Advanced laboratory I

Optical pumping of rubidium

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Contents

1.	. Physical fundamentals	3
	1.1. Fine structure	3
	1.1.1. Zeeman effect	4
	1.1.2. Paschen-Back effect	5
	1.2. Hyperfine structure	5
	1.2.1. Zeeman effect \ldots	5
	1.2.2. Paschen-Back effect	5
	1.3. Optical pumping	6
	1.4. Spectral line width	6
2.	2. Experimental setup	7
3.	B. Measurements and results	8
3.	B. Measurements and results 3.1. Pumping time	8 8
3.	B. Measurements and results 3.1. Pumping time 3.2. Earth's magnetic field	8 8 9
3.	B. Measurements and results 3.1. Pumping time 3.2. Earth's magnetic field 3.3. Zeeman splitting	8 8 9 10
3. 4.	 3. Measurements and results 3.1. Pumping time	8 8 9 10 13
3. 4. A.	 3. Measurements and results 3.1. Pumping time	8 8 9 10 13 14

1. Physical fundamentals

The alkali metals are comprised of the group 1 elements in the periodic table, with the exception of hydrogen (and possibly the still to be synthesised ununennium), which barely exhibits alkali properties. All these elements have only one weakly bounded outer electron, which allows us to approximate their energy levels by calculating them for hydrogen.

1.1. Fine structure



Fig. 1: Energy splittings of hydrogen without regard to the hyperfine splitting, adapted from [2]

Looking at the various energy levels and energy splittings of hydrogen without the hyperfine splitting, which we will discuss later, we get an overview as seen in Fig. 1. It should be noted that the "normal" Zeeman effect is just a relatively rare special case of the anomalous Zeeman effect, however, it can be explained semi-classically with the Bohr model. The term "anomalous" is therefore just historical, as it could not be understood properly at the time of discovery, and is in fact contradictory, as it is the normal case [3, p. 214]. In the following, we will simply ignore this separation and talk about the anomalous Zeeman effect as the Zeeman effect.

In the classical gross structure of energy spectra as described by Bohr, the energy levels of a hydrogenic atom depend only on the pricipal quantum number n with

$$E_n = -\frac{me^4}{8\epsilon_0^2 h^2} \frac{Z^2}{n^2}$$
(1)

This model, however, is not accurate. A better model can be obtained by solving the Dirac equation with regard of relativistic and spin effects, which split the spectral energy lines.

This fine structure can be split into three separate corrective terms, which leads to the following Hamiltonian [3, p. 197f.]

$$H = H_0 + H_{\rm kin} + H_{\rm so} + H_{\rm Darwinian} \tag{2}$$

The first corrective term is based on the special relativity, which changes the kinetic term in the Hamiltonian from $T = \frac{p^2}{2m}$ to $T = \sqrt{p^2c^2 + m^2c^4} - mc^2$. Expanding this in a Taylor series yields the first order correction as:

$$H_{\rm kin} = -\frac{p^4}{8m^3c^2}$$
(3)

The second corrective term arises from the spin-orbit coupling. Shifting the frame of reference from the nucleus to the electron results in the nucleus orbiting the electron and thereby functioning as a current loop, which generates a magnetic field coupling with the magnetic field of the electron due to its intrinsic angular momentum. Therefore, the corrective term should contain the scalar product of L and S. We get:

$$H_{\rm so} = \frac{1}{2} \left(\frac{Ze^2}{4\pi\epsilon_0} \right) \left(\frac{g_s}{2m^2c^2} \right) \frac{\boldsymbol{L} \cdot \boldsymbol{S}}{r^3} \tag{4}$$

The last, Darwinian correction term can be interpreted as a smearing of the interaction between electron and nucleus due to rapid quantum oscillations of the electron. It is given by:

$$H_{\text{Darwinian}} = \frac{\hbar^2 \pi}{2m^2 c^2} \left(\frac{Ze^2}{4\pi\epsilon_0}\right) \delta^3(\boldsymbol{r})$$
(5)

