Lab Report on **Pulsed Laser-Spectroscopy** on Na-Atoms

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1 Introduction

1.1 Principles of the Laser

The standard laser is a device for producing highly coherent, directed, and monochromatic light. It is based on the principles of absorption and emission in matter.

1.1.1 Emission and Absorption: Einstein Coefficients

Any material that has two or more energy levels can, in general, absorb photons to become excited from the lower level (E_k) into the higher level $(E_i = E_k + h\nu)$, and emit photons to relax from the higher into the lower level. This emission and absorption can be described by the three Einstein coefficients:

- B_{ki} Stimulated Absorption: The atom absorbs a photon of energy $h\nu$ and is excited into the higher level.
- B_{ik} Stimulated Emission: A photon of energy $h\nu$ causes the atom to relax to the lower level, emitting a second photon of the same energy that is coherent with the first photon.
- A_{ik} Spontaneous Emission: The atom relaxes into the lower level spontaneously, emitting a photon of the energy $h\nu$.

The probability of the atom to change from the lower level into the higher level (absorption) in a radiation field of photons with an energy density of w_{ν} is

$$P_{ki} = B_{ki} \cdot w_{\nu} \tag{1}$$

The reverse, emission, is described by

$$P_{ik} = B_{ik} \cdot w\nu + A_{ik} \tag{2}$$

Under normal conditions there is an equilibrium between absorption and emission, the states are occupied according to the Boltzmann-distribution. In this equilibrium, the induced absorption and induced emission can be shown to be equal, thus there is no amplification of the radiation field.

1.1.2 Inversion of Occupied States: Laser Emission

The purpose of a laser, however, is to *do* amplify the radiation field. So, the equilibrium of levels has to be inverted, so that induced emission becomes prevalent. This is known as the inversion of occupied states: A high occupation on the high energy level, and a low occupation on the low energy level.

Technically, in order to bring the electrons to the higher level 2, we need to use another level 3, higher than 2, to which the electrons are first "pumped" with any means of adding the required amount of energy to the system. From level 3, the electrons quickly decay into level 2, where they are stable for some amount of time. Thus we reach the desired inversion of states between level 1 and level 2.

At some point, spontaneous emission from level 2 to level 1 occurs, releasing a photon. Now, this photon can in turn produce stimulated emission in other atoms at level 2: Because of the inversion of states, we can have a cascade of stimulated emission, quickly producing a lot of coherent photons of the same frequency. While the normal delayed decay of the electron is known as fluorescence, the amplifying cascade is adequately called super-fluorescence. To enhance the effect even further, the photons are reflected back into the laser chamber by mirrors (resonance), inducing further emission. A small percentage of the photons is removed from the system to be used as the laser beam. The pumping is meanwhile continued, so that the laser produces a continuous beam.

The described model is the simplest case of a three-level-laser. Laser with more levels can be constructed, in which the process of pumping and emission is more complicated. The concept however remains the same in all cases.

1.2 Dye-Laser

In comparison to the simple laser which just emits monochromatic coherent light of a constant wavelength, the dye laser is tuneable within a certain wavelength range. The shematic setup of the device used in this experiment is shown in the lower part of Fig. 1. The difference to the standard laser is the active medium.



Fig. 1: Experiment Setup

In case of the dye laser a complex molecule with advanced properties is used to achieve the tuneability. The molecule still has discrete electronic states, but in addition many many possible vibrational states. The energy differences between these states are very small (within the natural linewidth), as shown in Fig. 2. This means that, if all these states are excited, one will not observe discrete lines, but a continuous spectrum within a certain wavelength range as fluorescent (spontaneous) emission.

As shown in Fig. 2, the dye used in this experiment has a three level structure, which is the minimum requirement for building a laser (cf. section 1.1). Electrons in the ground state S_0 are being pumped to the S_2 level by a pulsed nitrogen laser and decay in arbitrary substates of the excited state S_1 without emitting any radiation. The laser transition is between these substates and the S_0 states, induced by stimulated emission. The resulting "super-fluorescent



Fig. 2: Electronic Structure of the Dye Molecule

light" has the same properties as normal fluorescent light before but with additional coherence.

