Plasma Control of the Emission Spectrum of Mercury-Noble-Gas Discharges

PROEFSCHRIFT

ter verkrijging van de graad van doctor
aan de Technische Universiteit Eindhoven,
op gezag van de Rector Magnificus, prof.dr. M. Rem,
voor een commissie aangewezen door het College
voor Promoties in het openbaar te verdedigen
op donderdag 7 december 2000 om 16.00 uur

door

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geboren te Terneuzen
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This research is supported by the Technology Foundation STW and Philips Lighting Eindhoven, The Netherlands.

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ISBN 90-386-1669-4
NUGI 812
Trefw.: gasontladingen / fluorescentielampen / plasmadiagnostiek / kleur
Subject headings: discharges electric / fluorescent lamps / plasma diagnostics / colour

Drukwerk: Universiteitsdrukkerij Technische Universiteit Eindhoven
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1 Introduction

1.1 Colour temperature and its influence on humans
Lighting has a strong psychological impact on people. Even relatively small colour variations as different shades of white can have a significant influence. Since white light resembles the radiation of a blackbody at a certain temperature, we can use this (thermodynamic) temperature as a measure for the colour. This temperature is called the colour temperature. It expresses the shade of white in a quantitative way. People tend to perceive light as warm when its colour temperature is low, whereas light with a high colour temperature is perceived as cold.\(^1\) An example of this perception of warmth is a candle. It gives a warm feeling and its colour temperature is 2000 K, which is relatively low. Knez studied the influence of colour temperature and luminous flux on the mood of male and female subjects performing memory and problem-solving tasks.\(^2\) He found, among other things, that the preservation of positive and negative moods is depending on the colour temperature. Besides this, the intellectual performance also depends on the colour temperature. A remarkable difference has been found between male and female subjects: a positive mood is best preserved in males at 3000 K, while this mood is best preserved in females at 4000 K. These studies demonstrate that there is a need for efficient lighting systems of which the colour can be changed.

1.2 Fluorescent lamps and colour variation
Fluorescent lamps owe their popularity to their high efficiency in converting electrical energy into light. The plasma in fluorescent lamps has been studied extensively, both experimentally and theoretically.\(^3\)-\(^6\) The efficacy of a fluorescent lamp, i.e. the efficiency corrected for the sensitivity of the human eye, is at least three times higher than the efficacy of an incandescent lamp. Part of the reason for this high efficacy is the fact that the spectrum of a fluorescent lamp can be given almost any shape. The choice of the fluorescent layer determines the shape of the spectrum. This is very useful, since the spectrum can be shaped in such a way that useless radiation is omitted. The huge amount of invisible radiation, which is produced by an incandescent lamp, is part of the cause of its low efficacy. Besides this, the light for which the sensitivity of the human eye is relatively low, like red light, can also be partly omitted from the spectrum of a fluorescent lamp, increasing the efficacy of the lamp. On the other hand, the green part of the fluorescent lamp spectrum can be emphasised. This increases the efficacy since the human eye is very sensitive for green light.

The spectrum of a fluorescent lamp is normally adjusted for the environment in which the lamp is supposed to be used. For instance, the colour of lamps used for office lighting is different from the colour of lamps used in civic and social interiors. Museums need better colour rendering than industrial halls. Butchers need more red light in order to show their customers a healthy piece of red meat than social interiors. For all these examples, white light is needed. However, the shade of white differs for different applications.

Once the fluorescent layer is deposited, the colour of the fluorescent lamp is fixed. It cannot be changed anymore. However, for some applications, it would be convenient to change the colour temperature within the range of 2700 to 4000 K during operation. Changing the properties of the plasma in a fluorescent lamp offers the opportunity to change the emission spectrum of the lamp.
1.3 State of the art.
Variation of the colour of discharge lamps attracted the attention of the lighting industry during the 80’s of the last century. Pioneering work has been performed in the group of professor Itatani in Kyoto, Japan.\textsuperscript{7-10} Discharge lamps operating in a continuous wave mode turned out to emit light with a different spectrum than the same lamp operated in a pulsed mode. The explanation of this effect was found in the changing properties of the electron gas. Decreasing the pulse width and frequency increased the electron temperature.\textsuperscript{9} As a result, the emission from the higher energy levels increased. During the last decade, many novel techniques to change the colour of discharge lamps have been introduced.\textsuperscript{11-15} The research was mainly technology driven. It therefore concentrated on the control of the emission spectrum of the discharges, not on the underlying discharge processes. However, for the understanding of the important discharge processes, a more thorough investigation of the discharge is necessary.

Most of the discharges proposed for the use as a variable colour lamp are based on changing the properties of the electron gas in a fluorescent lamp. In a fluorescent lamp, the positive column of a mercury-noble-gas discharge converts the electrical energy into radiation. The resonant lines of mercury in the ultraviolet (UV) region dominate the emission spectrum of the discharge. Excitation of the noble-gas atoms requires much more energy than excitation of mercury. The emission of noble-gas lines is negligible, since the amount of electrons capable of exciting a noble-gas atom is very small. Selectively adding noble-gas emission to the discharge emission spectrum can be used to control the emission spectrum.

One way of changing the electron properties locally in such a way that noble-gas radiation is produced is igniting a second discharge in the discharge tube. The electrons in this second discharge are supposed to have a different energy distribution than the electrons in the main discharge. A capacitively coupled radio frequency (ccrf) discharge is suitable for this purpose. The electron energy in the sheath region of this discharge is higher than the energy of the electrons in the main discharge of the fluorescent lamp.\textsuperscript{16,17} Therefore, the emission of the ccrf discharge may contain much more noble-gas radiation than the emission of the main discharge. Capacitively coupled radio frequency discharges have been studied extensively in the past. Many studies are devoted to the electrical characterisation of these discharges,\textsuperscript{18-21} since the discharge impedance contains valuable information on the discharge.

The research on sodium- and caesium-noble-gas discharges as performed in the past suggests another way of changing the properties of the discharge in such a way that noble-gas radiation can be produced. Research by Van Tongeren showed that the metal-atom density in these discharges can be controlled locally by either changing the metal pressure, or by using the process of radial cataphoresis.\textsuperscript{22} Radial cataphoresis is the radial transport of atoms by a discharge. The emission spectrum of a metal-atom-noble-gas-discharge is dependent on the metal atom density in the discharge tube. This is demonstrated very clearly during the warm-up period of the low-pressure sodium-neon discharges used for street lighting. When the glass tube of these discharges is cold, the sodium pressure is low and the emission has a red colour characteristic for neon radiation. However, when the tube temperature is increased, the yellow sodium lines start to dominate the neon lines until at a certain temperature, the neon emission is decreased to a negligible level. The metal atom density, the properties of the electron gas, and other properties of the sodium- and caesium-noble-gas discharges have been studied by Van Tongeren\textsuperscript{25} and Cornelissen.\textsuperscript{23} Some data on radial cataphoresis in mercury-noble-gas discharges is also reported in the literature.\textsuperscript{24,25}

In conclusion, there are several options for the colour (temperature) variation of discharge lamps. A promising option is selectively adding noble-gas emission to the emission
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The research performed in the past on the two options for the addition of noble-gas emission to the discharge spectrum, the additional ccrf discharge and radial cataphoresis, was either application driven or did not concentrate on the production of noble-gas radiation. In contrast, the investigation described in this thesis does concentrate on the production of noble-gas radiation and the underlying discharge processes in mercury-noble-gas discharges.

1.4 Research goal and the chosen approach

This thesis deals with the investigation of the plasma control of the emission spectrum of mercury-noble-gas discharges. We are especially interested in the control of the amount of noble-gas radiation produced by these discharges. The main goal of the research is to obtain a better understanding of the processes involved in the production of noble-gas radiation in mercury-noble-gas discharges. The two most important questions to be answered are:

1. Which discharge processes and conditions are important for the production of noble-gas radiation in mercury-noble-gas discharges?
2. How can we produce this noble-gas radiation in an efficient way?

In order to answer these questions we performed an experimental investigation of two mercury-noble-gas discharges. This investigation comprises another important goal as well: the improvement of diagnostic techniques.

The starting point of the investigation was a hybrid discharge as invented by Itatani et al.\textsuperscript{12} This lamp is basically a standard fluorescent lamp, containing a positive column discharge with an additional capacitively coupled radio frequency discharge. We performed an optical and electrical characterisation of this hybrid lamp. For the electrical characterisation, we developed a new stray impedance characterisation technique. The electrical and the optical characterisation gave us more insight in the processes that are important for the production of noble-gas radiation in the hybrid discharge. Especially the influence of the mercury pressure on the emission spectrum of the discharge drew our attention.

The influence of the mercury pressure on the spectrum as found in the hybrid discharge, in combination with the potential of the radial cataphoresis process, led us to the introduction of the depleted discharge lamp\textsuperscript{†}. In this lamp, the mercury depletion due to the radial cataphoresis process is used to control the amount of noble-gas radiation produced by the discharge. The perspective for commercialisation of this depleted discharge lamp clearly was better than for the hybrid discharge lamp. Therefore, the research was narrowed down to the investigation of the radial cataphoresis process, with the emphasis on the properties of the electron gas and the emission spectrum. The depleted discharge was first studied by optical emission spectroscopy, electrical measurements, ultraviolet absorption measurements, and spatially resolved emission measurements. Combining the results leads to a rather complete picture of the discharge. However, apart from the information on the high-energetic electrons that can be obtained from the emission measurements, we need the electron density and temperature to complete this picture of the discharge.