1.1.1. Zeeman effect



wise the moments add to μ_j . Due to different g factors, μ_j precesses around **J**. From [3, p. 214] Looking deeper into the spin-orbit coupling [3, p. 215ff.], one finds a total magnetic moment as the (vector) sum of the orbital and spin magnetic moments:

$$\boldsymbol{\mu}_j = \boldsymbol{\mu}_s + \boldsymbol{\mu}_l \tag{6}$$

In contrast to the orbital and spin magnetic moments, where the vectors $\boldsymbol{\mu}_s$ and \boldsymbol{s} or respectively $\boldsymbol{\mu}_l$ and \boldsymbol{l} is antiparallel, one finds no similar coincidence for the total magnetic moment due to different g factors for spin and orbital magnetism. The vector $\boldsymbol{\mu}_j$ is in fact precessing (due to the strong coupling quite rapidly) around \boldsymbol{j} , which is fixed. We can therefore only observe the time average, which is it's projection on \boldsymbol{j} , written as $(\boldsymbol{\mu}_j)_{\boldsymbol{j}}$. Calculation yields, with multiple usage of the law of cosines:

$$|(\boldsymbol{\mu}_j)_{\boldsymbol{j}}| = g_j \sqrt{j(j+1)} \mu_B \tag{7}$$

with

$$g_j = 1 + \frac{j(j+1) + s(s+1) - l(l+1)}{2j(j+1)}$$
(8)

For multiple electron systems, on simply replaces s, l and j by S, L and J. The vector moment and its z component are:

$$(\boldsymbol{\mu}_j)_{\boldsymbol{j}} = -\frac{g_j \mu_B \boldsymbol{j}}{\hbar} \tag{9}$$

$$(\boldsymbol{\mu}_j)_{\boldsymbol{j},\boldsymbol{z}} = -m_j g_j \mu_B \tag{10}$$

Therefore with an applied magnetic field B we get an energy splitting of

$$\Delta E_B = (\Delta m_i) g_i \mu_B B \tag{11}$$

For optical transitions, the selection rules apply, with $\Delta m_j = 0$ for π transitions and $\Delta m_j = \pm 1$ for σ^{\pm} transitions.

1.1.2. Paschen-Back effect

In strong magnetic fields, where the energy level splitting due to the magnetic field is not small compared to the spin-orbit coupling, the Zeeman effect will not happen any more. With such a strong magnetic field, the fine structure coupling is dissolved and l and s are precessing independently around the magnetic field B. Therefore j becomes meaningless. The z components of the orbital and spin moments are now quantised individually, leading to a splitting of [3, p. 217]:

$$\Delta E = (\Delta m_l + 2\Delta m_s)\mu_B B \tag{12}$$

For optical transitions, the aforementioned selection rules now apply to Δm_l . Furthermore we have $\Delta m_s = 0$ as dipole radiation cannot cause a spin flip.

1.2. Hyperfine structure

The hyperfine structure works quite analogous to the spin-orbit coupling in the fine structure. Atomic nuclei possess an angular momentum I characterized the quantum number I [3, p. 362ff.]:

$$|\mathbf{I}| = \sqrt{I(I+1)}h\tag{13}$$

The observable z component is given as:

$$(I)_z = m_I h$$
 with $m_I = -I, -I+1, ..., I-1, I$ (14)

The connected magnetic moment is:

$$\boldsymbol{\mu}_{I} = \gamma \boldsymbol{I} = \frac{g_{I} \mu_{N}}{\hbar} \boldsymbol{I} \quad \text{with the nuclear magneton} \quad \mu_{N} = \frac{e\hbar}{2m_{P}} \tag{15}$$

The z component results as:

$$(\boldsymbol{\mu}_I)_z = g_I \mu_N m_I \tag{16}$$

Looking at the coupling between the angular momenta of the electrons J and the nucleus I, we can proceed analogous to the fine structure and get a total angular momentum F = J + I.

1.2.1. Zeeman effect

Applying a weak magnetic field B, this external field adds to the internal field B_J . If it is small enough to let the coupling of I and J remain intact, we get an energy splitting of [3, p. 371]:

$$\Delta E = g_F \mu_B B \Delta m_F \quad \text{with} \quad g_F = g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)}$$
(17)

As usual, for optical transitions, selection rules apply: $\Delta m_F = 0$ for π transitions and $\Delta m_F = \pm 1$ for σ^{\pm} transitions.

1.2.2. Paschen-Back effect

If however the external magnetic field B is strong enough to break the coupling of I and J, F becomes meaningless. The energy splitting now consists of three terms: The shell moment as explained in the Zeeman effect of the fine structure, a hyperfine splitting of these levels and the Zeeman energy of the nucleus [3, p. 372f.]:

$$\Delta E = g_J \mu_B \Delta m_J B + \frac{g_I \mu_N B_J}{\sqrt{J(J+1)}} \Delta m_I \Delta m_J - g_I \mu_N \Delta m_I B \tag{18}$$

For optical transitions the selection rules are $\Delta m_I = 0$ and $\Delta m_J = 0, \pm 1$.

