The relevant appendices can be found under 'Related Documents' for each experiment at this link:
http://sky.campus.mcgill.ca/Exp/exp.a5w#

Optogalvanic Spectroscopy

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Warnings:

- It is important not to bring your eyes at the height at the laser as it can severely damage your retinas, even before the dye module is placed, where the laser beam is invisible because it's in the ultraviolet.
- The Ne-Na hollow cathode lamp is extremely fragile. It is a sodium lamp packed under low pressure neon. If the glass lamp is damaged, it could implode. Please handle with care.

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1 Introduction

The optogalvanic effect is the change in the electrical properties of a gas discharge, induced by radiation having a wavelength corresponding to an atomic or molecular transition of the discharge medium. Thus, by measuring the voltage across the discharge, as the wavelength is tuned, one is provided with a powerful spectroscopic tool [1].

2 Theory

2.1 The Optogalvanic Effect

The optogalvanic effect is the observation of modified electrical properties of gas discharge under the effect of radiation. A gas discharge is a gas undergoing ionizing reactions, that is, reactions that strip some atoms of their electrons. These free electrons enable current to go through the gas.

Due to the quantization of energy levels of the electronic cloud of an atom, an electron needs a specific amount of energy to jump between a lower state to a higher energy state. This energy can be acquired through radiation. By shining radiation at a specific wavelength on an atom, one can enable a specific jump from one state to another.

This is used in the optogalvanic effect. The discharge gas is exposed to radiation, usually a laser; if a correct wavelength is used, atoms will jump to higher energy states. Being in an higher energy state reduces the energy required to attain ionization, but this state may also have a higher chance of radioactive decay back to a lower energy state than the original one. Therefore, depending on the nature of the jump, some laser wavelengths may reduce or increase the ionization probability of certain atoms. Increasing ionization probability will increase the current in the gas, thereby reducing the voltage discharge, while reducing the ionization probability has the inverse effect.

Usually, the optogalvanic effect is studied with very short pulses. Each pulse will upset the balance in the discharge gas, causing distinct voltage curves as the gas re-equilibriates. To properly understand the behavior of the voltage curves, you can picture them to be mathematically represented as a sum of exponentials [2]:

$$\Delta V(t) = \Sigma a_i e^{-b_i t} \tag{1}$$

Of course, change in the current flowing through the lamp will affect the shape of the signal. For example, a higher current will increase the probability of ionization, which may ionize atoms that would have decayed to a lower energy state given lower current.

2.2 Spin-Orbit Coupling

Transitions occur due to the presence of different electronic layers. These layers are governed by the orbital angular momentum and the principal quantum number of the electron (L and n). These layers can be divided into shells and sub-shells: the principal quantum number controls the main shell, while the angular momentum controls the sub-shell level and shape. The shells are labeled using the corresponding quantum number, while the subshells are labeled using s, p, d, f and g, which corresponds to $L = 0, 1, 2, 3, 4$. These layers, having different energy levels, require energy change for the concerned electrons to tunnel between two layers. This can happen through radiation, either by absorbing or emitting photons.

In our case, we are interested in the 3s – 3p transition of the electron. The splitting of the transition comes from the spin of the electron, and its interaction with the magnetic field induced by the nucleus. Therefore, depending on the orientation on the spin, two electrons in the same 3p layer will have different energy levels, and will thus emit photons of different energy when they jump to the 3s state. The energy stored in intrinsic spin is much smaller than the energy required to jump from 3p to 3s layer to another. Therefore, we observe a small splitting in photon wavelength when looking at where the 3p -> 3s transition should be located.

