity.
been used to create linearized frequency axes for atomic spectra produced by ECDL scans on many occasions in recent years \cite{15–17} with a different spectroscopy technique.

Next, we plot the saturated absorption data against the new frequency axis \( f(x) \) and fit them to a model function containing a sum of Lorentzian line shapes. As the saturated absorption spectra are Doppler-free use of the Voigt profile was not necessary. A sample fit of spectra containing both isotopes is shown in figure 6.

3.2. Single isotope spectra

Hyperfine splitting values for \(^{85}\text{Rb}\) and \(^{87}\text{Rb}\) spectra are derived in three independent ways, \( \nu_3-\nu_1 \) (direct), \( 2(\nu_3-\nu_2) \) (indirect), and \( 2(\nu_3-\nu_2) \) (indirect), where, \( \nu_1, \nu_2, \) and \( \nu_3 \) are the first, second, and third peak positions respectively. The indirect measurements use the fact that 2nd peak (cross-over) is located right in the middle of the two hyperfine peaks. These direct and indirect values are a good systematic error check for our frequency axis. For each single isotope spectrum, direct and indirect methods yielded similar results reassuring that our frequency axis linearization is done correctly. We extract several hundred hyperfine splitting values by fitting our single isotope spectra in this manner.

3.3. Dual isotope spectra

The dual isotope spectrum shown in figure 6 can be used to determine hyperfine splittings for individual isotopes as well as the transition isotope shift (TIS) between them. Using this spectrum individual hyperfine splittings are extracted using a similar analysis done for the single isotope spectra. According to the level diagram described in the figure 1, transition isotope shift can be determined by the following equation.
Table 1. Hyperfine splittings and transition isotope shift measurements.

| Hyperfine splitting | $\Delta V^{85}$ | $\Delta V^{87}$ | TIS |
|---------------------|-----------------|-----------------|-----|
| Final result (MHz)  | 362.37          | 815.49          | 77.56 |
| Stat. error (MHz)   | 0.39            | 0.79            | 0.51 |
| Systematic error source: |
| Scan direction      | 0.67            | 0.73            | 0.85 |
| Scan speed          | 0.10            | 0.19            | 0.24 |
| Excitation scheme   | 0.36            | —               | —   |
| Scan linearization  | 0.02            | 0.24            | 0.16 |
| Combined error (MHz)| 0.86            | 1.13            | 1.03 |

\[
TIS = \left( \frac{5}{12} \Delta V^{85}_g + \frac{7}{12} \Delta V^{85}_e \right) - \left( \frac{3}{8} \Delta V^{87}_g + \frac{5}{8} \Delta V^{87}_e \right) + (\delta \nu_8^{85} - \delta \nu_8^{87}) \tag{3}
\]

Here, $(\delta \nu_8^{85} - \delta \nu_8^{87})$ term can be identified as the difference between 1st and 4th peak in the dual isotope spectrum shown in the figure 6. Similar to hyperfine splitting values, here we can extract the isotope shift values in several independent ways. For example, the term $(\delta \nu_8^{85} - \delta \nu_8^{87})$ can be determined using various peak positions with the aid of $\Delta V^{85}_g$ and $\Delta V^{87}_g$. The ground state splittings $\Delta V^{85}_g$ and $\Delta V^{87}_g$ are taken from [18] and [11] respectively.

Compared to the single isotope spectrum shown in figure 5 we noticed a relatively rippled/noisy baseline for our dual isotope spectra as shown in figure 6. However, we tested the fitted peak positions against the individual peak fits versus full spectrum (6-peaks) fits and found no statistically resolvable differences. Therefore, we conclude that the fluctuating baseline (mostly associated with our gain setting of the differential photo-diode) didn’t affect our peak positions extracted by full spectrum fits.

3.4. Systematic error search and calibration

In order to assess any potential systematic effects that occurred during the experiment we divided our data files into different categories (subsets) and determine the hyperfine splittings based on laser sweep direction (up scan versus down scan), scan speed (slow versus fast), excitation transition (lower versus upper), and scan linearization (direct versus indirect measurements). The calculated averages of each subset converged to the final average value within their statistical uncertainty and found no evidence of any bias or trends associated with our subset measurements conducted for both isotopes as well as the isotope shift measurement.

During the Fabry–Perot data fitting process described in the data and analysis section, we used the nominal value of 500 MHz as the cavity’s FSR. However, this value does not guarantee the actual FSR value present during the experiment. In order to ensure the frequency calibration is done correctly, we independently measured the FSR a few times during the course of the experiment. Each time, this has been done by carefully observing frequencies that correspond to cavity resonances while the laser is operating in single mode. Using the fact that differences of all pairs of these frequencies must be an integer multiple of FSR, the determination of the best fit integer corresponding to each pair resulted in an independent FSR measurement. Then we compared these measured FSR values with a range of possible values (estimates) in the vicinity of 500 MHz/$\chi^2$ method. All of our FSR measurements correspond to the $\chi^2$ minimum resulted in similar and consistent values and found the average of 500.42 (15) MHz. Based on this value we corrected all of our final (uncalibrated) measurements by multiplying a correction factor defined as $c_{\text{cal}} = 500.42/500.00 = 1.0008(3)$. It is also important to note that throughout the course of the experiment the laser beam alignment through the Fabry–Perot cavity was maintained fixed as it could cause a slight change in FSR.