1.3. Optical pumping

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Fig. 3: Energy level schema for ⁸⁷Rb with optical transitions, from [6]

In Fig. 3 we can see the fine, hyperfine and hyperfine Zeeman splitting of ⁸⁷Rb. Note that the energy is not to scale, in fact, the fine splitting is about five orders of magnitude larger than the hyperfine splitting, which in turn is about three orders of magnitude larger than the hyperfine Zeeman effect with an external field of some mT.

Assuming we are now inducing optical transmissions with σ^+ polarized light, which means that m_F will be increased by one. This intermediate state then decays with $\Delta m_F = 0, \pm 1$ into an excited state. Therefore, we are effectively emptying the initial state, as the excited state will in many cases not be the initial state, and thereby causing a population inversion.

In reality, due to collisions between atoms and between atoms and the walls, the atoms will relax from the excited state back into a thermodynamic balance. In effect, we will reach a state of equilibrium between the relaxation of excited states into thermodynamic balance and the creation of new excited states by optical pumping.

This equilibrium can be tuned in two ways: First, introduction of noble gases can be used to minimize collisions. Second, the intensity of the

light used to induce the optical pumping can be increased. Both possibilities lead to a higher proportion of excited states.

The time dependency of the number of atoms in an excited state can be calculated on the basis of a two level system. Assuming N_+ to be the number of atoms in the excited state and N_- to be the number of atoms in the ground state, with $N = N_+ + N_-$ the total number of atoms, we get the following differential equation:

$$\frac{dN_+}{dt} = \frac{N - N_+}{\tau_{\text{pumping}}} - \frac{N_+}{\tau_{\text{relaxing}}}$$
(19)

With
$$\tau := \left(\tau_{\text{pumping}}^{-1} + \tau_{\text{relaxing}}^{-1}\right)^{-1}$$
 and $N_0 := \frac{N}{1 + \frac{\tau_{\text{pumping}}}{\tau_{\text{relaxing}}}}$ we get:
$$\frac{dN_+}{dt} = \frac{N_0 - N_+}{\tau}$$
(20)

With the initial condition $N_+(0) = 0$ this is solved by:

$$N_{+}(t) = N_0 \left(1 - \exp\left(\frac{-t}{\tau}\right) \right)$$
(21)

1.4. Spectral line width

In this experiment, we will use a rubidium spectral lamp to induce optical pumping of rubidium. One might think that this should not be possible, as the lamp emits sharp spectral lines, which cannot induce all permissible transitions between the various Zeeman levels, as they are by definition of varying energy.

However, several mechanisms broaden the spectral lines, therefore, they are not ideally sharp.

The first mechanism is Heisenberg's uncertainty principle. This leads to a homogeneous broadening dependent on the lifetime τ of the energy levels. As is well known, the energy uncertainty of one such energy level is given as:

$$\Delta E = \frac{\hbar}{\tau} \tag{22}$$

The broadening of the emitted frequency is therefore given as

$$\Delta f = \frac{1}{2\pi} \left(\frac{1}{\tau_1} + \frac{1}{\tau_2} \right) \tag{23}$$

with the lifetimes τ_1 and τ_2 of the lower and upper energy levels.

Further homogeneous broadening comes from collisions with other atoms. This leads to a broadening of

$$\Delta f = \sqrt{\frac{3}{4mk_BT}}pd^2 \tag{24}$$

with the atomic mass m, the atomic diameter d and the pressure p.

An other source of broadening is the Doppler effect. This broadening is inhomogeneous and depends on the velocity of the gas atoms. It is given non-relativistically as:

$$\Delta f = f \frac{v}{c} \tag{25}$$

2. Experimental setup



Fig. 4: Schematic of our experimental setup, from [1]

Our experimental setup as seen in Fig. 4 consist of a high-frequency rubidium gas discharge lamp (1), whose light is collected by a lens (2). It then passes an interference filter (3), which transmits the D_1 line and suppresses the D_2 line. It is then first linearly polarized (4) and then passes a quarter-wave

plate (5), thereby becoming circularly polarized. Afterwards, the light enters the rubidium absorption chamber (10), which can be heated (11) and which is located between two pairs of Helmholtz coils (6) and the coils (12) of a high frequency transmitter (7). In the end, the light gets collected by a lens (8) and detected by a photo transistor (9).