For the determination of the electron density and temperature, we had to choose a diagnostic technique. In the past, electron density and/or temperature measurements in fluorescent lamps and other metal-vapour-noble-gas discharges were performed using

Langmuir-probes,\textsuperscript{26-29} microwave techniques,\textsuperscript{30} absorption spectroscopy,\textsuperscript{31} laser absorption,\textsuperscript{32} Stark shift measurements,\textsuperscript{23} UV absorption by mercury ions,\textsuperscript{33-35} and other techniques. The main disadvantage of all these techniques is that they are either intrusive, spatially integrating or necessitating the use of theories based on certain assumptions for the discharge or the measurement. Unlike the techniques mentioned above, incoherent Thomson scattering is non-intrusive, it can be used spatially resolved, and the interpretation of the measurements is straightforward. Therefore, Thomson scattering is preferred above the other techniques to measure the electron density and temperature.

The main problem of Thomson scattering is the low intensity of the scattered signal. Using a powerful laser and a sensitive detector can increase the Thomson scattering signal to a detectable level. However, the intensity of parasitic scattering will also increase with the laser power. This is especially a problem in a fluorescent lamp, where the electron density is low (in the order of $10^{18}$ m$^{-3}$), and the parasitic scattering (stray light) intensity is high. The Thomson scattering signal is expected to be at least 6 orders of magnitude lower than the stray light.

Thomson scattering has been used extensively in fusion plasmas like Tokamaks.\textsuperscript{36-38} Furthermore, it has been used in relatively high-density plasmas as inductively coupled plasmas,\textsuperscript{39,40} and arcs.\textsuperscript{41-43} More recently, the technique has also been used for plasmas with an electron density down to $10^{17}$ m$^{-3}$.\textsuperscript{44,45} Substantial effort is made to lower the detection limit even further.\textsuperscript{46} Because of the high stray light level and the relatively low electron density, Thomson scattering has never been used to determine the electron density and temperature in a fluorescent lamp.

Redesigning the plasma-reactor using baffles and beam-dumps can reduce the stray light level.\textsuperscript{47} However, in fluorescent lamps this is not possible, since the plasma parameters will change when the discharge geometry changes. The only solution is the introduction of a notch-filter in the detection branch. This notch-filter should block the stray light (at the laser wavelength) while at the same time transmitting the Thomson scattering signal (Doppler-broadened because of the velocity distribution of the electrons). Interference filters are not suitable for this purpose because of their high spectral width. Therefore, a special notch filter has to be designed. One candidate is a double monochromator used in a dispersion-subtractive-dispersion arrangement.\textsuperscript{48-50} A second type of special notch filter is a ruby crystal, in combination with a ruby laser.\textsuperscript{51} Another promising candidate is an atomic notch filter.\textsuperscript{52,53} We chose to use a sodium vapour absorption cell. This cell should be able to completely suppress the stray light from the discharge tube, when Thomson scattering is performed at a laser wavelength of 589 or 589.6 nm, i.e. the resonant absorption lines of sodium.

In conclusion, an experimental investigation comprising Thomson scattering and other diagnostics is chosen to reach the goal and to answer the two main questions stated in the beginning of this section. The development of a novel Thomson scattering set-up is a major part of this investigation.

1.5 Thesis outline
Throughout the whole thesis, we will discuss the two variable colour discharge lamps mentioned above. However, the emphasis is put on the lamp based on radial cataphoresis. Chapter 2 describes colourimetry, the conventional fluorescent lamp and the two variable colour fluorescent lamps we studied: the hybrid discharge lamp and the depleted discharge lamp. The theory of radial cataphoresis and the resulting mercury depletion is discussed in detail. Chapter 3 describes the diagnostic techniques we used to investigate the discharge. Chapter 4 discusses the experimental results. Chapter 5 synthesises these results. The
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synthesis consists of cross-linking of separate results, a phenomenological description of the discharges, and a comparison between the two variable colour discharge lamps. The final chapter, i.e. chapter 6 sums up the most important conclusions that can be drawn from the results of the investigation.

1.6 Publications
The main part of this thesis has already been published in journal papers. This thesis is based on the following nine publications and two patents.


- L.P. Bakker, and G.M.W. Kroesen, “Method for adjusting the light spectrum of a gas discharge lamp, gas discharge lamp, and luminaire for said lamp”, International application published under the patent cooperation treaty (PCT), International publication number WO 00/16376, filing date 9 September 1999. Also: USA Patent application, 09/396563 filing date 15 September 1999.
2 Variable colour fluorescent lamps

2.1 Introduction
This chapter briefly describes the theory of colourimetry, the conventional fluorescent lamp and the two types of variable colour fluorescent lamps we investigated. We will discuss the working principle, the gas mixture, the electrodes, the fluorescent powders and the geometry of fluorescent lamps. The emphasis of this chapter is on the variable colour fluorescent lamp based on mercury depletion. The process of radial cataphoresis is discussed, and some useful analytical expressions describing this radial cataphoresis and the resulting mercury depletion are derived. We will start with the discussion of colourimetry.

2.2 Colourimetry
Colour is closely related to the vision of human beings. It is used to describe the appearance of a visible stimulus with a certain spectral shape. The human eye uses three different kinds of cones to detect visible radiation in colour: one for blue, one for green, and one for red light. In this way, the spectral shape of a visible stimulus is converted into three physiological signals. Obviously, information is lost, since an infinite number of spectral shapes is represented by only three numbers, defined as the tristimulus values. Therefore, different spectra can have the same colour. The tristimulus values can be calculated using colour-matching functions, which implicitly contain the relative sensitivity of the three cones in the human eye. In order to calculate the tristimulus values, the spectrum should be multiplied with the corresponding colour-matching function, and subsequently integrated over the whole wavelength range for visible radiation. The colour of a spectrum is assumed to be independent of the intensity. Therefore we can normalise the total value of the tristimulus values to unity. The result of this normalisation is that we can represent the colour of a spectrum using just two quantities: the chromaticity coordinates. A diagram containing these two chromaticity coordinates is called a chromaticity diagram. The most frequently used chromaticity diagram is the one defined by the Commission International de l’Eclairage (CIE) in 1931. In this diagram, the chromaticity coordinates are defined as x and y.

Figure 2.2.1 shows the chromaticity diagram, along with the three colour-matching functions \( \Xi(\lambda) \), \( \Upsilon(\lambda) \), and \( \Xi(\lambda) \). In the chromaticity diagram, the x and y values of the monochromatic stimuli are plotted. Furthermore, figure 2.2.1 shows the colour of the monochromatic stimuli and the colour of the mixtures of these stimuli. We note that in the CIE 1931 system, the colour-matching function \( \Upsilon(\lambda) \) is equal to the overall sensitivity of the eye for visible radiation. Due to limitations in the reproduction process, the colour in figure 2.2.1 is only an approximation of the real colour. It will also vary with the illumination of this page. In figure 2.2.1, we also plotted the colour of a special source of radiation: the blackbody. A blackbody at a certain temperature emits radiation with a spectrum described by Planck’s radiation law. When the temperature of the blackbody is high enough, it will start emitting visible light. In figure 2.2.1 the position on the blackbody locus is marked with the temperature. At a temperature of 1000 K, the colour of this radiation is red, at 4000 K it is white and at higher temperatures it starts becoming blue. Therefore the temperature of a blackbody can be used as a measure for the colour of the emitted radiation. Extending this idea for a lamp with the same chromaticity coordinates as a blackbody at a certain temperature, we can use the temperature of the blackbody as a measure for the colour of the lamp. The colour temperature of a lamp is defined as the temperature of the blackbody with the same colour. Furthermore, when the colour of a lamp is not exactly on, but close to the
blackbody locus in the chromaticity diagram, the term correlated colour temperature is used. For the sake of clarity, we also use the term colour temperature for the correlated colour temperature. For a more detailed discussion on colourimetry, see Wyszecki et al. For a physiological discussion on human vision, see Kaiser et al.

Lamps should have a colour resembling the colour of a blackbody. This ensures that the emitted light is perceived as white. In general, the colour temperature of fluorescent lamps ranges from 2700 to 4000 K. The colour temperature of a lamp can be changed by adding light with a different colour to the emission spectrum. When the light of a lamp with a colour temperature of 4000 K lamp is mixed with light with a wavelength of 590 nm, the colour temperature of the resulting radiation shifts from 4000 to 2000 K. This is clear in figure 2.2.1: the colour of the mixture is positioned on a straight line between the positions of the colours of the 4000 K stimulus and the 590 nm stimulus. The colour of a neon discharge resembles the colour of a blackbody at 1000 K. So by adding neon radiation to a 4000 K fluorescent lamp, the colour temperature of this lamp can be decreased. We note that by using the colour temperature concept, we first decreased the number of quantities necessary to describe the colour from three (tristimulus values) to two (chromaticity coordinates), and finally to one (colour temperature).

### 2.3 Conventional fluorescent lamps
In a conventional fluorescent lamp, the positive column of a low-pressure mercury-noble-gas discharge converts approximately 70% of the electrical power input into ultraviolet radiation. The electrical energy is first converted into kinetic energy of the free electrons in the discharge. The electrons then collide with mercury atoms, which are excited to a higher energy level. After some time, the excited atoms lose their energy by spontaneous emission of an ultraviolet quantum. The ultraviolet radiation is then absorbed by a fluorescent layer, which is deposited on the inner wall of the discharge tube. This layer converts the absorbed ultraviolet photon into a visible photon. This whole process of generation of visible radiation from electrical energy is schematically drawn in figure 2.3.1.
Variable colour fluorescent lamps

Fig. 2.3.1: Schematic drawing of the working principle of a fluorescent lamp.

The electrical current is injected into the discharge by means of two electrodes that are positioned at the ends of the discharge tube. The electrodes provide the electrons necessary to maintain the discharge. The electrodes are covered with a mixture of the oxides of barium, strontium and calcium. These materials are known for their relatively high electron emission, even at low temperatures as 800°C. The electrodes emit the electrons by means of thermionic emission. The electrodes are incorporated in an electric system, which contains a stabilising device, a ballast. Stabilisation is necessary since the voltage-current characteristic of the discharge has a negative slope.

