# STUDIES OF RF-OPTOGALVANIC LASER SPECTROSCOPY.

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LRAP - 41

Diploma paper at the department of physics
Lund institute of technology
Lund, Sweden 1985

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# INVESTIGATIONS OF RF-OPTO-GALVANIC LASER SPECTROSCOPY.


## ABSTRACT

Employing a single-mode dye laser acting on an atomic gas in a rf discharge the technique Doppler-free OptoGalvanic Spectroscopy (OGS) has been compared with POLINEX (POLarization INtermodulated EXcitation) and PS (Polarization Spectroscopy). During the course of the work it was also discovered that a frequency shift occured in the driving rf oscillator as the laser was tuned to a transition in the discharge medium. The possibilities of using this shift for detection of atomic resonances was briefly investigated.


## INTRODUCTION

After the rediscovery of the optogalvanic effect by Green et al. [1] in 1976, OGS has rapidly become a powerful tool for investigating atomic and molecular properties. There are some apparent advantages of rf OGS compared to optical methods. The technique is free from background stray-light and the oscillator can simultaneously be used both for driving the discharge and for detecting the signals [2]. Hollow-cathode discharges have been rather thoroughly examined [3-5], whereas there is less documentation about rf-discharge work. One advantage with rf as compared to hollow-cathode discharges is that the background noise caused by leak-currents between the anode and the cathode are avoided as the rf-cell does not have any electrodes inside. This also makes it possible to investigate chemically aggressive elements which tend to react with the electrodes as pointed out in Ref.[6]. The rf oscillator can also maintain a discharge at low gas pressure which minimizes pressure broadening. Doppler-free rf OGS measurements have been reported earlier [2] and it was decided to use a Colpitt oscillator of the same type as that used by Lyons, Schawlow and Yan [2]. The Doppler-free signals were obtained in neon at 588.2 nm and compared with signals obtained with PS and POLINEX [7].

EXPERIMENTAL SET-UP FOR RF-OGS.

The experimental set-up used in the present experiments is shown in Fig. 1. An argon ion laser (CR-12) is pumping a CW dye laser (CR 599-21) operating in a single cavity mode. The beam is split into two parts with a beamsplitter, the two beams are then mechanically chopped at two different frequencies and directed into a neon cell. A lock-in amplifier is employed to extract OGS signals on the reference frequency from the chopper. The lock-in signal is registered on the recorder as the dye laser frequency is scanned. The signal from a Fabry Pérot interferometer with a free spectral range of 50 MHz is connected to the recorder to supply a relative frequency scale.

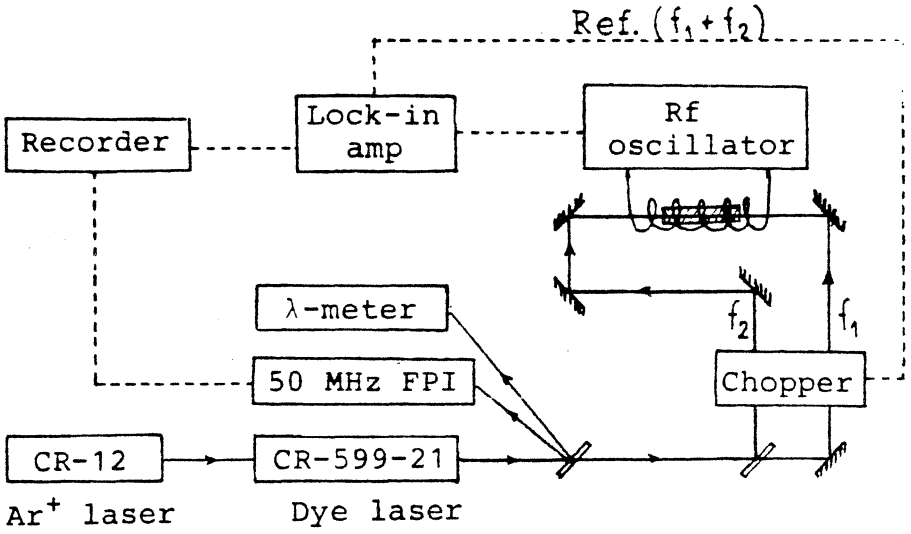


Fig.1 Experimental set-up for IMOGS. The best signals are obtained when the counterpropagating beams overlap perfectly in the cell.

A discharge in the neon cell is driven by a Colpitt oscillator with an oscillator frequency of about 10 MHz. The neon cell is placed in the 2.4 μH coil in the resonance circuit.