2.3 Linewidth

Any light source, which includes coherent light sources such as lasers, have an uncertainty related to their emitted wavelength. This uncertainty can be caused by impurity in the source used, but even in completely pure samples, there is a lingering uncertainty that can be explained by the Heisenberg uncertainty principle: $\Delta t \Delta E \geq \frac{\hbar}{2}$. This uncertainty in the laser's wavelength takes the form of a Gaussian distribution when looking at the emission wavelength. Recall that the equation for a Gaussian is:

$$f(x) = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{(x-\mu)^2}{2\sigma^2}} \tag{2}$$

We know that μ is the mean of the distribution and σ its variance. We also know that maximum is attained at $x = \mu$, or when the exponential vanishes to one:

$$f_{max} = \frac{1}{\sigma\sqrt{2\pi}} \tag{3}$$

We then want to equate half of that to equation 2, and simplify on both sides to obtain:

$$\frac{1}{2} = e^{-\frac{(x-\mu)^2}{2\sigma^2}}$$

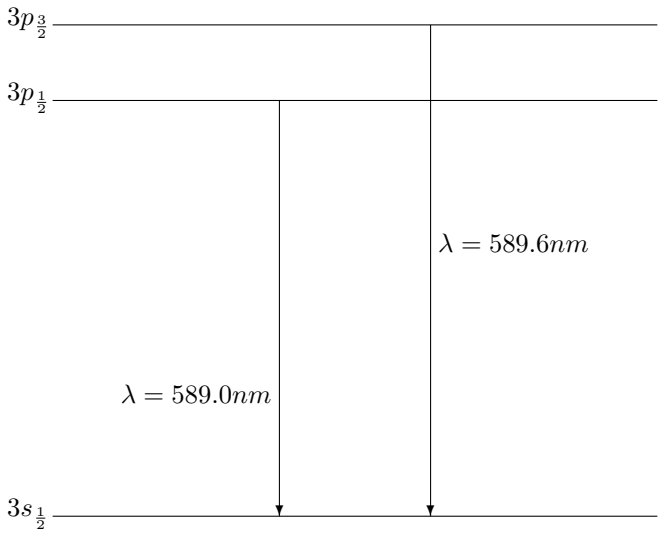

Figure 1: Splitting of the sodium doublet. The value in subscript to the left represents the J, the total angular momentum quantum number

$$\sqrt{ln(2)} = \left|\frac{x-\mu}{\sqrt{2}\sigma}\right|$$
$$\sigma\sqrt{2ln(2)} = |x-\mu|$$

This yields the half-width at half-maximum, or the distance between the half-maximum point and the maximum point. In spectroscopy, the full width at half maximum is usually used, also known as the linewidth. We then double our previous result to obtain:

$$l = 2\sigma\sqrt{2ln(2)} \tag{4}$$

3 Apparatus

The resistor and capacitor on the right side of figure 2 are in the orange box. You simply need to make the connections with the rest of the equipment. Do not force in the connection between the orange box and the lamp; make sure that the notch on the lamp's plastic end is properly aligned with its hole on the connector. Use the two rotor stands and clamps to hold the lamp and the photodetector at the right level. You will want to plug in the photodetector in CH2 of the oscilloscope, while the orange box will be plugged in CH1. Make sure that the nitrogen laser and the dye module are aligned with their respective black notches on top of them. Finally, make sure that the shudder isn't closed on the nitrogen laser.

The ammeter is plugged into the orange box. Remember the convention: positive on the red slot. Since the ammeter is analog and has no negatives, plugging it the opposite way will damage it. **Do not turn on the DC power supply until you read section 3.3.**

3.1 Alignment of photodetector

Please leave the DC power supply offline for this section.