The light entering the absorption chamber fulfils two purposes. On the one hand, it is initiating a pumping process. On the other hand, a optically pumped absorption chamber transmits more light. Measuring the transmitted light via a photo transistor, we therefore get an information about the population of energy states in the chamber. The current – and via a resistor the voltage – measured at the photo transistor is proportional to the light intensity.

3. Measurements and results

3.1. Pumping time

We measured the pumping time during the initial heating up phase of the rubidium lamp. Therefore, we get multiple intensities quite easily.



Fig. 5: Pumping processes at increasing lamp temperatures and intensities. Intensities increase from (a) to (d)



Fig. 6: First fully visible pumping processes from Fig. 5 with exponential fit

We get the following pumping times with $I_1 < I_2 < I_3 < I_4$:

Intensity	Pumping time τ [ms]
I_1	3.798 ± 0.021
I_2	4.353 ± 0.020
I_3	3.555 ± 0.013
I_4	4.056 ± 0.010

We cannot detect any correlation between the light's intensity and τ . Therefore, τ cannot be the pumping time from equation 21. Interestingly, the values are different from each other even considering multiples of their respective error.

3.2. Earth's magnetic field

In this part, we determine the horizontal intensity of earth's magnetic field by applying a rectangularly varying field with an added constant field. If the resulting magnetic field negates earth's magnetic field, every second pumping process is omitted. By changing the polarity of the constant magnetic field, we become independent of the varying field:

$$B_{\text{earth}} + B_{\text{AC}} - B_2 = 0 \tag{26}$$

$$B_{\text{earth}} - B_{\text{AC}} + B_1 = 0 \tag{27}$$

Adding these two equations, we get:

$$B_{\text{earth}} = \frac{B_2 - B_1}{2} \tag{28}$$

The strength of the constant magnetic field can be derived from the applied current by:

$$B = \mu_0 \mu_r \left(\frac{4}{3}\right)^{\frac{3}{2}} \frac{nI}{R} \tag{29}$$

The used coils have n = 80 and R = 90 mm. As copper has nearly no magnetic susceptibility[4], we can assume $\mu_r = 1$. All put together, we get:

$$B_{\text{earth}} = \mu_0 \mu_r \left(\frac{4}{3}\right)^{\frac{3}{2}} \frac{n}{2R} \cdot (I_2 - I_1) \qquad \Delta B_{\text{earth}} = \mu_0 \mu_r \left(\frac{4}{3}\right)^{\frac{3}{2}} \frac{n}{2R} \sqrt{(\Delta I_2)^2 + (\Delta I_1)^2} \tag{30}$$

We measured at three different intensities of the AC field:

I_1 [A]	I_1 [A]	B_{earth} [µT]
1.24 ± 0.02	1.30 ± 0.02	51 ± 24
1.73 ± 0.02	1.78 ± 0.02	43 ± 24
2.25 ± 0.02	2.26 ± 0.02	9 ± 24

Averaging yields a result of $B_{\text{earth}} = (34 \pm 24) \,\mu\text{T}$. Obviously, the error is very large. We assumed an error of 0.02 A for the currents because the displays were varying by 0.01 A and we assumed the same value as a measurement error of the indicator.

However, the determined value fits well with the calculated value of $B_{\text{earth}} = 19.0 \,\mu\text{T}$ at a latitude of 52°2′, a longitude of 8°29′ and an elevation of 132 m on the date of measurement, as calculated with 11th IGRF [5].

3.3. Zeeman splitting

To determine the Zeeman splitting, we apply a triangular current to the Helmholtz coils and additionally apply a high frequency field. If the energy of the high frequency field equals the Zeeman energy splitting, the pumping process is disturbed. By looking up the voltage/current applied at the Helmholtz coils in that moment and thereby determining the current magnetic field, we get two equations for the energy splitting:

$$\Delta E = hf \tag{31}$$

$$\Delta E = \mu_B g_F \Delta m_F B \tag{32}$$

As we are measuring the voltage applied at the Helmholtz coils with an oscilloscope, we first need to determine their resistance to be able to calculate the current. We do this by applying various direct currents to the coils:



Fig. 7: Voltages and currents applied to the Helmholtz coils

With linear regression we get a resistance of $R = (3.749 \pm 0.031) \Omega$.