The discharge tube is filled with a mixture of noble gases and mercury, the latter being present in both liquid and vapour form. The pressure of the noble-gas mixture is a few hundred Pa, the mercury vapour pressure is approximately 1 Pa. This is the vapour pressure at a temperature of 42°C. Although the noble-gas pressure is much higher than the mercury pressure, the discharge is dominated by mercury. Virtually no excitation and ionisation of the noble-gas atoms occurs. This is due to the fact that the energy of the excited states of the noble-gas atoms is much higher than the energy of the excited states of mercury. This is clearly illustrated in figure 2.3.2, which shows the energy schemes of mercury and neon.

Fig. 2.3.2: The energy schemes of the atoms of mercury (left) and neon (right). Note the different energy scales.

Compared to the energy of the excited states, the electron temperature is rather low. This results in the presence of much more inelastic collisions of electrons with mercury than with neon. For example, in a Maxwellian electron energy distribution with a temperature of 1 eV, $9 \times 10^4$ times more electrons are involved in inelastic collisions with mercury than with neon. Moreover, the amount of high-energy electrons is reduced by the inelastic collisions with mercury. This depletion of high energetic electrons favours the inelastic collisions with
mercury with respect to the inelastic collisions of electrons with neon atoms. Concluding, we can assume that, despite the fact that we have two or three orders of magnitude more rare-gas atoms than mercury atoms, the discharge is dominated by mercury. We will only have mercury ions and hardly any rare-gas ions in the discharge.

The noble gas serves three functions. First, the noble gas decreases the mean free path of the charged particles. Second, it reduces sputtering and evaporation of the emitter material by reducing the ion bombardment of the electrodes. The last function of the inert gas is providing easier breakdown.

The fluorescent layer consists of three phosphors. Each phosphor has a characteristic emission spectrum. By mixing them in the right amounts, the desired colour can be achieved. In this way the lamp colour can be in the range from warm-white (2700 K) to cool-white (4000 K). However, once the fluorescent layer is deposited on the tube wall, the colour can not be changed anymore. For this special purpose, i.e. colour variation during operation, the variable colour fluorescent lamp is needed. We note that a detailed discussion of electric discharge lamps can be found in the book of Waymouth.57

2.4 Variable colour fluorescent lamps

There are several ways to change the colour of a fluorescent lamp. A simple solution is to use two differently coloured lamps and vary the relative power input to the two lamps. This idea can be extended, resulting in one discharge tube with two discharge paths, differing in colour.11,14 We can also make use of the fact that the decay time of phosphors can differ significantly.13 The most common way to change the colour of a fluorescent lamp is changing the electron energy distribution function (eedf) in the lamp. This changing of the eedf is usually done by using sophisticated electrical driving schemes like pulsing or ramping.8-10,15 The emission of the lamp depends heavily on the eedf, so changing the eedf results in a change in the lamp colour. Another option for colour variation is introducing a second discharge in the lamp, for instance a capacitively coupled radio frequency discharge.12 This second discharge locally modifies the eedf. Some lamps use the emission spectrum of the noble gas,8-10,12 while in other lamps mercury is the only emitting species.15

We will concentrate on the addition of noble-gas radiation to the emission spectrum of fluorescent lamps. Two noble gases are candidates for the use in a variable colour fluorescent lamp. The first one is neon, since addition of its red light to the emission of a cool-white fluorescent lamp results in a warm-white colour. The second one is xenon because of its strong ultraviolet resonance line at 147 nm. This line can be used to excite a selective phosphor. The choice of the phosphor now determines the shift in colour of the fluorescent lamp. So xenon offers more freedom in the choice of the colour shift.

We investigated two options for the addition of noble-gas radiation to the emission spectrum of mercury-noble-gas discharges. First, we studied the lamp with the additional capacitively coupled radio-frequency discharge, the hybrid discharge lamp. Subsection 2.4.1 discusses this hybrid discharge lamp in more detail. Second, we developed a new type of variable colour fluorescent lamp. The colour variation in this lamp is also based on the addition of noble-gas radiation to the emission spectrum of the lamp. The mercury density in the discharge is varied in order to change the emission spectrum of the lamp. Subsection 2.4.2 discusses this type of variable colour fluorescent lamps in detail.
2.4.1 The hybrid discharge

The hybrid discharge consists of a conventional fluorescent lamp, as described in section 2.3, with an additional capacitively coupled radio-frequency (ccrf) discharge. There are two main differences between the conventional fluorescent lamps and the hybrid discharge lamps. These differences are:

- In the hybrid discharge lamp neon is used as a buffer gas whereas in the conventional tubes a mixture of noble gases is used.
- In the hybrid discharge lamp an additional capacitively coupled radio frequency (ccrf) discharge is implemented.

The working principle of this lamp is as follows. In the electrode regions of the ccrf discharge, the electrons can gain enough energy to be able to excite and ionise neon atoms. The subsequent spontaneous emission of neon atoms results in the production of red light. By adjusting the power dissipated in the ccrf discharge, the amount of red light can be controlled, and the colour of the lamp can be changed.

The ccrf discharge is struck between two electrodes that are attached to the outer surface of the discharge tube. The driving frequency of the ccrf discharge is 13.6 MHz. At this frequency, the discharge tube acts as a capacitor, so a displacement current can flow through the tube wall. For the measurements, two kinds of electrodes were used. For the impedance measurements, brass electrodes were used. For the optical measurements, aluminium tape was attached to the glass surface. The advantage of the brass electrodes is the fact that the lamp and the electrodes could be separated. The advantage of the aluminium electrodes is the good mechanical contact between glass and electrodes, which minimises stray impedance effects. The length of the electrodes is 55 cm, their width is 5 mm. The distance between the electrodes is 5 mm for the aluminium and 25 mm for the brass electrodes. Figure 2.4.1 shows a cross-section of the hybrid discharge lamp.

![Fig. 2.4.1: A schematic drawing of the tubular fluorescent lamp, with the two brass electrodes at the outer glass surface of the tube (1), and the cross section of the lamp. The neon radiation (2) is produced near the electrodes. The mercury radiation (3) is produced in between the electrodes.]

An impression of light emission pattern is also plotted in figure 2.4.1. This pattern is observed by visual inspection of a lamp without the fluorescent layer. The neon light is produced only in the regions near the ccrf electrodes. In a lamp with fluorescent powder coating on the inner glass surface, the red light pattern looks uniform over the whole tube. This is because of the fact that the fluorescent powder coating is a diffuse reflector.

2.4.2 The depleted discharge

Getting rid of the mercury atoms in the mercury-noble-gas discharge seems to be the most straightforward way to make the discharge produce a significant amount of noble-gas radiation. There are two ways to change the mercury density in the discharge. First, the overall mercury pressure can be controlled by controlling the temperature of the coldest spot of the discharge. This implies that the temperature has to be controlled at the position where liquid mercury is present. Second, we can change the mercury density by introducing radial cataphoresis. Cataphoresis is defined as the partial segregation of gas components that occurs...
when a gas mixture is subjected to an electrical discharge. In other words, the gas discharge can control the mercury density. Especially radial cataphoresis is suitable for the control of the mercury density in a mercury-noble-gas discharge.

**A. Radial cataphoresis**
The electron-ion pairs in the positive column of a mercury-noble-gas discharge are created by ionisation in the bulk and lost due to ambipolar diffusion to the walls where they recombine. The result is a net flux of mercury from the plasma region to the walls. This phenomenon is called radial cataphoresis. We note that due to the low electron density, bulk recombination can be neglected. The net flux of mercury to the walls results in a depleted mercury density profile. The depletion of mercury will increase until the resulting diffusional flux of mercury atoms from the walls back into the plasma region compensates the ambipolar flux. The positive column of a low-pressure mercury-noble-gas discharge is a non-equilibrium plasma, which means that the ambipolar diffusion coefficient is much higher than the diffusion coefficient of neutral atoms. Therefore, the diffusion of mercury atoms from the wall into the plasma region is much slower than the ambipolar diffusion of mercury ions and electrons in the opposite direction. The result is that although the mercury ion density is much lower than the mercury atom density, the depletion of mercury can be significant. Radial cataphoresis in sodium- and caesium-noble-gas discharges has been studied extensively in the past. This phenomenon is also well known in mercury-noble-gas discharges.

We can increase the mercury depletion by either lowering the mercury pressure, or increasing the electron/ion density. The first option is based on the fact that less mercury has to be transported to the walls in order to have a significantly depleted mercury density profile. We note that in general, a decrease in the mercury density will not lead to a decrease in the electron/ion density, so the ambipolar flux will not decrease. The second option is based on the fact that at a higher electron/ion density the ambipolar flux of ions and electrons will be higher. The electron density can be increased in a very simple way: by increasing the electrical current through the lamp.

**B. Mathematical description**

We can derive some useful expression from the mass-balance of mercury. The mass balances for mercury atoms and mercury ions are

\[
\frac{\partial n_{\text{Hg}^0}}{\partial t} = -n_{\text{Hg}^0} n_e k_{\text{ion}} + \nabla \left( D_0 \nabla n_{\text{Hg}^0} \right),
\]

and

\[
\frac{\partial n_+}{\partial t} = +n_{\text{Hg}^0} n_e k_{\text{ion}} + \nabla \left( D_a \nabla n_+ \right),
\]

respectively. In these expressions, \( D_a \) is the ambipolar diffusion coefficient, \( n_+ \) is the mercury ion density, \( D_0 \) is the diffusion coefficient for mercury atoms, \( n_{\text{Hg}^0} \) is the mercury atom density, \( n_e \) is the electron density, and \( k_{\text{ion}} \) is the ionisation reaction coefficient. We assumed the particle temperatures to be independent of position. In the stationary situation, both expressions are equal to zero. Adding them up results in the following expression

\[
\nabla \left( D_a \nabla n_+ \right) = -\nabla \left( D_0 \nabla n_{\text{Hg}^0} \right).
\]

The gradient of the ambient flux to the wall is equal to the gradient of the flux of neutral mercury from the wall. We also assume the diffusion coefficients to be independent of position. Then expression 2.4.2 results in

\[
n_+ (r) = \frac{D_0}{D_a} \left[ n_{\text{Hg}^0} (R) - n_{\text{Hg}^0} (r) \right],
\]

(2.4.3)
where we assumed the mercury ion density at the wall to be equal to zero, i.e., we have total recombination at the wall. The mercury density at the wall is fixed. We assume the wall to be the coldest part of the discharge, the cold spot. Expression 2.4.3 can be rewritten in
\[
n_+(r) = k \cdot \mu_0 \cdot T_0 \cdot T_e \cdot \left[ n_{H_g} (R) - n_{H_g} (r) \right],
\]
(2.4.4)
where we used the Einstein relation for the diffusion coefficients, \( T_0 \) is the heavy particle temperature expressed in Kelvin, \( T_e \) is the electron temperature expressed in eV, \( \mu_+ \) is the mercury ion mobility, and \( \mu_0 \) is the mobility of neutral mercury. We now define the degree of mercury depletion
\[
\chi \equiv \frac{n_0 (R) - n_0 (0)}{n_0 (R)} = e \cdot \frac{\mu_+}{\mu_0} \cdot T_e \cdot n_+(0),
\]
(2.4.5)
where \( p_{H_g} \) is the mercury pressure at the wall, as determined by the wall temperature. We note that the presence of temperature gradients can also result in depletion of mercury. This depletion due to temperature differences is also present in the noble-gas atom density profile.