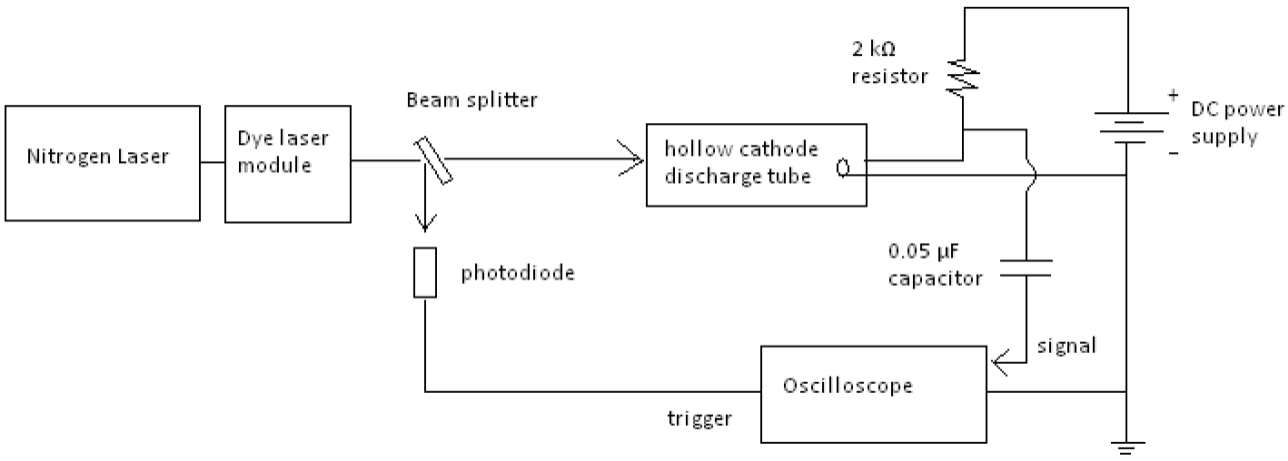

Figure 2: Block diagram of the apparatus. Note that there are no electrical connections between the nitrogen laser, the dye module, the beam splitter, the photodiode and the lamp – the arrows represent light rays.

On the oscilloscope, pressing *CH1 menu* and *CH2 menu* twice will show and hide the respective channel. For now, hide channel one and only show channel two. Press *Trigger menu*. Set the following options:

```
Edge
Slope:    Falling
Source:   CH2
Mode:     Normal
Coupling: AC
```

Set the trigger level to around -40mV. Unless the laser is powered on, the oscilloscope should not be triggering.

Now, turn on the laser, and turn the frequency knob until you hear it has reached maximum frequency. If you're lucky, your oscilloscope will start to trigger, but you probably won't be lucky. If you're using a glass prism as splitter, remember that not all the side faces are perfectly perpendicular to the the two large ones. Make sure that your prism is upright, as this makes the rest of the setup way easier. You may need to elevate your splitter to align it with the hole in the dye module.

The dye module has a knob on its front. It increases the intensity of the resulting laser. Turn it all the way up. Align the photodiode with the laser beam. This can be done using a white paper in front of the photodiode to show where the beam is hitting. This may require turning off the lights of the room to properly see what you're doing.

Adjust the position of the photodiode until you see the oscilloscope triggering. Once it triggers, you should see a negative peak on the oscilloscope. Continue to adjust the position of the photodiode until you can get the highest possible intensity on the peak. You want the photodiode to be rather close to the prism to make adjustment easier. Note that changing the resolution of the oscilloscope may interfere with the triggering. Just adjust the trigger level to get it working again.

3.2 Alignment of the Ne-Na lamp

Optimizing the position of the lamp is arduous. If you (carefully) pick up the lamp in your hands and look through the glass end of the lamp, you should see a dark circle surrounded by a metal rim. Note that this dark piece is hollow. You want your laser to hit the black surface, close to the middle; optimally, it would hit the

hollow center where the gas concentration is the highest. Due to the placement of the apparatus, you will have to look through the curved side of the lamp to see where the beam is hitting. Keep in mind that you will probably see a lot of "ghosting" from the curved glass – the red dot may not lie where it seems to be, and may appear to be at multiple places at the same time. If possible, turning off the lights of the room may be useful.

3.3 Turning on the lamp

To power up the lamp, you need to reach a rather high potential difference between the two ends. Doing so however may expose the lamp to a high current. The power supply has two knobs: voltage and current. These act as limiters, i.e. it prevents the current or voltage from going over a threshold. This can be used to safely power on the lamp. The following steps, while a bit long, ensures you never go over 8 mA while still getting a clear signal on the oscilloscope.