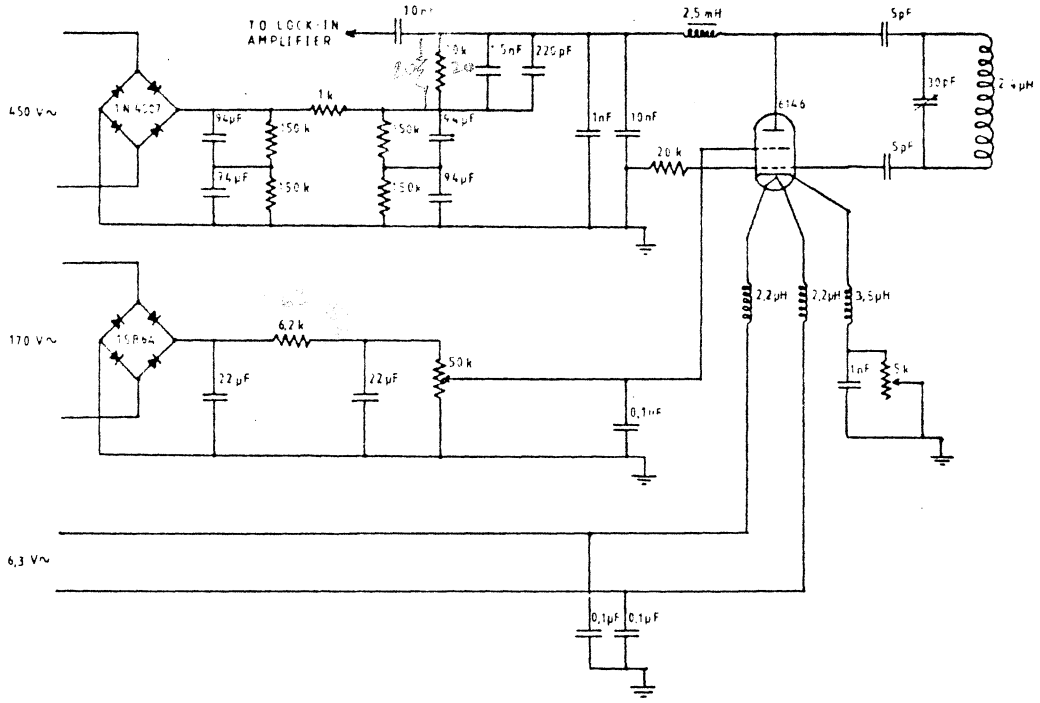


Fig.2 Circuit diagram of the Colpitt oscillator.

When the CW dye laser is in resonance with a neon transition the signal will appear as a voltage change over the 10 kΩ load resistor in the rf oscillator. The signal on the sum frequency $f_1+f_2$ is first picked out (among $f_1$, $f_2$, $f_1+f_2$ and $f_1-f_2$ where $f_1$=225 Hz and $f_2$=750 Hz) and amplified by the lock-in amplifier and then displayed on a paper recorder. The power of the rf oscillator could be regulated with the 50 kΩ potentiometer and the feed-back could be varied with the 5 kΩ potentiometer. The power needed to start the discharge varied between 20-50 W but after ignition only a few watts were needed for driving the discharge.

Measurements could not be done at high rf-power, because the disturbing rf field from the oscillator made the rest of the instruments in the laboratory go crazy. The best signals were often obtained when the discharge was close to cease. The feed-back factor was then very low which caused the oscillator to be more sensitive for a change of load in the resonance circuit. Fig. 3 (to the right) shows the output voltage from the lock-in amplifier as a function of the rf-power. One can clearly find a region where the signal to noise ratio is optimum.

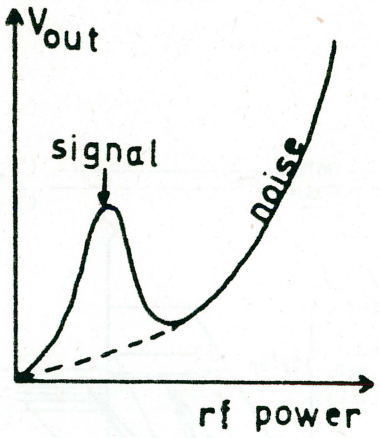


Fig.3 The variation of the lock-in signal when the rf power is swept manually by varying the 50KΩ potentiometer. The signal is recorded with the experimental set-up in fig.12. The dye laser is in resonance with the transition $1.5s_5$-$2p_2$ (Paschen notation) in neon and the chopper frequency is 975 Hz.