It is clear from expression 2.4.5 that an increase in the mercury ion density or the electron temperature will result in a higher degree of depletion. On the other hand, when the mercury pressure is high, then the degree of depletion will be low. In physical terms, the first conclusion results from the fact that a higher ambipolar flux has to be compensated by a higher degree of depletion. The second conclusion results from the fact that at higher mercury pressures, the amount of mercury on the tube axis is high in comparison with the amount transported to the walls. The degree of depletion is not an explicit function of all the relevant discharge parameters. The degree of depletion is an implicit function of discharge parameters like the current and the noble-gas pressure. Their influence is felt in the electron density and the electron temperature.

In order to investigate the trends in the onset of the production of noble-gas radiation, we assume that the production of noble-gas radiation occurs in the situation where we have an almost completely depleted mercury density profile, i.e. \( n_{H_g} (r) = 0 \). This assumption is justifiable when we realise that the current through the discharge without noble-gas ionisation is limited to a critical value \( I_c \). This is due to the fact that the electron density is limited when we assume to have only mercury ions, as can be concluded from expression 2.4.4: the mercury ion density is at its maximum when the mercury atom density is zero, locally. The current can only be larger than the critical value \( I_c \) when apart from the mercury atoms also noble-gas atoms are ionised. The expression for the critical current can be obtained by substituting \( n_{H_g} (r) = 0 \) in expression 2.4.4, assuming the electron density to be equal to the mercury ion density, and using the following expression for the current density in the positive column
\[
j = e \cdot n_e \cdot \mu_e \cdot E,
\]
(2.4.6)
where \( j \) is the current density, \( n_e \) is the electron density, \( \mu_e \) is the electron mobility, and \( E \) is the electric field in the positive column. Note that we neglect the ionic contribution to the current. This can be done since the ion mobility is much lower than the electron mobility. The resulting expression for the critical current is
\[
I_c = \pi R^2 \cdot \frac{\mu_0}{\mu_+} \cdot p_{H_g} \cdot \frac{\mu_e \cdot E}{T_e},
\]
(2.4.7)
where \( R \) is the inner radius of the discharge tube. The mercury contribution to the electron mobility is negligible at low mercury pressures, the electron mobility can thus be written as
\[
\mu_e = \frac{\xi}{p_{Ne}}.
\]
(2.4.8)
where $p_{Ne}$ is the noble-gas pressure expressed in Pa, and the value of $\xi$ is equal to the electron mobility at a noble-gas pressure of 1 Pa. Expression 2.4.7 can be rewritten in

$$I_c = \pi R^2 \frac{\mu_0}{\mu_e} \xi \frac{p_{He}}{p_{Sc}} \left( \frac{E}{T_e} \right).$$

(2.4.9)

The ratio of the electric field and the electron temperature is the only unknown quantity in this expression. From measurements done by Verweij in an argon-mercury positive column, we can conclude that this ratio remains constant within 20%, while varying the mercury pressure. This means that the critical current is approximately proportional to the mercury pressure at the wall. On the other hand, the ratio of the electric field to the electron temperature is not constant while varying the noble-gas pressure. Therefore, we cannot assume the critical current to be inversely proportional to the noble-gas pressure.

C. Power modulation

For the application of the depleted discharge as a light source, there is one complication. Increasing the current in order to change the colour of the lamp will also result in an increased luminous flux produced by the lamp. This is due to the fact that the power deposited in the discharge increases with increasing current. However, we can de-couple the discharge current and the dissipated power by modulating the amplitude of the discharge current. Amplitude modulation (AM) allows us to control the current independently from the dissipated power. The simplest way of amplitude modulation is turning the discharge on and off and repeating this sequence with a certain modulation frequency. During the time that the discharge is off, the mercury atoms will diffuse back into the centre region of the tube. The depletion of mercury will decrease. When the discharge is turned on again, the mercury will be transported back to the walls. This transport takes some time. Assuming that noble-gas radiation can only be produced when the mercury density is low enough, we can conclude that during a fraction of this transport time, the discharge is not producing noble-gas radiation. So the time the discharge is on should be longer than the characteristic transport time. This determines the upper limit for the modulation frequency.

The characteristic transport time can be estimated using the mass balance for mercury ions and atoms, expression 2.4.1. Appendix A describes the derivation of a zero-dimensional time-dependent model for the transport of mercury. The resulting characteristic time for the transport of mercury is

$$\tau = \frac{\Lambda^2}{D_0} \left( \frac{2 n_{He}(\infty)}{n_{He}(R)} \right) = \tau_D \left( \frac{2 n_{He}(\infty)}{n_{He}(R)} \right),$$

(2.4.10)

where $\tau_D$ is the characteristic time for the diffusion of mercury atoms:

$$\tau_D = \frac{\Lambda^2}{D_0},$$

(2.4.11)

$n_{He}(\infty)$ is the mercury density at $t \to \infty$, and $n_{He}(R)$ is the mercury density at the wall, which is assumed to be constant in time. It is clear from expression 2.4.10 that when the resulting mercury atom density at the tube axis, at $t \to \infty$ is lower than half the density at the wall, then the cataphoresis process is faster than the ‘free’ diffusion of mercury.

The characteristic time for the diffusional transport of mercury is in the order of a few milliseconds, as can be calculated from the characteristic length for diffusion, and the diffusion coefficient of mercury. Therefore, the modulation frequency should be in the order of 100 Hz or lower. On the other hand, the lowest modulation frequency that can be used is approximately 75 Hz, since lower frequencies result in visible flickering of the discharge emission.
D. Phenomenological description and conclusions

We will end the discussion in this section with a simplified phenomenological description of the radial cataphoresis process and the subsequent onset of the production of noble-gas radiation. The expression for the degree of depletion (expression 2.4.5) shows that the ionisation degree of mercury is the main discharge parameter determining the shape of the mercury density profile. Therefore, a high discharge current and a high noble-gas pressure, both resulting in a high electron density, along with a low mercury density together result in a significantly depleted discharge. When the mercury is removed, less energy is dissipated by means of inelastic collisions with mercury. This results in a higher electron temperature, since the electron heating by the electric field will not decrease. Schematically, the following steps are assumed to be important:

1. High current or low mercury pressure,
2. High ionisation degree,
3. Significant mercury depletion,
4. Less inelastic collisions of electrons with mercury,
5. Increase of the electron temperature,

Concluding, the radial cataphoresis process will be guided by a decrease of the mercury density, an increase in the electron temperature, and the onset of the production of noble-gas radiation. The increase in the electron temperature will be accompanied by an increase of the electric field. The next chapter describes the experiments that are performed in order to observe the influence of radial cataphoresis on the quantities mentioned above.
3 Diagnostic techniques

3.1 Introduction
In this chapter we will discuss the diagnostic techniques we used to measure the relevant discharge quantities. We will start with the emission spectroscopy and the electrical measurements performed on the hybrid discharge. Then we will discuss the experiments on the depleted discharge: emission spectroscopy, electric field measurements, ultraviolet absorption measurements, spatially resolved emission spectroscopy, and incoherent Thomson scattering. We investigated the positive column of alternating current (AC) driven discharges, see appendix B. The discharge currents as reported in this chapter and the next chapters are root mean square values. Unless explicitly stated different, we measured time-averaged quantities in order to limit the time necessary to perform the experiments. The time-averaged measurements are supposed to result in time-averaged quantities like a time-averaged emission spectrum, time-averaged absorption and so on. For the absorption measurements and the Thomson scattering measurements, this is not as straightforward as it seems. For these measurements the implications of not measuring time-resolved are discussed in Appendix B. Besides the time-averaged measurements, the modulation of the emission spectrum and the electron density and temperature due to the non-zero driving frequency is studied by performing time-resolved emission measurements and Thomson scattering measurements, respectively.

3.2 Emission spectroscopy
This section describes the experimental set-up for the emission measurements. The emission spectrum of a discharge can give important information on the discharge. This is especially the case for variable colour discharge lamps. The set-up consists of a discharge tube with power source, a diaphragm with a diameter of 1 mm, a lens with a focal length of 77 mm, an optical fibre with its aperture in the focal plane of the lens, and an optical multichannel analyser. The distance between the lens and the diaphragm is 22 mm. The input cross section of the fibre is a circle with a diameter of 1 mm, the output cross section is a straight line of 4 mm length. Figure 3.2.1 shows a schematic drawing of the input optics. Using these optics, radiation is captured in a cone with a full-angle of $1.3 \times 10^{-2}$ rad. All the measurements are performed with the lamp within 50 mm of the diaphragm. The resulting spatial resolution is better than 2 mm in the plane perpendicular to the optical axis.