- Start by powering on both the pulse laser and the oscilloscope. The oscilloscope should be triggering.
- Hide channel two and show only channel one. Assuming the power supply is offline (it should be), you will see a dead line.
- *Before turning on the power supply*, turn both voltage and current knobs all the way to zero (counter clockwise).
- Turn on the power supply. You may see things on the oscilloscope, but disregard them.
- While keeping the current knob all the way to zero, increase the voltage until you see the ammeter jolt up to a non-zero value. This will happen at around 250 volts, maybe higher. When the lamp is under a high enough voltage, the discharge happens in the lamp. You will see a yellow glow inside. Once the lamp is ignited, the voltage will significantly drop. It may now also be reduced to an even lower value.
- The ammeter should show a low value of current, around 2 mA. If this value is over 8 mA, dial the current knob on the power supply all the way to zero. If the current is still over 8 mA, shut everything down and ask the technician for help.
- On the oscilloscope, you should see a large oscillatory motion. If you don't see it, zoom in to about 20 mV.
- This oscillation is caused by the current limiting feature of the power supply. Slowly (*very slowly*) turn the current knob until the ammeter reads between 5 and 6 mA.
- Reduce the voltage until you read 4 mA on the ammeter to maintain the discharge, then increase the current knob again back to 6 mA. Repeat these steps until you have a clear signal on the oscilloscope, without any large oscillatory motion.
- What we now have is a DC current that is not under the current limiting effect of the power supply. By turning the voltage knob, set the current to 6 mA. Be careful however: increasing the voltage wil allow you to go over 8 mA.

Unless you're actively using the lamp, it would be better to keep it turned off to increase it's lifetime. Turning the power supply off then back on will probably not have enough voltage to cause the discharge to happen without increasing the voltage. In order for an ionization to happen in the lamp, the voltage required will incur a much higher current than 8 mA. Therefore, every time you power up the lamp, please follow the steps provided in this section.

4 Experiment

The optogalvanic effect can be studied in a variety of manner. It can be used for spectroscopic purposes, or to study energy transitions of materials. This section will often call for currents that may go over the prescribed maximum of 8 mA for the lamp. Such high currents will damage the lamp if left on for extensive periods of time. Whenever required, please quickly make your measurements at high currents and refrain from letting the lamp powered on for too long.

4.1 Calibration

Once the apparatus is set up, you will want to sweep the dye module's micrometer and search for peaks on the oscilloscope. Peaks can have an amplitude of 5mV to 30mV. Note the micrometer value and the intensity of each peak. For that last task, you may want to use the average function on the oscilloscope. On the top of the oscilloscope, press "acquire", select the number of averages, then push the "average" option on the screen. This will average the last few entries, and make it much easier to read. If you want to revert back to non-averaged view, press "sample" in the same menu. If you want to go back to regular options, press "display" on the top of the oscilloscope. Once you swept through all the micrometer length, you can compare your data to the values seen in appendix F, and extract a relationship between the micrometer and the emitted wavelength. This sweep should be done at around 6 mA, where most of the peaks encountered will be from the neon in the lamp.

4.2 Calibration verification

To check if your calibration is good, we will compare the reading of a sodium discharge tube with the laser output. Put up the spectrometer and connect it to the computer. The spectrometer is the small black box with the fiber optic cable. Open the SpectraSuite program on the computer; it should work right away. Point the end of the fiber optic cable to the fluorescents in the room to see a bunch of peaks appear.

Now, turn on the sodium discharge tube, and let it heat up for a minute or two. Use the spectrometer to find the biggest peak. The spectrometer may be uncalibrated, hence why we use the sodium tube to have an idea of where to look. Once you know where are the sodium lines on the spectrometer scale, you can check your calibration scale. Find what micrometer reading would correspond to the sodium doublet (589-590nm), then set the dye module to it. Use the spectrometer to see if it's in the same area as the discharge tube reading. Even though the spectrometer is useful, it is not nearly precise enough to be used as the main reading tool in this experiment.

4.3 Optogalvanic signal

You may have noticed that different currents will yield differently shaped voltage curves as you studied the peaks for calibration. As seen in section 2.1, the voltage curves follow a behavior similar to a sum of real exponentials. Pick a well-defined peak seen in your calibration list, and change the applied current. Note the differences in the shape of the curves, and attempt at explaining them.