1.2.1 Color Selection

To conclude the setup of a laser a resonator is needed. As described in section 1.1.2, this is done by adding a mirror on the one side and, specifically for the dye laser, a reflection grating on the other side of the dye container. Depending on the orientation of the lattice, only one specific sharp wavelength is reflected back into the dye causing stimulated emission. By rotating the lattice one can now select the wavelength within the range given by the molecule structure.

In addition to the properties mentioned, the dye laser used in our experiment is *pulsed*, i.e. it only emits very short bursts (3.2 ns) of laser light. This is done by using a second nitrogen laser for pumping which is also pulsed.

1.3 Absorption in Sodium

Such a pulsed dye laser can be used to examine the emission spectrum in sodium. The short laser pulse is absorbed by the Sodium atoms, which reemit the light as fluorescence.

The principles of emission and absorption are the same ones described in section 1.1.1.

The specific absorption/emission spectrum that we intend to observe in sodium is described by the atom's fine structure. The specific two lines are the transitions $3^2P_{1/2} \longrightarrow 3^2S_{1/2}$ and $3^2P_{3/2} \longrightarrow 3^2S_{1/2}$ with a wavelength of 589.6591 nm and 589.0617 nm in air, respectively (cf. [2]).

2 **Objectives**

We expect the experimental setup to provide the possibility of time-resolved measurements within a nanosecond time scale. We will be building a dye laser pumped by a pulsed nitrogen laser and observe the laser spectrum the timedependant evolution of the fluorescence in the pulse.

After setting up the laser we excite the $3s^2S_{1/2}$ and $3p^2P_{1/2}$ states in a sodium sample and examine the lifetime of these states as well as the linewidth of the sodium D line emission.

3 **Experimental Setup**

The general setup is shown in Fig. 1. First of all, we recorded the time-development of the super-fluorescence of the dye when excited by the nitrogen laser at three different sample rates.

In the next step, we measured the fluorescence intensity within a range of 560 nm to 620 nm. The result will be compared to the absorption spectrum of the dye later.

Now, we set up the laser by adjusting the grating manually. After that, we could clearly observe a speckle pattern, confirming the laser activity. A Fabry-Perot-Etalon was used to observe and calibrate the wavelength tuning of the dye laser. For this we mapped the position of the step motor controlling the laser grid to the corresponding wavelength measured with the monochromator.

The next part of the experiment was the excitation of the sodium. We focused the laser beam into the center of the heated sodium cell (temperature inside: 116.4 °C) and started time-resolved measurements using the fast sampler due to the high time-resolution of 200 ps. The observed time delay was 100 ns. We used the step motor to change the wavelength of the dye laser and recorded the fluorescence intensity at the wavelength position of the D lines and at a position of zero fluorescence. After that, we measured the intensity in dependence of the step motor position to observe the frequency separation between both lines. In order to map a frequency interval to a step interval, we did an additional calibration measurement with the Fabry-Perot-Etalon.

$\mathbf{4}$ Results

Fluorescence of the Dye 4.1

The decay constants of the fluorescence of the dye in the first measurements are presented in the table below:

samples	decay constant	$``tastverh\"altnis"$
3	$(224 \pm 4) \text{ ns}$	$2.24 \cdot 10^{-6}$
30	$(476 \pm 10) \text{ ns}$	$4.76 \cdot 10^{-6}$
300	$(1624 \pm 30) \text{ ns}$	$1.62 \cdot 10^{-5}$

The results obtained clearly show that the time-evolution of the dye's fluorescence seems to depend on the sample rate. This cannot be true and has to be explained by the way the measurement devices operate. The boxcar averager calculates the intensity by averaging over a certain amount of recent samples, given by the sample rate. This is an effective way to reduce noise but it can also lead to wrong results, because a situation may occur where no intensity is left after a certain period of time, but the averaging process which uses data from the past still calculates a value greater than zero. This is what happend in the case of 300 samples. We decided to use 30 samples as a value providing good noise reduction while introducing a still acceptable error into the measurements.