![Fig. 3.2.1: The input optics.](image)

The optical multichannel analyser consists of a detector interface (EG&G Princeton Applied Research 1461), and an intensified photodiode array (1420IR gated detector), mounted on a spectrograph (Acton Research Corporation SpectraPro 275). The fibre is connected to the spectrograph with an imaging input optics assembly (Acton Research Corporation model FC-459-180). The slit height is 4 mm, the slit width is 50 microns. In the spectrograph a 68:68 mm$^2$ grating with 150 lines/mm is used. The focal length of the
The spectrograph is 275 mm. The detector is cooled to –10°C in order to reduce the dark-current. It is connected to a 14 bits analogue to digital converter. The optical multichannel analyser can detect the whole visible spectrum from 400 to 775 nm in one go, using 1024 channels. The full width at half maximum of the apparatus profile of the system is 1.7 nm. The wavelength calibration is performed using a low-pressure mercury lamp. The transmission of the optics is measured by replacing the discharge tube with a calibrated tungsten filament lamp (Osram Wi 17/G) at a temperature of 2600 Kelvin. We used the data by De Vos for the emissivity of tungsten. 61

From the measured spectra, we calculated the \((x,y)\) values by multiplying the spectra with the colour matching functions as given by the CIE, 55 and by subsequent normalising. From the \((x,y)\) values, we calculated the correlated colour temperature \(T_c\).

In order to represent the colour of the lamp on the computer screen and on a printer, a series of calculations is performed. The three phosphors of the screen can be seen as primary stimuli of a new colour system: the screen-\((r,g,b)\) system. In order to transform \((x,y)\) values to the screen-\((r,g,b)\) system, the \((x,y)\) values of the red, green and blue phosphors of the screen have to be known. These values are shown in table 3.2.1 for the PAL television standard. 62

| Red   | 0.64 | 0.33 |
| Green | 0.29 | 0.60 |
| Blue  | 0.15 | 0.06 |

Table 3.2.1: The \((x,y)\) values of the red, green, and blue phosphors used to calculate the screen-\((r,g,b)\) values.

By mixing the light emitted by the three phosphors, we can have colour stimuli with a colour point positioned in between the straight lines connecting the three phosphor colour points. In figure 3.2.2 this principle is shown in the CIE 1931 diagram.

![Fig. 3.2.2: The stimuli of the screen phosphors in the CIE 1931 diagram.](image)

The screen-\((r,g,b)\) values each have an integer value from 0 to 255. They are normalised to the maximal intensity; the tristimulus values are multiplied by a constant in such a way that the highest tristimulus value is equal to 255. Because of the fact that no
negative tristimulus values can arise in the screen-(r,g,b) system, these systems cannot define all the possible colours. The screen cannot produce the colours with a colour point positioned outside of the triangle in figure 3.2.2. For this, the colour of the points outside the triangle have to be approximated. The approximation is that whenever a tristimulus value becomes negative, the corresponding screen r, g or b value is set to zero. In this way, the screen-(r,g,b) values of every visual stimulus can be calculated. With the calculated screen-(r,g,b) values, we can make a bitmap file (.BMP format). These bitmap files can be used in other Dos or Windows applications. The coloured figures in chapter 4 are produced in this way.

3.3 Electrical measurements hybrid discharge

Electrical measurements are relatively easy to perform. The results of these measurements give overall information on the discharge. The electrical measurements we performed can be divided into two categories, the low-frequency (lf) and the radio frequency (rf) measurements.

3.3.1 The low-frequency network

A schematic drawing of the low frequency network is shown in figure 3.3.1. Because of the fact that the lf discharge has a negative (V,I) characteristic, an impedance has to be placed in series with the lamp to make the total differential impedance connected to the lf generator positive. In the present lf network an inductance is used for this purpose. The lf generator and this inductance together are called the lf ballast.

In order to prevent the current from being rectified by the lamp, a capacitance (LCC 1000 pF, 20 kV) is placed in series with the lamp. The lamp can behave as a rectifier due to the fact that it is possible that one electrode gets a bit hotter than the other, which leads to a higher current through the hot one and a lower current through the cold one. The current difference amplifies the temperature difference, until one electrode is hot and the other is so cold that hardly any electron can be emitted from it. Then the current can flow normally in one direction, in the opposite direction no current can flow. The capacitance prevents this effect from occurring.

The current in the lf circuit is measured with a current probe (Pearson electronics inc. Wideband current monitor model 2877) at position 3 in the network (figure 3.3.1). This current probe is connected to a digital oscilloscope (Tektronix DSA 601 digitising signal analyser). The voltage at the ballast output (position 1) and just after the capacitance (position 2) are measured with high-voltage probes (Tektronix Tek P5100 100x) connected to the oscilloscope. The influence of the rf discharge on the lf network is minimised by using a transformer (2⋅27 mH) and placing two inductances (403 µH) in series with the lamp.
3.3.2 The RF network: Discharge impedance measurements

Impedance measurements are widely used as a diagnostic technique for radio frequency (rf) discharges. Its advantages are the simplicity of the instrumentation and the possibility to monitor the discharge impedance real-time and ex-situ. The main problem with rf impedance measurements is the characterisation of the stray impedances in the electrical circuit. The behaviour of an rf network is different from a low frequency network. The first reason for this is the effect of the time-dependent magnetic fluxes enclosed by an electric circuit. Usually, the time derivative of these fluxes is negligible for low frequencies. Then the contour integral of the electric field is equal to zero, according to Faraday’s law of electromagnetic induction. However, at high frequencies the time derivative of the enclosed magnetic flux cannot be neglected, therefore the contour integral of the electric field is not equal to zero, and Kirchoff’s voltage law is not valid anymore. Another reason for the different behaviour of high frequency networks is the skin effect for conductors. These high-frequency effects can be represented in the network by stray impedances.

There are several methods to measure the impedance of an rf discharge. The most common method is using current and voltage probes to determine the discharge impedance. The disadvantage of this method is the difficulty of finding a proper position for the electric probes, since the probes can influence the network. For our measurements, we use the fact that the impedance of an impedance-matched network is equal to the internal impedance of the power source. For this method to work, all the impedances in the network need to be known in order to derive the discharge impedance from the total impedance of the matched network, so also the stray impedances have to be characterised. When the stray impedances are known, then the discharge impedance can be obtained from the settings of the matching network in the matched situation. Commonly, one of two methods is used for the determination of the stray impedances. Firstly, the stray impedances can be measured with a network analyser. A second way to determine the four-terminal network behaviour is making use of known impedances instead of the discharge impedance. The stray impedances can then be determined from the total impedance in the matched situation. The known impedances are placed at the position of the discharge. The disadvantage of these two ways to characterise the stray impedances is the need for extra equipment. A network analyser and well-defined dummy loads are usually not present. The way to characterise the stray impedances presented here uses no extra equipment, unlike all the previously mentioned techniques. We do not need a network analyser, voltage and current probes, oscilloscopes and dummy loads. We only need a rf power source, a bi-directional power meter (or a SWR meter), a matching network and a plasma. This equipment is already present in a normal rf discharge set-up.

The impedance measurements are carried out with the set-up consisting of a 13.6 MHz rf power source (Hewlett Packard 8116A 50 MHz pulse/ function generator with ENI 3100 LA power amplifier), a coaxial cable with a bi-directional power meter (Bird series 4410 rf directional Thruline wattmeter), an L-type matching network with a fixed inductance and two variable capacitances, and a fluorescent lamp. The impedance representation of this set-up is shown in figure 3.3.2.
In this figure, the power flows are plotted also. The stray impedances are not plotted in this figure. The lamp is a conventional tubular fluorescent lamp with two additional brass electrodes attached to the outside of the glass tube as described in section 2.4.1. One electrode is connected to the inner and one is connected to the outer conductor of the second coaxial cable.

The determination of the stray impedances is based on the reflection of power at the matching network end of the coaxial cable (transmission line). The reflection coefficient for this process is a function of the impedance of the network terminating the coaxial line. A bi-directional power meter measures either the forward power \( P_F \), or the reflected power \( P_R \). The expressions for \( P_F \) and \( P_R \) are:

\[
P_F = \frac{V_F^2}{2R_0},
\]

and

\[
P_R = \frac{V_F^2}{2R_0} \left| \frac{Z - R_0}{Z + R_0} \right|^2,
\]

where \( V_F \) is the amplitude of the voltage wave travelling from the power source to the matching network and \( R_0 \) is the characteristic resistance of the coaxial line and \( Z \) is the impedance of the terminating network. The ratio of \( P_R \) to \( P_F \) is equal to

\[
\frac{P_R}{P_F} = \left| \frac{Z - R_0}{Z + R_0} \right|^2,
\]

which is a function of \( Z \). This means that we can get information of \( Z \) by measuring the power flows. If except for the stray impedances we know all the components in the network that terminates the coaxial line, then we can write down an expression for the impedance \( Z \). This expression will be a function of the stray impedances. The discharge impedance cannot be a part of this network, because it is dependent on the power the discharge dissipates. In our case, the output of the matching network is short-circuited, the rest of the network is disconnected. In this way, we can characterise the stray impedances in the matching network. The stray impedances can be represented as two-terminal or four-terminal networks. We have chosen to use only two-terminal networks in order to give the stray impedances a physical meaning. The reflection coefficient of the short-circuited matching network, terminating the coaxial line, can be expressed as a function of the stray impedances, the inductance and the capacitances. The terminating network is shown in figure 3.3.3. In this figure, the stray impedances are shown in dashed lines.
Fig. 3.3.3: The experimental set-up for the determination of the inductance $L$ and the stray impedances $R$ and $jb$. The stray impedances are plotted dashed.