Looking at Fig. 8, we see two distinct minima in the transmitting intensity. They belong to the two natural isotopes of rubidium, ⁸⁵Rb and ⁸⁷Rb. Putting equations 31 and 32 together with the determined resistance and the voltage of the minima, which we can easily read in the oscilloscope data file, we get:

$$g_F = \frac{hfRr}{\mu_B\mu_0\mu_r \frac{4}{3}^{\frac{3}{2}}nU} \qquad \Delta g_F = g_F \cdot \sqrt{\left(\frac{dU}{U}\right)^2 + \left(\frac{dR}{R}\right)^2 + \left(\frac{df}{f}\right)^2} \tag{33}$$



Fig. 8: Zeeman splitting at different frequencies of the high frequency emitter

f [Hz]	U [V]	q_F	Isotope
340112 ± 1	0.31 ± 0.16	0.339 ± 0.170	$^{87}\mathrm{Rb}$
340112 ± 1	0.63 ± 0.16	0.169 ± 0.042	$^{85}\mathrm{Rb}$
527450 ± 50	0.63 ± 0.16	0.263 ± 0.066	$^{87}\mathrm{Rb}$
527450 ± 50	1.25 ± 0.16	0.131 ± 0.016	$^{85}\mathrm{Rb}$
860845 ± 5	1.72 ± 0.16	0.156 ± 0.014	87 Rb
860845 ± 5	3.13 ± 0.16	0.086 ± 0.004	85 Rb
1028990 ± 1	2.34 ± 0.16	0.137 ± 0.009	$^{87}\mathrm{Rb}$
1028990 ± 1	4.06 ± 0.16	0.079 ± 0.003	$^{85}\mathrm{Rb}$
1188351 ± 1	2.81 ± 0.16	0.132 ± 0.007	87 Rb
1188351 ± 1	5.00 ± 0.16	0.074 ± 0.002	$^{85}\mathrm{Rb}$

One can notice, that the g factor seems to be dependent of the frequency: The higher the frequency, the lower the g factor. Interestingly, an other group had the inverse phenomenon: The higher the frequency, the higher the g factor. As the g factor should be constant, there seems to be a systematic error somewhere in the setup.

Additionally one can notice, looking at the applied voltage at the Helmholtz coils in Fig. 8, that this voltage has quite a lot of noise. This might indicate a larger error in the voltage than assumed.

Ignoring the frequency dependency and averaging over the g factors yields:

$$g_F(^{87}\text{Rb}) = 0.21 \pm 0.09$$
 $g_F(^{85}\text{Rb}) = 0.11 \pm 0.04$ (34)

The theoretical value can be calculated from equation 17 with $S = \frac{1}{2}$ and L = 0 leading to $J = \frac{1}{2}$ as well as $g_J \approx 2$, $I = \frac{3}{2}$ for ⁸⁷Rb and $I = \frac{5}{2}$ for ⁸⁵Rb:

$$g_F(^{87}\text{Rb}) = \frac{1}{2} \qquad g_F(^{85}\text{Rb}) = \frac{1}{3}$$
(35)

Obviously, our results do not match the theoretical values even with regard to multiples of their errors.

Assuming no knowledge of the spin of rubidium, one would calculate it by transposing equation 17:

$$I = -\frac{1}{2} + \frac{1}{2} \frac{g_J}{g_F} \qquad \left(\frac{\Delta I}{I}\right)^2 = \left(\frac{\Delta g_F}{g_F}\right)^2 \tag{36}$$

Applying our results for g_F leads to:

$$I(^{87}\text{Rb}) = 4.26 \pm 1.83$$
 $I(^{85}\text{Rb}) = 8.59 \pm 3.12$ (37)

As expected, the values are way off from the true values.

4. Conclusions

In this experiment, we were able to optically pump rubidium. We determined a relaxation time, which however is not the relaxation time from optical pumping, as it is intensity independent.

We were further able to successfully measure earth's magnetic field, albeit the uncertainty of our determined values of roughly 70% is not satisfying.

We were also able to observe the hyperfine Zeeman splitting by applying a high frequency field. However, the determined g factors do not match with theory. Furthermore, there seems to be a systematic problem, as our g factors are frequency-dependent, which they should not be. As the g factors do not match, the calculated nuclear spins do not match with theory as well.

A. References

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B. Data

This protocol as well as the recorded data is available online at http://www.sknust.de/sci/2012/protocols/rubidium/.