Three cells were made of Pyrex glass (length 70 mm and diameter 20 mm) with end windows of quartz were filled with neon to a pressure of 3, 0.6 and 0.12 torr respectively. At first the cells were evacuated and baked out in an oven at 400-500 °C for 10 hours. After this they were placed in the rf-coil and while the pressure of neon increased from 0.01 to 100 torr the fluorescence from the discharge was measured at 588.2 nm with a photomultiplier and a spectrograph. For this line the strongest fluorescence signals were obtained at a pressure of 3 torr. As could be expected the 3 torr cell also gave the best signal-to-noise ratio for the transitions in the 3s-3p array.

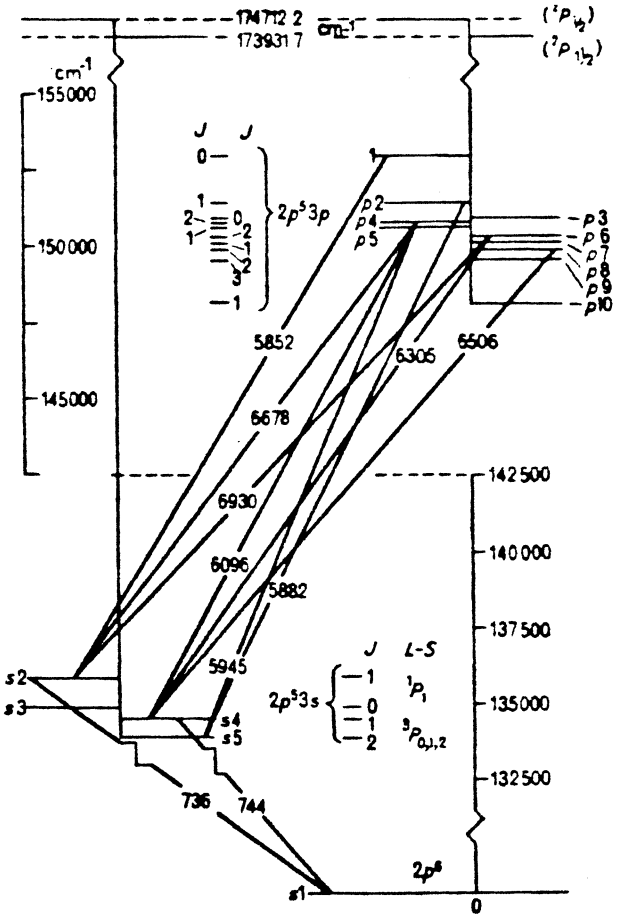


Fig. 4 Energy-level diagram showing the ground state and the two lowest excited configurations in neon [9].

Neon atoms with their high lying metastable states are excited and ionized by the rf field. The equilibrium between ionized and neutral atoms will change when the laser is tuned to an atomic transition. For picturing the impedance change in the discharge in a simple way one can say that the ionization of the Ne atoms will increase if the laser is exciting atoms from a metastable state to a higher lying state which is more easily ionized by the rf field. Another more sophisticated point of view is that when the atoms have been excited, super elastic collisions between electrons in the discharge and the excited state atoms will increase the electron temperature in the discharge. The energy is then rapidly transferred to all different species in the cell, thus modifying the impedance of the discharge [4]. The change of load for the rf oscillator can be detected either as a voltage change over the 10 kΩ resistor or alternatively as a change in the resonance frequency of the oscillator.

DOPPLER-FREE SATURATION SPECTROSCOPY

A laser beam is split up into two, the one with higher intensity being the saturating beam and the other the probe beam.

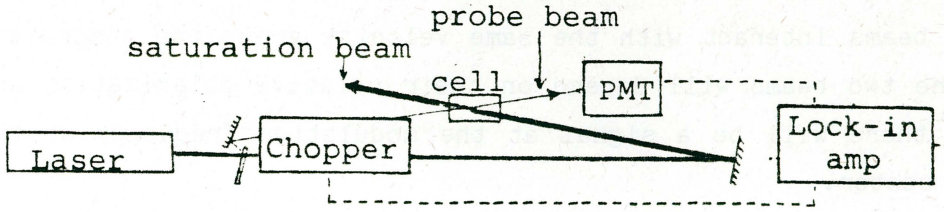


Fig.5. Experimental set-up for saturation spectroscopy.