The expression for the impedance of this network is

$$Z = \frac{Z_1 \cdot Z_2}{Z_1 + Z_2},$$

where

$$Z_1 = \frac{1}{j \omega C_1} + j \cdot b,$$

and

$$Z_2 = \frac{1}{j \omega C_2} + j \omega L + R.$$  

The unknown quantities in these expressions are $L$, $R$ and $b$. These quantities are determined using the following procedure:

1. The network is matched to the power source; i.e. the reflected power is minimised.
2. The settings of the two capacitances, $C_1$ and $C_2$ are recorded.
3. The value for $C_1$ is slightly adjusted, while keeping $C_2$ constant.
4. The forward power $P_F$ and the reflected power $P_R$ are measured.
5. The setting of the capacitance, $C_1$ is recorded.
6. The preceding three steps are repeated.

The capacitances are measured with an automatic RCL meter (Fluke PM 6303A). In this way, the power reflection coefficient is measured as a function of the capacitance $C_1$. The parameters of this function are the stray impedances $R$ and $b$, the inductance $L$ and the capacitance $C_2$. The reflection coefficient, as given by expression 3.3.3 is fitted to the measured values. The parameters of this fit are $R$, $b$ and $L$. In figure 3.3.4, this fit is plotted, along with the measured values for the power reflection coefficient.

Fig. 3.3.4: The measurements and the fit for $P_R/P_F$. 

The fit is good if $P_R/P_F < 0.2$. This is the situation in which the coaxial cable is almost properly terminated. In the matched situation, when the reflection coefficient is low, the voltage at any point in the cable is the same as the voltage at the end of the cable, except for a certain phase factor. The same holds for the current in the matched situation. When the cable is not properly terminated, this is no longer true. Parasitic effects in the coaxial cable are dependent on the position in the cable and cannot be taken into account by just assuming a ‘simple’ impedance terminating the cable. Therefore, the fit will only be valid at low reflection coefficients. The fit parameters, i.e. the extra impedances are:

\[
\begin{align*}
    b &= 5.249 \ \Omega, \\
    L &= 4.24 \ \mu\text{H}, \\
    R &= 0.6383 \ \Omega.
\end{align*}
\]

With this unique solution for the stray impedances, all components of the matching network are known. In the matched situation, the impedance $Z_a$ at the output terminals of the matching network is

\[
Z_a = \frac{Z_1 \cdot R_0}{Z_1 - R_0} - Z_2.
\]  

(3.3.7)

The discharge electrodes are connected to the matching network with a coaxial cable. This cable transforms the impedance of the discharge according to the following expression

\[
Z_a = R_0 \cdot \frac{Z_{\text{dis}} + j R_0 \cdot \tan(k \cdot l)}{R_0 + j Z_{\text{dis}} \cdot \tan(k \cdot l)},
\]

(3.3.8)

where $Z_{\text{dis}}$ is the discharge impedance, $k$ is the magnitude of the wave vector in the coaxial line ($k = 0.43 \ \text{rad} \cdot \text{m}^{-1}$), $l$ is the length of the line ($l = 0.65 \ \text{m}$), and $R_0$ is equal to 50 $\Omega$. Using the expressions 3.3.7 and 3.3.8, the discharge impedance can be calculated from the settings of the matching network capacitances.

In order to check the measurement method, the voltage between the electrodes is calculated from the settings of the matching network capacitances and the power dissipated in the network, with the discharge on. This voltage is also measured with a voltage probe on the electrodes. We note that in contrast to the discharge voltage, the discharge current is difficult to determine independent from the impedance measurements. Due to the high level of stray radiation from the electrodes a current probe will not work properly. Figure 3.3.5 shows the results of the two different discharge voltage measurements as a function of $P_F$.

---

![Graph](image)

Fig. 3.3.5: Comparison of the voltage probe measurements (line) with the voltage calculated from the impedance measurements with (black square) and without (circle) taking into account the stray impedances.
We also calculated the lamp voltage without stray impedance calibration, assuming \( b=0, R=0 \), and taking the value of \( L \), measured with the LCR meter at 1 kHz (\( L = 3.72 \, \mu\text{H} \)). This voltage is also shown in figure 3.3.5. It is clear that the addition of stray impedances in the impedance representation improves the impedance measurements. Having performed this stray impedance characterisation, we are able to measure the discharge impedance.

3.4 Emission spectroscopy and electric field measurements in the depleted discharge

In order to study the influence of mercury depletion on the emission spectrum and on the electric field, we performed emission measurements and electric field measurements in the depleted discharge. A 100 kHz power source (ENI Plasmaloc 1-HF) is used to sustain the discharge. The current through the discharge is measured with a current monitor (Pearson electronics inc. Wideband current monitor model 150). The discharge voltage is measured with a high voltage probe (Tektronix Tek P5100). The current monitor and the voltage probe are connected to an oscilloscope (Tektronix TDS 360 digitising signal analyser), which measures the root mean square value of the current and the voltage. The temperature of the discharge tube is controlled by a transparent water jacket. The temperature of the inner wall of the discharge tube at the position of the water jacket determines the mercury pressure in the tube. However, we can only control the outer wall temperature. Since there is some heat transport through the glass tube, the inner wall temperature will be somewhat higher than the outer wall temperature. This temperature difference is in the order of 0.1 to 2°\( \text{C} \), depending on the power dissipated in the discharge. The temperature difference can be estimated by assuming that the power deposited in the positive column minus the power radiated by the positive column as light flows through the glass as heat. Furthermore, we use Fourier’s law of heat conduction and the thermal conductivity of the glass. For the mercury pressure we used the vapour pressure curve given by Alcock \textit{et al.} The water jacket is connected to a thermostat bath (Haake FE). The water jacket is an 80 mm diameter perspex cylinder with brass flanges at the ends. A rubber O-ring is pressed around the discharge tube with a second brass flange. The water jacket is positioned in the middle of the discharge tube. The length of the jacket is approximately 4 cm less than the length of the lamp, so the lamp is almost completely covered by the jacket.

The optical system for the emission measurements is already described in section 3.2. The transmission of the optics is measured by replacing the discharge tube with a calibrated tungsten filament lamp (Osram Wi 17/G). After this calibration, the discharge emission is measured. We measured the emission originating from a small area of the phosphor coating through the transparent water jacket. The measured emission is corrected for the transmission of the water jacket. From the measured spectrum, we calculated the chromaticity coordinates \( x \) and \( y \), along with the correlated colour temperature \( T_c \), and the strength of some relevant spectral lines of mercury and neon.

For the determination of the electric field in the positive column, we use the fact that the positive column is homogeneous, i.e. the characteristics of the plasma are the same along the length of the positive column. So by measuring the voltage across the positive column, we can determine the homogeneous electric field in the column. However, the voltage across the lamp consists of the positive column voltage and the voltage drop in the electrode regions. By using two lamps of different length, we can separate these two components and determine the electric field in the positive column. We used lamps of 200 mm and 400 mm long. The length of the water jackets around these lamps is proportional to the length of the lamp. The electric field is obtained by subtracting the root mean square (rms) values of the two voltages,
and dividing this difference by the difference in column length. We can use the difference of the rms values, since the waveform of both voltages is the same.

### 3.5 Ultraviolet absorption measurements

The depletion of mercury can be observed indirectly from the addition of neon radiation to the spectrum of the lamp. To give direct proof of the fact that mercury depletion takes place, we have to measure the mercury density profile in the lamp. Mercury atoms in the ground state absorb UV radiation with a wavelength of 253.7 nm. Therefore, the absorption of this radiation by a discharge can be used as a probe for the mercury ground state density. In order to measure the density profile we built an ultraviolet (UV) absorption set-up. In this set-up, a neon-mercury discharge lamp confined in a quartz tube is used as a UV source. The source lamp is filled with 500 Pa neon† and 5 mg mercury. The current through this lamp is 400 mA. The radiation from the source lamp is guided through a 2.3 kHz optical chopper (Scitec instruments 300 CD) and two diaphragms of 1 mm diameter. The distance between these two diaphragms is 100 mm. The beam is then guided through a second neon-mercury discharge lamp. This probe lamp is also made of quartz, it is filled with 1000 Pa neon and 5 mg mercury. The cold-spot temperature is 18 °C. This temperature is controlled by a thermostat bath (Endocal refrigerated circulating bath model RTE-5DD) and two cooling fluid jackets adjacent to the beam. The length of the region in between the jackets was 8 cm. After passing the probe lamp, the beam is guided through another two diaphragms of 1 mm diameter. Then the 253.7 nm part of the beam is detected by a photomultiplier (Radio Corporation of America type 1P28), mounted on a monochromator (Oriel model 7240). The detected signal is fed into a lock-in amplifier (EG&G Princeton Applied Research model 5209), which discriminates the source radiation (chopped) from the emission of the probe lamp. The lock-in signal is plotted on a \((x,t)\) writer (Kipp&zonen BD112) in order to see the time evolution of the transmission through the probe lamp. Both lamps are operated using a standard 35 kHz ballast (Philips BRC411/01). Figure 3.5.1 shows a schematic drawing of the set-up. The positions of the probe lamp and the two diaphragms in front of the monochromator can be changed as indicated in figure 3.5.1 in order to measure the transmission at several heights.

![Fig. 3.5.1: Schematic drawing of the UV absorption set-up.](image)

The measurements are performed in the same way as Van Tongeren measured the Cs density in a Cs-Ar discharge.\(^{22}\) In order to calibrate the set-up, the transmission of the lamp at the lamp-axis is measured in the lamp-off situation for mercury densities ranging from \(6.4 \times 10^{18}\) to \(4.4 \times 10^{19}\) \(m^{-3}\). The temperature of the cooling fluid is varied in order to control the mercury density. The wall temperature at the position of the UV beam during the calibration

---

† The noble-gas pressures as reported in this chapter and the subsequent chapters are filling pressures at a temperature of approximately 25°C.
measurements was 21°C. The calibration curve is shown in figure 3.5.2. In this plot, the output voltage of the lock-in amplifier is plotted as a function of the line integrated density $B$.

![Calibration Curve](image)

Fig. 3.5.2: The calibration curve for the mercury ground state density measurements.