The oscilloscope you are using can output pictures straight to a computer. Connect it with the RS 232 cable to the computer, and start the *OpenChoice* program. You may want to take a screen capture, or output the data to get a clearer/simpler image. In either case, using the average function again to maintain a clearly defined curve would be useful. You can also skip that last step by drawing yourself the shape of the curves. Of course, computer-assisted drawings would be preferred. You may want to use piecewise continuous functions, or simply a sum of exponentials to redraw the curves. More simply, you could note the coordinates of 5 to 10 points on the curve that capture all the features of the curve, and then fit a smoothing spline on it. The applied current can be anywhere between 1 and 10 mA. Do not exceed 10 mA.

4.4 Sodium lines

The big problem with sodium lines is that they require a massive voltage on the lamp to be seen on the oscilloscope. Currents of 15-17 mA are required, while manufacturer's recommended maximum is 8 mA. Needless to say, looking for the sodium lines is very damaging for the lamp. You will want to minimize as much as possible the uptime on the lamp at such voltage. The sodium lines are routinely observed around 300v (read on the power supply) for this apparatus.

Since the currents will be well over 10 mA, you need to **disconnect the ammeter** before proceeding with high voltage. The ammeter is an analog component, and will be damaged by that kind of current. Use the short in the orange box.

Using the relation you found in the calibration section, estimate where is located the sodium doublet, and perform a sweep of the area. This needs to be done in a consistent fashion – you will want to pick a value reasonably under the doublet location, then increment the micrometer by a consistent amount each time. Note that a division will probably be too large; a third, a quarter or even a fifth of a division would be much more appropriate. Once you start incrementing, do not turn the micrometer in the other direction. This is to prevent backlash in the equipment – when slightly dialing back the micrometer, the presence of slack in the gears of the dye module will have the internal component turn less than what is actually read on the micrometer.

You will want to sweep through both spectrum lines, and extend a bit beyond to give a firm "anchor point" for your fit to rely on later on. Note the voltage amplitude at each increment point. Again, the use of the average function on the oscilloscope will greatly increase the precision of your readings. Remember that the average function takes the average of the last n triggers – you will need to wait for n pulses to happen after you modified the micrometer value in order to have an average made exclusively of your new wavelength, and can note down the amplitude.

Given the clarity of your data, fitting two Gaussians separately may prove enough to obtain a clear picture. If you so desire, you may also use a sum of two Gaussians and fit the whole dataset. This may give you better results if you have very clear data, especially between the two peaks. You may want to use the same variance for each peak and see how the data fits, since the variance should be similar. Use your judgment when deciding which method to use. Remember that having few points on your peaks will make it harder for the fitting routine to properly fit that area, especially with multiple parameters to play on. **Again, please, do not leave the lamp powered on for long times**, especially at such high voltages. It will damage the lamp over time.

4.5 Linewidth measurement

The acquisition of the natural linewidth of the nitrogen laser is one of the applications of optogalvanic spectroscopy. Pick one of the larger peaks you encountered while calibrating with a 6 mA current, and apply the same consistent sweeping described in the previous section. Note the different amplitudes as you go through the voltage peak. After fitting a Gaussian, you can extract the linewidth of the distribution, which should be slightly higher than that of the nitrogen laser, but within one order of magnitude.

5 Goals

To summarize, here are the goals of the experiment:

- Observe the optogalvanic effect for both neon and sodium lines.
- Determine the wavelength seperation of the sodium doublet using the optogalvanic effect.
- Observe the relaxation rates of optogalvanic signals at different currents.
- Determine the linewidth resolution of certain peaks
- Expand on the experiment however you can. You may want to try to observe different areas of the electromagnetic spectrum by using a dye different than Rhodamine 6G. If you desire to use another dye, please ask the technician for help.

References

[1] J. E. M. Goldsmith & J. E. Lawler, "Optogalvanic spectroscopy", *Contemporary Physics*, 1981, 22:2, p. 235-248.

[2] R. Shunker, A. Ben-Amar & G. Erez "Inverted population observation using optogalvanic effect", *Optics Communications*, 1982, 42:1, p. 29-33.

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