4.2 Absorption and Emission Spectrum of the Dye

Fig. 3 shows in arbitrary units the position of the absorption spectrum of Rhodamin 6G (first peak) and the fluorescence emission (second peak) obtained in the second measurement, as well as the laser emission measured (third peak). The values where obtained using the calibration curve shown in Fig. 4 and interpolated using cubic splines for better comparison.



Fig. 3: Absorption and Emission of Rhodamin 6G

One can clearly see that the position of the fluorescence and laser emission peak differs from the position of the absorption peak. This can be explained by radiationless vibrational transitions. During the absorption process not only electronic states but also molecule vibrations are excited, which the decay into the ground state without emitting radiation. This leads to an energy shift which can be observed as a difference in the position of absorption and emission peak.

4.3 Width of the Dye Laser Pulse

After setting up the laser we made several time-resolved measurements to observe the fluorescence of the sodium D lines.

The full-width-at-half-maximum of the laser pulse was

$$t = (6.7 \pm 0.6) \,\mathrm{ns}$$

 $T_{10 \,\mathrm{Hz}} = (6.7 \pm 0.6) \cdot 10^{-8},$



Fig. 4: Calibration of the Step Motor Position

which is a lot smaller than for the super-fluorescence.

4.4 Results for the Sodium D-Lines

4.4.1 Frequency Difference Between the Lines

By tuning the laser, we successfully observed resonance fluorescence of the sodium D-lines in the chamber. Using the calibration curve for mapping the grating position to a wavelength, we calculated the frequency difference of the lines to be

$$\Delta \nu_{\text{exp}} = (515 \pm 41) \,\text{GHz}.$$

The theoretical prediction calculated from the wavelength values in section 1.3 is

$$\Delta \nu_{\text{theo}} = 516 \,\text{GHz}.$$

The values are identical.

The calibration curve and its theoretical prediction are shown in Figure 5. It is clear that the measured calibration curve is a lot broader than the prediction, because of the finite bandwith of the laser discussed below.

4.4.2 Bandwith of the Laser Emission

A good approximation of the expected bandwidth of the laser emission can be obtained by only taking into account the finite resolution of the reflection grating. The angle between laser beam and grating is given by the Bragg condition to the fifth order.

$$2a\sin(\theta) = 5\lambda\tag{3}$$



Fig. 5: Calibration Curve with Theoretical Prediction

Assuming a wavelength of $\lambda=590$ nm and a beam diameter of d=1 mm one gets a resolution of

$$\frac{\lambda}{\Delta\lambda} = 5\frac{d}{a\cdot\cos(\theta)}$$
$$\approx 6.4\cdot10^3$$

This means that the bandwidth should be about

 $\delta \nu_{\rm L, theo} \approx 160 \, {\rm GHz}$

The values we read directly from our measurements are, for comparison:

$$\delta \nu_{\rm L, D1} = (133 \pm 15) \,{\rm GHz}$$

 $\delta \nu_{\rm L, D2} = (124 \pm 13) \,{\rm GHz}$

The Doppler broadening in our experiment was $\delta\nu_{\rm D} = 1.5$ GHz and can therefore be neglected.

4.4.3 Absorption in Sodium

Using the oscillator strength f and assuming a Gaussian bandwidth of $0.42\delta\nu_D$, we can calculate the absorption coefficient (cf. [1]) as follows:

$$\int_{0}^{\infty} \alpha_{ik} \cdot e^{\frac{(\nu-\nu_0)^2}{2 \cdot (0.42\delta\nu_D)^2}} = n\frac{h\nu}{c} \cdot \frac{e^2}{4m_e\epsilon_0h\nu} f$$

$$\Rightarrow \quad \alpha_{ik} = \frac{1}{\sqrt{2\pi} \cdot 0.42\delta\nu_D} \cdot n\frac{h\nu}{c} \cdot \frac{e^2}{4m_e\epsilon_0h\nu} f_{D2}$$

$$= \frac{ne^2}{\sqrt{2\pi} \cdot 0.42\delta\nu_{D2} \cdot 4cm_e\epsilon_0} f \qquad (4)$$