The two beams are now sent through a cell from opposite directions and overlapped in the sample. The laser having a small linewidth interacts only with atoms of a specific velocity group. If the laser now is tuned close to resonance the counter-propagating beams will burn holes in the ground state velocity distribution according to fig.6.

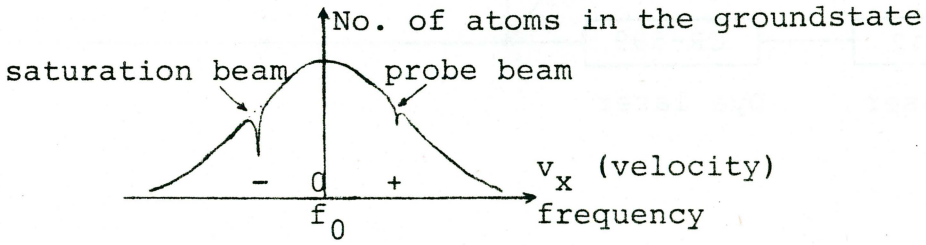


Fig.6. Ground state velocity distribution after "hole burning".

The two holes are due to atoms with the same velocity $v_x$ but travelling in opposite directions. The velocity causes a Doppler-shift ($\delta f$) equal to the difference between the photon energy ($f_\lambda$) and the energy corresponding to the atomic transition for a zero velocity atom ($f_0$).

$$\delta f = f_\lambda \cdot v_x \div c \qquad \text{where} \qquad f_0 = f_\lambda + \delta f$$

That is, when the laser is swept over the transition there will be a narrow signal at the line center since the absorption of the probe beam will be less as the two beams interact with the same velocity group ($v_x \approx 0$) at this specific frequency and the saturating beam saturates the transition. I.e. it excites a non negligible percentage of the ground state atoms in the $v_x=0$ velocity subgroup to the upper state, thus making the sample more transparent to the probe beam.

POLINEX.

The Pockels cell (see experimental set-up below) modulates the beam between left hand and right hand circular polarization. When both beams interact with the same velocity group the absorption of the two beams will depend on their relative polarization and thus there will be a signal at the modulation frequency at the line center.

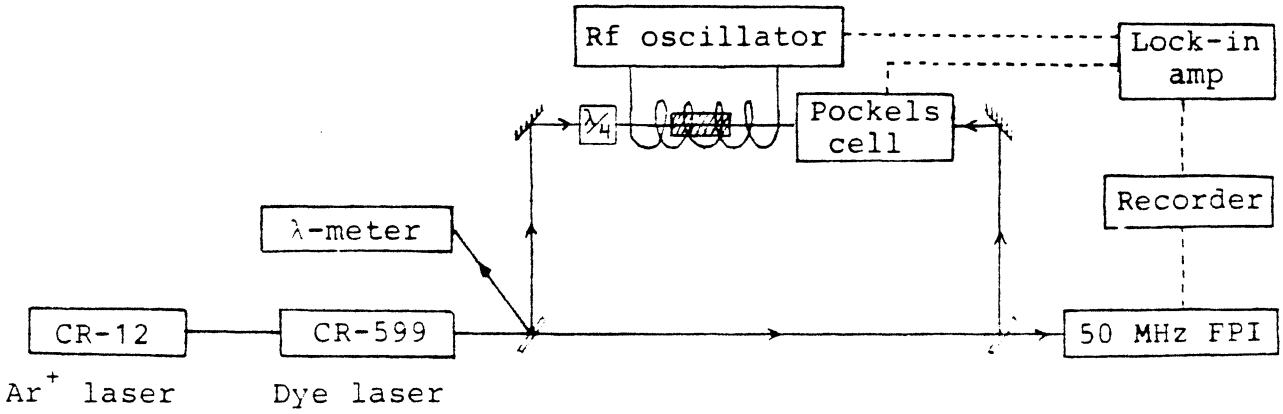


Fig.7. Experimental set-up for POLINEX.

POLARIZATION SPECTROSCOPY.

The saturating beam (having 80-90 % of the laser power) is circularly polarized to orient the atoms in the sample. This induced optical anisotropy is probed by the counter-propagating linearly polarized beam. The linearly polarized probe beam can be described as a superposition of a left and a right hand circularly polarized wave. At the line center where the two beams interact with the same atoms, this balance will be disturbed by the oriented atomic sample and part of the probe beam will pass through the second polarizer at 90° angle with respect to the first.