The measurements are performed as follows. At first the lamp is switched on. We waited until it burned stable. Then, we measured the transmission. After this measurement, we turned the lamp off and we immediately measured the transmission again. From the difference in transmission with and without plasma in the probe lamp, we calculated the line-integrated mercury density $B_{on}$, using the following procedure. First, the theoretical line-integrated density $B_{off}$ is calculated for the lamp-off situation, where the mercury density profile is uniform. For this calculation, we used the mercury vapour pressure curve as given by Alcock et al.\textsuperscript{[71]} Second, the transmission at this line-integrated density $B_{off}$ is determined from the calibration curve. Third, the transmission in the lamp-on situation is determined from the measured change in the transmission between the lamp-off and the lamp-on situation. Finally, the line-integrated density $B_{on}$ in the lamp-on situation is obtained from the calibration curve.

The use of quartz tubes leads to a complication. The discharges produce 185 nm photons, which can convert oxygen in the surrounding air into ozone. Ozone itself absorbs the 254 nm photons. So not only the mercury in the discharge but also the surroundings can absorb the 254 nm photons. The discharges are placed in a closed box. The ozone density can be regarded as constant during the two transmission measurements, therefore its influence cancels out.

We measured the lateral profile of the line-integrated mercury density. This lateral profile $B_{on}(y)$ is transformed into a radial density profile $n(r)$. This transformation is performed using two methods. In the first method, we assumed the following analytical expression for the radial density profile $n(r)$

$$n(r) = a + b \cdot r^m,$$  \hspace{1cm} (3.5.1)

where $a$, $b$, and $m$ are fitting parameters. Using the Abel integral

$$B(y) = 2 \int_y^R n(r) \frac{r}{\sqrt{r^2 - y^2}} dr,$$  \hspace{1cm} (3.5.2)

we calculated the lateral profile corresponding to the radial profile in expression 3.5.1. This lateral profile is fitted to the measured lateral profile using a least squares routine. We note that the Abel integral is solved numerically. In the second method, we calculated the radial profiles directly from the measured lateral profile, using a method similar to the method of
Bockasten.\textsuperscript{72} The discharge cross-section is divided into shells. In every shell, we assume the density to be constant. The Abel integral is calculated by summing the density $n_i$ in the $i$th shell multiplied with the length $l_i$ of the path of the probe beam in the same shell.

$$B_j = 2 \sum_{i=j}^{n} n_i \cdot l_i ,$$

where the expression for $l_{ij}$ is

$$l_{ij} = \sqrt{r_{i+1/2}^2 - y_j^2} - \sqrt{r_{i-1/2}^2 - y_j^2} , \quad \text{for } i>j,$$

$$l_{ii} = \sqrt{r_{i+1/2}^2 - y_i^2} , \quad \text{for } i=j. \quad (3.5.4)$$

In this way we obtain a matrix equation that describes the transformation from the radial profile to the lateral profile. The inverse transformation can be obtained by calculating the inverse of the transformation matrix. We note that assuming a higher order polynomial function instead of a constant value for the density in a shell will decrease the error we make in the Abel transformation. However, our calculation results in an error that is negligible compared to the experimental errors. Therefore, assuming the density to be constant in a shell is an acceptable simplification of the problem. For the directly transformed profiles we used a Fast Fourier Transformation filter that eliminates oscillations with a spatial period lower than 2 mm in order to reduce the noise. The filter function is a parabola equal to one and horizontal at zero frequency, and equal to zero at the cut-off frequency.

Note that the method used explicitly assumes a radially constant spectral absorption profile. In principle, this is not the case in our lamp because of a temperature gradient. However, because of the relatively low mercury density, the influence of this temperature gradient is small. Another complication is the fact that the gas temperature during the calibration was different from the gas temperature during the transmission measurements. We estimated the maximum influence of the temperature gradient and also the influence of the fact that the calibration and the measurements are taken at different gas temperatures: the total error in the lateral profile was found to be less than 5%. We note that the influence of the modulation of the discharge current is discussed in Appendix B.

### 3.6 Spatially resolved emission spectroscopy

This section describes the experimental set-up we used for measuring the spatially resolved emission spectrum of the neon-mercury positive column. From the results of these measurements, we can also obtain the density profiles of the excited states of neon and mercury. In the first subsection, the emission measurements are described. The second subsection discusses the absorption measurements done in order to check whether the spectral lines we measured are optically thin.

#### 3.6.1 Emission measurements

The set-up consists of a discharge tube, a diaphragm, two lenses, an optical fibre, and an optical multichannel analyser. Figure 3.6.1 shows the set-up schematically. The tube axis is positioned in the focal plane of the first lens. The height of the imaged area in the focal plane is determined by the height of the aperture of the fibre, along with the magnification of the two lenses. The radius of the diaphragm determines the height of the imaged area outside the focal plane of the first lens. The temperature of the discharge tube is controlled by a transparent water jacket. The temperature of the inner wall of the discharge tube at the position of the water jacket determines the mercury pressure in the tube. However, we can
only control the outer wall temperature. Since there is some heat transport through the glass tube, the inner wall temperature will be somewhat higher than the outer wall temperature. This temperature difference is in the order of 0.5°C. The water jacket is connected to a thermostat bath (Haake FE). The water jacket is made from an 80 mm diameter perspex cylinder. The discharge is sustained by standard 35 kHz ballast (Philips BRC 411/01).

![Diagram](https://via.placeholder.com/150)

**Fig. 3.6.1:** The experimental set-up for the emission measurements.

The diaphragm has a diameter of 4 mm. It determines the solid angle of the emission that is captured by the detector. The focal distance of the first lens is 200 mm. The axis of the discharge tube is positioned in the focal plane of this first lens. The second lens has a focal distance of 300 mm. The aperture of the optical fibre is positioned in the focal plane of this second lens. The aperture is circular, with a diameter of 1 mm. This optical system of a diaphragm and two lenses captures the radiation emitted in the detection cone as shown schematically in figure 3.6.1. It focuses this captured radiation on the aperture of the optical fibre. The solid angle in which the radiation is detected is $3 \times 10^{-4}$ sr. The spatial resolution in the focal plane is 0.67 mm. However, since the detection cone widens when moving away from the focal plane, the real spatial resolution is approximately 1 mm. The position of the discharge tube can be changed in order to measure a lateral profile of the plasma emission. The plasma emission is measured through the transparent water jacket. The optical axis is perpendicular to the axis of the tube. On the one hand, in the plane constructed by the optical and the tube axis, the solid angle and the image size do not change. On the other hand, in the plane perpendicular to the tube axis the water jacket acts as a negative lens. However, it can be shown that the absolute emission measurements are not affected by this negative lens, see appendix C. The optical multichannel analyser has already been described in section 3.2. The slit width is 50 microns. The full width at half maximum of the apparatus profile of the spectrograph is 1.7 nm.

The intensity calibration is performed by replacing the discharge tube with a calibrated tungsten ribbon lamp (Osram Wi 17/G). During the calibration, the tungsten ribbon is positioned in the focal plane of the first lens. The optical system is not changed after the calibration procedure, except for the water jacket that is placed around the fluorescent lamp. The transmission of the lamp with this water jacket is measured in a separate experiment, using a tungsten ribbon lamp and a helium-neon laser. For these measurements, we turned the fluorescent lamp off. It is difficult to get an absolute value for the transmission of the water jacket using a tungsten ribbon lamp, since the jacket acts as a strong negative lens. Therefore, we measured a normalised transmission profile at the tube axis with the tungsten ribbon lamp, and scaled this profile to the measurement with the helium-neon laser. This transmission profile can be used to correct the emission measurements. However, in the transmission measurements, Fresnel reflection losses occur at the surfaces. On the other hand, in the emission measurements these reflection losses do not occur. Although the radiation
emitted in the direction of the detector is partly reflected at the tube and the water jacket, this reflection has no influence. The reflection loss is counterbalanced with the extra contribution of the radiation emitted in the opposite direction, which is reflected in the direction of the detector. This is the case for all lateral positions. We corrected the transmission profile of the water jacket for these reflection losses.

From the measured spectra, the line-of-sight integrated intensity for several spectral lines is determined. The measured spectrum is integrated in a 5 nm band around the relevant spectral lines. We determined the radial profile of the emission coefficient \( \varepsilon_{ki}(r) \) from the lateral profile of the line-integrated emission coefficient \( I(y) \) using two different methods. In the first method, we used a fifth order polynomial function as a model for the radial profile. The lateral profile that corresponds to this radial profile is calculated using the following expression

\[
I(y) = 2\int_{y}^{R} \varepsilon(r) \frac{r}{\sqrt{r^2 - y^2}} dr.
\] (3.6.1)

This lateral profile is then fitted to the measurements. We note that the integral can be solved analytically for a polynomial function for \( \varepsilon(r) \). We assumed the intensity gradient at the cylinder axis equal to zero. In the second method, we used the matrix method as described in the preceding section to obtain the radial profile of the experimental data directly. The noise is reduced by applying a Fast Fourier Transform (FFT) filter to the raw data, which eliminates spatial structures narrower than 1 mm. Using the transition probabilities as given in the National Institute of Standards and Technology database, we can calculate the density \( n_k \) of the states from which the radiation originates. In order to do this, we use

\[
n_k = \frac{4\pi \cdot \lambda}{hc \cdot A_{ki}} \varepsilon_{ki},
\] (3.6.2)

where \( \lambda \) is the wavelength of the transition. Note that \( \varepsilon_{ki} \) is expressed in Watt/sr\cdotm^3, so the density is expressed in m\(^{-3}\). The uncertainty in the transition probabilities is 10% for the lines of neon and 10 to 50% for the lines of mercury.