The density of sodium atoms in the chamber at T = 116.4 °C is about $n \approx 10^{16} \,\mathrm{m^{-1}}$. Therefore, the absorption coefficient for the two lines is

$$\alpha_{\rm D1} = 5.5 \,\mathrm{m}^{-1}$$

 $\alpha_{\rm D2} = 11 \,\mathrm{m}^{-1}$

The average absorption coefficient within the interval $[\nu_0 - 1.3\delta\nu_D, \nu_0 + 1.3\delta\nu_D]$ resulting from an integration over the Gauss curve is then

$$\bar{\alpha}_{D1} = 2.2 \,\mathrm{m}^{-1}$$

 $\bar{\alpha}_{D2} = 4.4 \,\mathrm{m}^{-1}$

Using the geometry of the chamber, we can calculate the probability of absorption.

$$I_{D1} = 1 - e^{-0.06 \cdot 2.2} = 12\%$$

$$I_{D2} = 1 - e^{-0.06 \cdot 4.4} = 23\%$$

The probability for a fluorescence photon to be emitted at the center of the chamber and to travel 25 cm to the detector is then about

$$P_{\rm D1} = 10\%$$

 $P_{\rm D2} = 5\%$

These results can be used to guess the total absorption of the sodium D_2 line assuming a triangular shaped laser intensity distribution. The maximum intensity occurred at 592 nm. The total range was 25 nm. The line is at $\lambda =$ 589.0617 nm with a broadening of $\Delta \lambda = 0.153$ nm. This means that the position of maximum intensity I_0 where absorption takes place is

$$I_{\rm D2} = 0.38 I_0$$

The intensity absorbed by the D_2 line is 23% of this value, i.e.

$$I_{\rm abs, D2} = 0.09 I_0$$

This is 0.4% of the total intensity $(I_{\text{tot}} = 25I_0)$.

4.4.4 Natural Linewidth

In the last experiment we examined the lifetime of the resonance-fluorescence. The measured fluorescence is shown in Fig. 6 The measurement was influenced by a high frequency disturbance, which resulted in a high error for the calculation of the decay constants. However, we linearized the plot (Fig. 7, 8) and calculated the decay constants which correspond to the lifetime of the excited sodium states.

The measured lifetimes of both states are

$$\tau_{D1} = (25 \pm 3) \text{ ns}$$

 $\tau_{D2} = (29 \pm 3) \text{ ns}$

which is much higher then the theoretical values

$$\tau_{\rm D1, \ theo} = 16.38 \ {\rm ns}$$

 $\tau_{\rm D2, \ theo} = 16.36 \ {\rm ns}$

The reason for that may be a reabsorption of photons within the chamber. This would increase the measured lifetime, because the electrons can populate



Fig. 6: Exponential Decay of the Fluorescence



Fig. 7: Linearized Decay of the Sodium D_1 Line



Fig. 8: Linearized Decay of the Sodium D_2 Line

the excited states multiple times. Another reason for the error may be the disturbances observed during the experiment.

The natural linewidth calculated from our values is

$$\Delta \nu_{D1} = (6.4 \pm 0.6) \text{ MHz}$$

 $\Delta \nu_{D2} = (5.5 \pm 0.5) \text{ MHz}$

5 Conclusion

In the experiment, we could clearly show that spectroscopy with a pulsed dye laser is an excellent tool to resolve both small energy and time differences. We were successful in constructing the dye laser and examining its properties. We could then very accurately resolve the two sodium D-lines. The measurement of the states' life-times was not quite as successful due to the difficulties described above.

References

- [1] DEMTRÖDER, Wolfgang: Laser Spectroscopy. Springer, 1988
- [2] WEAST, Robert: Handbook of Chemistry and Physics. CRC Press, 1990