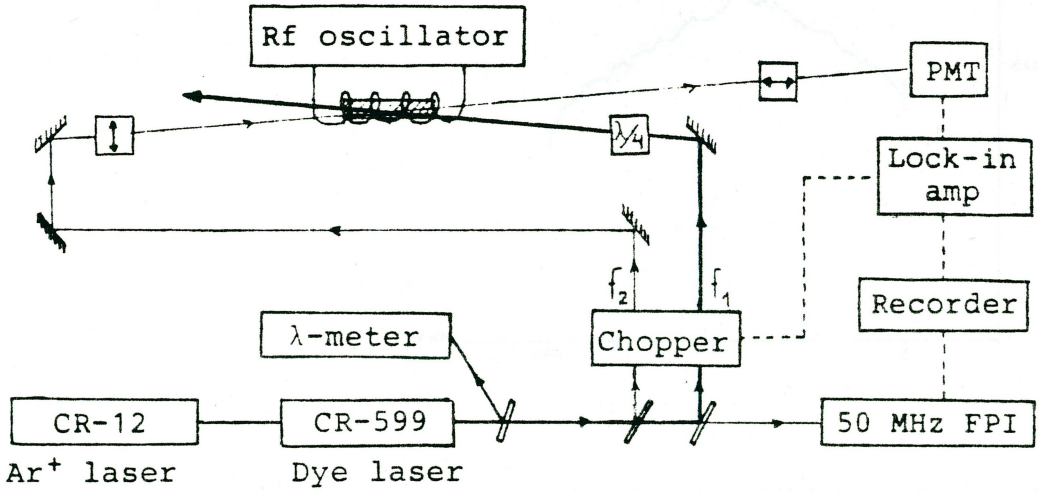


Fig.8. Experimental set-up for PS.

MEASUREMENTS AND RESULTS

To get an opinion of the possibility to obtain a good signal-to-noise ratio for Doppler-free signals with rf-IMOGS (Inter-Modulated OptoGalvanic Spectroscopy on a rf discharge) the method was compared with two other Doppler-free methods, rf-POLINEX and PS. All the figures (9-11) show the same neon line at 588.2 nm with the 0.12 torr cell, and they were recorded with a lock-in amplifier time constant of 1 sec.

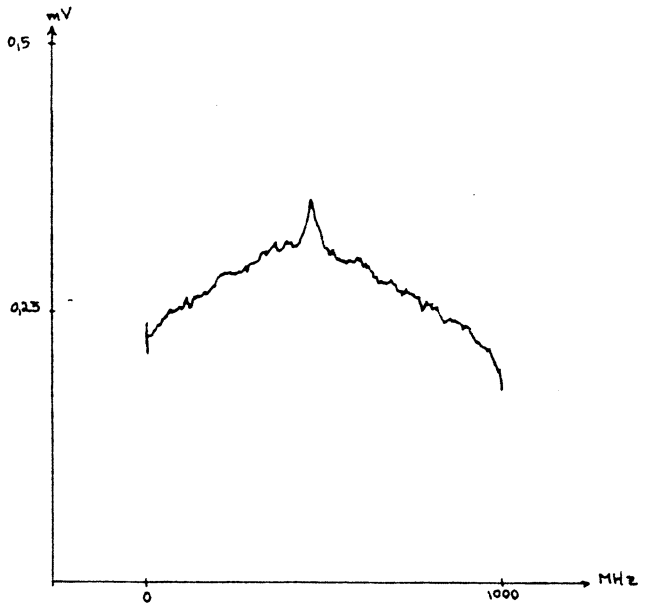


Fig.9. The rf-IMOGS signal with a peak due to $^{20}Ne$. Note the huge Doppler-broadened pedestal arising from velocity-changing collisions.