Our final (calibrated) hyperfine splittings and isotope shift results are summarized in table 1 along with their statistical and systematic uncertainty contributions based on our subset analysis described above. The use of equation (1) along with the hyperfine splittings found in table 1 allows us to determine hyperfine $A$ (magnetic-dipole) coupling constants for the rubidium $5P_{1/2}$ excited state. These results are listed in table 2 along with the previous measurements that exist in the literature.

4. Discussion

In laser spectroscopy experiments, one of the major issues with using ECDL in scanning mode is the frequency drifting and the reliability of obtaining frequency change proportional to the applied voltage ramp. The frequency drifting may have an effect on averaging a large number of spectra taken continuously during a considerable period of time, however, in this experiment we analyzed each spectrum separately and during one
scan period, zero to negligible drifting was observed. Furthermore, we analyzed each up-ramp and down-ramp scan separately so the duration during each scan is half of the scan period. Also, we took extreme care in the linearization procedure explained in section 3.1 by trying several polynomial orders and found that the 1st, 2nd or 3rd order polynomial was not sufficient to realize a linearized frequency axis. Use of 4th order polynomial fully linearized each of our spectra. The Fabry–Pérot etalon used in this experiment was a Burleigh (Burleigh instruments inc., serial no: 1176) high-precision etalon with custom mirrors to form a confocal cavity configuration. It is built with Invar rods that have an extremely low thermal expansion coefficient (∼4 × 10⁻⁷ K⁻¹). Even though, a few degrees of room temperature fluctuation has no effect on the extent of considerable FSR change at the level of precision considered in this experiment, a negligible or less than 1-degree room temperature change was observed during the course of the experiment.

5. Conclusion

The SAS cross-beam technique with differential detection has been demonstrated previously for the 5P₃/₂ state of ⁸⁷Rb using a 780 nm diode laser [19]. Here, SAS has been used to excite the relatively difficult ⁵S₁/₂ → ⁵P₁/₂ transition of rubidium and measure hyperfine splittings of ⁵P₁/₂ excited state for both isotopes using a homemade ECDL operating at 795 nm. The experiments and/or techniques used in the previous investigations listed in table 2 are significantly different from ours. Furthermore, even though a similar data analysis procedure may have been used with Doppler-free two-step two-color spectroscopy, this is the first time it has been used for SAS spectra.

Using our measured splittings hyperfine A magnetic dipole coupling constants have been extracted for both isotopes. According to table 2, even though the previous measurements of ⁸⁵Rb hyperfine constants are in reasonable agreement with each other it is clear that there are two groups of results that exist for ⁸⁷Rb in the literature. One group claims values close to 406 MHz and the other claims values close to 408 MHz. Our value supports the 408 MHz group and is in good agreement with the measurements done in [7] and more recently [10] within the stated error bars.

Even though our uncertainty estimates are an order of magnitude larger compared to the uncertainties claimed in those experiments our uncertainty is small enough to clearly resolve the discrepancy between the two groups. One of the motivations behind our experiment was to resolve this ⁸⁷Rb hyperfine constant discrepancy and clearly, we accomplished that goal. It is also important to note that the theoretical calculations support the higher-value group with 408.53 MHz [20], and 410.06 MHz [21]. It is possible some calibration errors that occurred in experiments described in [6, 8, 9] may have caused this discrepancy. The use of a frequency comb may improve the precision of our result by a factor of 100.

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Data availability statement

The data cannot be made publicly available upon publication because no suitable repository exists for hosting data in this field of study. The data that support the findings of this study are available upon reasonable request from the authors.

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**Table 2. Magnetic-dipole coupling constant measurements.**

| Source          | ⁸⁵Rb     | ⁸⁷Rb     |
|-----------------|----------|----------|
| This work (MHz) | 120.79(29)| 407.75(57)|
| [7] (MHz)       | 120.499(10)| 408.328(15)|
| [10] (MHz)      | 120.500(13)| 408.330(56)|
| [6] (MHz)       | 120.640(20)| 406.147(15)|
| [8] (MHz)       | 120.72(25) | 406.2(8)  |
| [9] (MHz)       | 120.645(5) | 406.119(7)|
ORCID iDs

P M Rupasinghe @ https://orcid.org/0000-0003-4656-332X

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