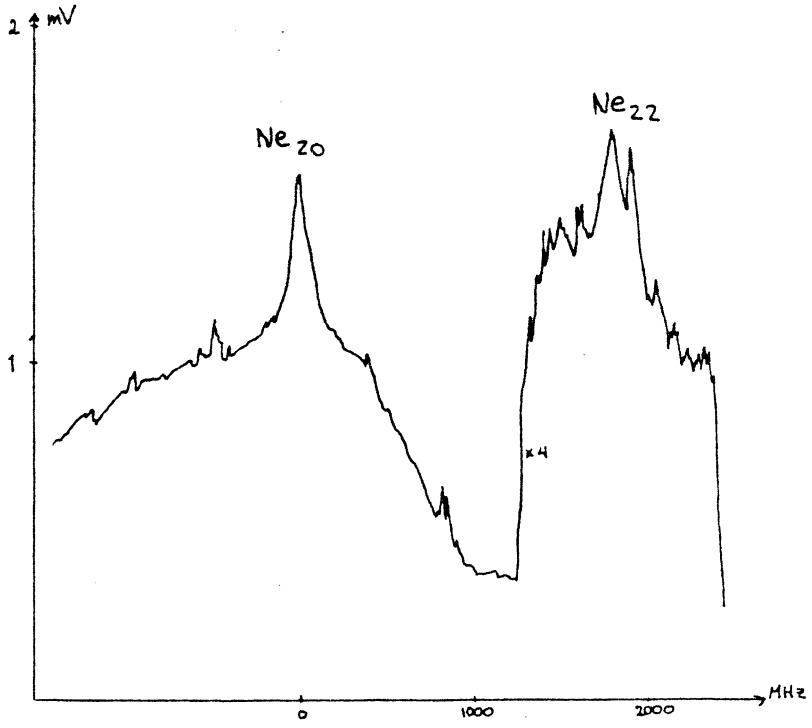


Fig.10. The signal obtained using POLINEX.

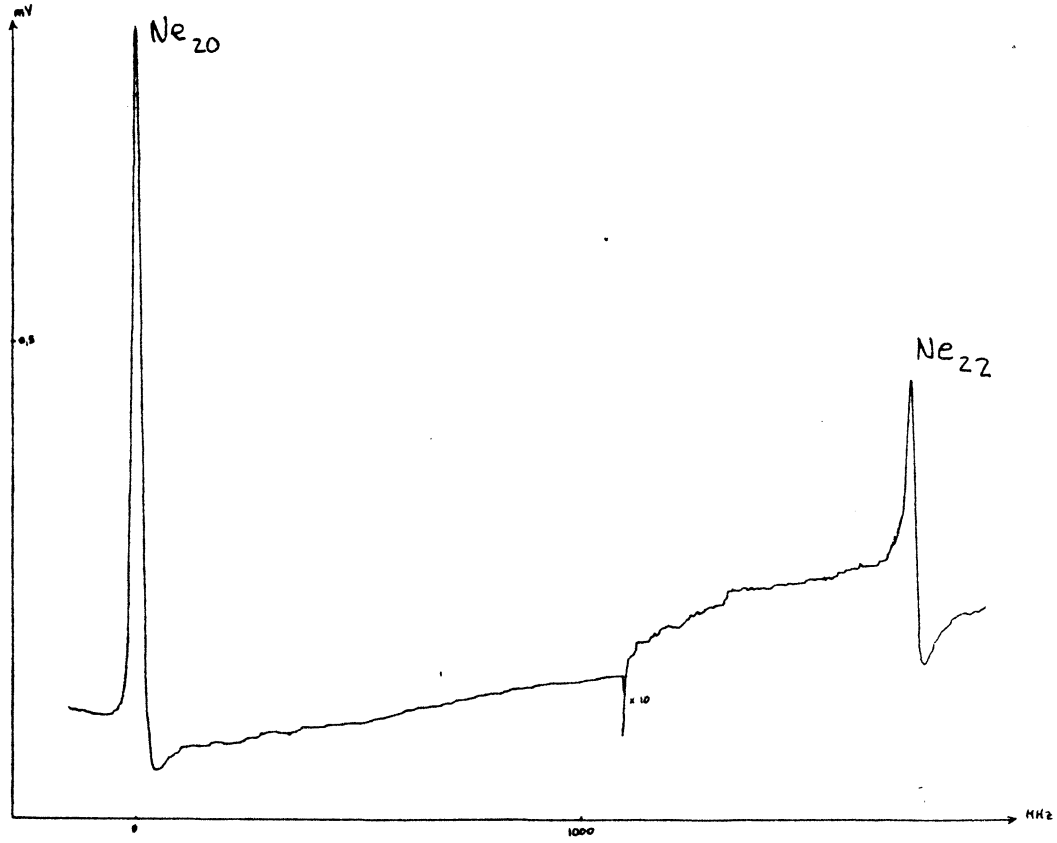


Fig.11. The signal when PS was employed. This obviously gave the best signal-to-noise ratio. The dispersion form of the PS signal appears as the polarizers are not perfectly crossed, which improves the signal-to-noise ratio [8].

## USING THE FREQUENCY CHANGE OF THE RF OSCILLATOR.

A to our knowledge new type of getting rf-OGE detection was also discovered and briefly investigated. The rf-field from the oscillator was picked up by a (crystal oscillator) reciever tuned to a frequency $f_c$ close to the frequency of the rf oscillator $f_{rf}$. A signal with a frequency corresponding to the difference between the two oscillator frequencies $|f_{rf}-f_c|$ was generated by the reciever and measured with a digital frequency counter. When the laser is scanned through a neon transition the plasma temperature and the number of ionized species change and thus the oscillation condition in the rf oscillator resonance circuit changes resulting in a change of load (it is also possible to regard it as a change of inductance see below). Thus when we scan the laser the frequency of the signal with frequency $|f_{rf}-f_c|$ will have a maximum or minimum at the line center.

## THEORY

To find a mathematical description of what is happening let us assume that the frequency change depends on a change in inductance. The resonance condition for the rf oscillator circuit can be derived to give us the desired expression:

$$\omega = \frac{1}{\sqrt{LC}} \quad \Rightarrow \quad \frac{d\omega}{dL} = -\frac{1}{2L\sqrt{LC}} = -\frac{\omega}{2L} \quad \Rightarrow$$

$$\Rightarrow \quad \frac{\Delta L}{L} = -\frac{2\Delta\omega}{\omega} = -\frac{2\Delta f}{f} \quad \because$$

One might alternatively regard the change as a change in load for the oscillator [10]. Thus we make a simple equivalent scheme of the oscillator with the discharge consisting of a resistance (R) in series with an inductance (L). Where R changes as the laser is tuned to the atomic transition.

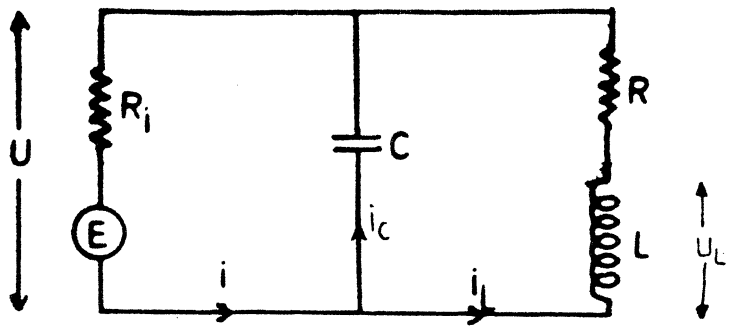


Fig.10 Equivalent scheme of the rf oscillator.

The following four equations can be obtained by using Ohm's and Kirchoff's laws.

$$i = i_L + i_C \tag{1}$$

$$E = (i \cdot R_i + U) = i \cdot R_i + i_L(R + j\omega L) \tag{2}$$

$$U = i_C/j\omega C = i_L(R + j\omega L) \Rightarrow i_C = i_L(j\omega RC - \omega^2 LC) \tag{3}$$

$$U_L = i_L \cdot j\omega L \tag{4}$$

Use (1) to eliminate i in (2).

$$E = (i_L + i_C)R_i + i_L(R + j\omega L)$$

$i_C$ can be expressed in terms of $i_L$ by using (3).

$$E = i_L(R_i + j\omega R_i RC - \omega^2 R_i LC + R + j\omega L)$$

And finally using (4) we can get an expression with no dependence of the currents.

$$E = \frac{U_L}{j\omega L} \cdot (R_i + j\omega R_i RC - \omega^2 R_i LC + R + j\omega L)$$

The only possibility for E to get the same (or opposite) phase as $U_L$ (a criterion for resonance) is when the imaginary parts cancel.

$$R_i - \omega^2 R_i LC + R = 0 \qquad \omega^2 = \frac{R+R_i}{R_i LC}$$

$$\omega = \omega_0\sqrt{1 + R/R_i} \quad \text{where} \quad \frac{1}{LC} = \omega_0^2 \quad \text{is constant}$$

Differentiation of $\omega(R)$ results in.

$$\frac{d\omega}{dR} = \frac{\omega_0}{2R_i} \cdot \frac{1}{\sqrt{1+R/R_i}} = \frac{\omega}{2(R_i+R)} \quad \Longrightarrow$$

$$\frac{\Delta R}{R+R_i} = 2 \cdot \frac{\Delta\omega}{\omega} = 2 \cdot \frac{\Delta f}{f}$$

Using the simple equivalent scheme above for modelling the rf cirquit we have shown that the relative change in frequency is directly proportional to the change of load (inductance) induced by the laser radiation.

EXPERIMENTAL SET-UP.

The broadband rf-OGE signal is monitored on the lock-in amplifier to indicate when the laser is tuned to the neon transitions. When we had maximum signal strength on the lock-in the crystal oscillator was tuned near the rf oscillator frequency with an offset of approximately 500 Hz and then the laser was blocked manually and the frequency on the counter was noted with the beam blocked and unblocked. The difference between the two frequencys gives the resulting signal. When the laser is tuned off resonance the frequency shift disappears.

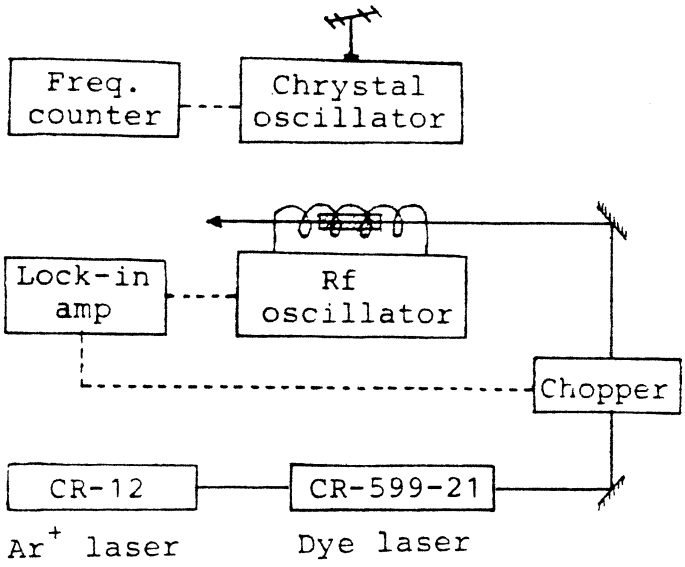


Fig.12. Experimental set-up for measuring the frequency shift of the rf oscillator. The chopper frequency was 975 Hz and the laser beam was blocked for 2-5 s. depending on how fast the frequency counter would sample.

RESULTS

The neon transition $1.5s_5$-$2p_2$ (588.2 nm) was investigated with the dye-laser running in a single cavity mode. Ten measurements were done for three different laser powers given in table 1 below.

| laser power | frequency shift | standard deviation |
|---|---|---|
| 2 mW | 100 Hz | 11 Hz |
| 10 mW | 180 Hz | 62 Hz |
| 20 mW | 430 Hz | 17 Hz |

Table 1.

This possibly indicates that the signal strength increases in a nonlinear way with increasing laser power (at these low powers). It was also noted that the best signals were obtained when the rf-oscillator had low feedback (i.e. operating at low power). The shift was now measured for some neon transitions with a laser power of 20 mW but with lower accuracy.

| transition | wavelength (nm) | frequency shift (Hz) |
|---|---|---|
| $1.5s_5$-$2p_2$ | 588.2 | 480 |
| $1.5s_5$-$2p_4$ | 594.5 | 670 |
| $1.5s_5$-$2p_5$ | 597.5 | 200 |
| $1.5s_5$-$2p_6$ | 614.3 | 900 |
| $1.5s_4$-$2p_2$ | 603.0 | -100 |
| $1.5s_3$-$2p_2$ | 616.3 | 140 |

Table 2. Observed frequency shifts in neon with the experimental set-up in Fig. 12.

A further study of the above described effect is outside the scope of this diploma paper. However further investigations will be made shortly. In particular the potential of this technique in atomic spectroscopy or other possible applications will be explored. The method might also help to give some insight in the physics of rf-discharges.

## CONCLUSION

We have not been able to get Doppler-free rf-OGS signals with enough strength to recommend it for atomic Doppler-free spectroscopy. Other Doppler-free techniques like polarization spectroscopy then seem more efficient. An interesting observation was that the resonance frequency of the rf oscillator changed when laser energy was absorbed in the sample cell. However, if this can lead to some fruitful results or developements remains to be seen.

ACKNOWLEDGEMENTS

This diplomawork would never have been completed if it were not for the help of some people at the Lund Institute of Technology department of Physics. I especially would like to thank my supervisor Stefan Kröll who never failed to solve my problems, professor Sune Svanberg who inspired me to the work and taught me the necessary atomic spectroscopy, Åke Bergkvist who helped me with the electronic equipment and recomended the measurement of the frequency shift, and the people at the mechanical workshop.

I am also most grateful to my fiancée Ewa Jönsson for encouraging me and patiently listening to me trying to explain what I was doing all the late nights at the laboratory.

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