In order to visualise the emission in the lamp, we re-created the radial profile of the total visible spectrum by using the radial profiles of the emission coefficients for the spectral lines of mercury and neon. From the constructed spectra, we calculated the chromaticity coordinates \( x \) and \( y \). These chromaticity coordinates can be transformed into red, green, and blue (RGB) values that can be used to create a bitmap containing the radial profile of the colour of the visible emission. The chromaticity coordinates of the red, green, and blue colour are equal to the ones used in the European PAL television standard. The RGB values are maximised conserving their mutual ratios. Therefore, the cross-sections of the tube we construct do not contain intensity information.

### 3.6.2 Absorption measurements.

It is important to realise that we cannot use the Abel transformation as given in expression 3.6.1 when the spectral lines are not optically thin. In order to check whether the spectral lines we used are optically thin, we measured the transmission of one lamp for the radiation produced by a second, similar lamp. In the absorption set-up, both lamps are operated under the same experimental conditions using a standard ballast (Philips BRC 411/01), their temperature is controlled by the same thermostat bath (Haake FE). A chopped beam of 1 mm diameter, produced by the first (source) lamp is guided through the second (probe) lamp. We used a chopper at a frequency of 1.8 kHz (Princeton Applied Research Model 191 Variable Speed Chopper). The phases of the driving waves of the two lamps are uncorrelated. The
influence of the modulation on the absorption measurements is described in appendix B. The transmission is measured at $y = 0$, i.e. through the axis of the lamp. The radiation is detected by a photomultiplier (Radio Corporation of America type 1P28). This photomultiplier is mounted on a monochromator (Oriel model 7240). The chopped radiation produced by the source lamp is discriminated from the radiation produced by the probe lamp by a lock-in amplifier (EG&G Princeton Applied Research model 5209). The transmitted radiation when the probe lamp is on is divided by the transmitted radiation in the situation where the probe lamp is off, giving the transmission of the plasma. When this transmission is high enough, then the line is optically thin. When the absorption in the tube is linear, then the maximum relative error we make in the radial profile of the emission coefficient is one minus the square root of the measured transmission.

We measured the transmission for some of the spectral lines of mercury and neon in a lamp with a neon pressure of 1000 Pa, cold-spot temperatures of 18 and 30°C, and an electric current of 400 mA. We will start with the mercury lines at a cold-spot temperature of 18°C.

Table 3.6.1 shows the measured transmission of several spectral lines of the mercury atom. It is clear that the spectral lines corresponding to the transitions to the resonant states $6^3P_1$ (435.83 nm) and $6^1P_1$ (576.96 nm) are optically thin.

Table 3.6.1: The wavelengths of the measured spectral lines, the corresponding transitions, the transition probabilities $A_{\text{hi}}$, the transmission $t$, and a summary of the conclusions of the absorption measurements: whether or not a line is optically thin.

<table>
<thead>
<tr>
<th>$\lambda$ (nm)</th>
<th>Transition</th>
<th>$A_{\text{hi}}$ ($10^8$ s$^{-1}$)</th>
<th>$t_{18^\circ C}$ (%)</th>
<th>$t_{30^\circ C}$ (%)</th>
<th>$I \leq 400$ mA, $T_{\text{cold}}=18^\circ C$, $p_{\text{Ne}}=1000$ Pa</th>
<th>$I=400$ mA, $T_{\text{cold}}=18^\circ C$, $p_{\text{Ne}}=1000$ Pa</th>
<th>$I=400$ mA, $T_{\text{cold}}=18^\circ C$, $p_{\text{Ne}}=300$, 1500 Pa</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hg 404.66</td>
<td>$7^3S_1-6^3P_0$</td>
<td>0.21</td>
<td>90</td>
<td>75</td>
<td>?</td>
<td>No</td>
<td>?</td>
</tr>
<tr>
<td>435.83</td>
<td>$7^3S_1-6^3P_1$</td>
<td>0.557</td>
<td>100</td>
<td>86</td>
<td>Yes</td>
<td>No</td>
<td>?</td>
</tr>
<tr>
<td>546.07</td>
<td>$7^3S_1-6^3P_2$</td>
<td>0.487</td>
<td>68</td>
<td>48</td>
<td>?</td>
<td>No</td>
<td>?</td>
</tr>
<tr>
<td>576.96</td>
<td>$6^1D_2-6^1P_1$</td>
<td>0.236</td>
<td>100</td>
<td>95</td>
<td>Yes</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>491.61</td>
<td>$8^3S_0-6^1P_1$</td>
<td>0.058</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Yes</td>
</tr>
<tr>
<td>Ne 588.19</td>
<td>$2p_2-1s_1$</td>
<td>0.115</td>
<td></td>
<td></td>
<td>Yes</td>
<td>Yes</td>
<td>?</td>
</tr>
<tr>
<td>594.48</td>
<td>$2p_2-1s_1$</td>
<td>0.113</td>
<td>96</td>
<td></td>
<td>Yes</td>
<td>Yes</td>
<td>?</td>
</tr>
<tr>
<td>614.31</td>
<td>$2p_0-1s_1$</td>
<td>0.282</td>
<td>93</td>
<td></td>
<td>No</td>
<td>?</td>
<td>?</td>
</tr>
<tr>
<td>640.22</td>
<td>$2p_0-1s_1$</td>
<td>0.514</td>
<td>81</td>
<td></td>
<td>No</td>
<td>?</td>
<td>?</td>
</tr>
<tr>
<td>609.62</td>
<td>$2p_4-1s_1$</td>
<td>0.181</td>
<td>94</td>
<td></td>
<td>No</td>
<td>?</td>
<td>?</td>
</tr>
<tr>
<td>650.65</td>
<td>$2p_6-1s_1$</td>
<td>0.300</td>
<td>90</td>
<td></td>
<td>No</td>
<td>?</td>
<td>?</td>
</tr>
<tr>
<td>724.52</td>
<td>$2p_{10}-1s_1$</td>
<td>0.0935</td>
<td></td>
<td></td>
<td>Yes</td>
<td>Yes</td>
<td>?</td>
</tr>
<tr>
<td>626.65</td>
<td>$2p_5-1s_1$</td>
<td>0.249</td>
<td>87</td>
<td></td>
<td>No</td>
<td>?</td>
<td>?</td>
</tr>
<tr>
<td>585.25</td>
<td>$2p_{1}-1s_2$</td>
<td>0.682</td>
<td>95</td>
<td></td>
<td>Yes</td>
<td>Yes</td>
<td>?</td>
</tr>
<tr>
<td>534.11</td>
<td>$4d_{5}-2p_{10}$</td>
<td>0.110</td>
<td></td>
<td></td>
<td>Yes</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>748.89</td>
<td>$3d_{5}-2p_{10}$</td>
<td>0.349</td>
<td></td>
<td></td>
<td>Yes</td>
<td>Yes</td>
<td>Yes</td>
</tr>
</tbody>
</table>

The two transitions to the metastable states (546.07 and 404.66 nm) are not optically thin. However, the density of the $7^3S_1$ state can be obtained from the 435.83 nm line. The 546.07 and 404.66 nm lines originate also from this $7^3S_1$ states, so we can construct these lines from the density of the $7^3S_1$ state. The density of the resonant and the metastable states increases with increasing electric current through the lamp. Therefore, we can assume that the transitions to the resonant states are optically thin for currents lower than 400 mA. At a
cold-spot temperature of 30°C, the situation is quite different. Only the transitions resulting in the $6^1P_1$ state are optically thin. The density of the $7^3S_1$ state cannot be obtained at this cold-spot temperature. Based on calculations by Vriens et al.,\textsuperscript{75} who used the results from Koedam et al.,\textsuperscript{76} we can assume that at different neon pressures and a cold-spot temperature of 18°C, the transitions to the $6^1P_1$ state are optically thin.

The absorption of the neon lines is measured at a cold-spot temperature of 18°C. Table 3.6.1 also shows the transmission for several neon lines. We used the Paschen notation for the identification of these lines. Figure 3.6.2 shows the measured transmission as a function of

$$\kappa = A_{k_i} \cdot \lambda^3 \cdot \frac{g_k}{g_i}, \quad (3.6.3)$$

which scales with the absorption coefficient. In this expression, $A_{k_i}$ is the transition probability, $\lambda$ is the wavelength of the transition, and $g_k$ and $g_i$ are the statistical weights of the upper and the lower state, respectively. We separated the spectral lines in four groups. These groups correspond to the four lower states of the transitions, either $1s_2$, $1s_3$, $1s_4$, or $1s_5$. For each lower state, we measured at least one spectral line. We assumed the absorption coefficient to correlate positively with $\kappa$. Therefore, we can give a maximum value for the absorption of lines which have a lower $\kappa$ than the lines for which we measured the transmission. When the transmission of a line is higher than 95%, then we assume the line to be optically thin.

![Fig. 3.6.2: The transmission of the spectral lines of neon as a function of $\kappa$. The lower state of the corresponding transition is either $1s_2$ (circle), $1s_3$ (square), $1s_4$ (up-triangle), or $1s_5$ (down-triangle)](image)

We also measured the emission of two very weak lines at 534.11 and 748.89 nm corresponding to transitions to the $2p_{10}$ state. The density of the $2p_{10}$ state can be estimated from the emission of the spectral lines originated from this state. This density is low enough to assume the 534.11 and the 748.89 nm lines to be optically thin.

At lower electric currents and at higher cold-spot temperatures, the neon emission is lower. This is because of the fact that the degree of depletion is lower. Therefore, we can assume that the density of the metastable and the resonant states is also lower. So for the absorption of the neon spectral lines, the 400 mA, 18°C measurements are the worst-case. For different neon pressures we can still use the 534.11 and the 748.89 nm lines. Table 3.6.1 gives a summary of the conclusions, drawn in this subsection.
3.7 Time-resolved emission spectroscopy

As mentioned in the preceding sections, the driving frequency of the discharge is a few tens of kilohertz. The discharge emission will be modulated with the doubled frequency. This section describes the phase-resolved emission measurements we performed to study this modulation. Besides this, we also discuss the measurement performed to investigate the influence of amplitude modulation of the discharge current.

The set-up for the measurements consists of the optical multichannel analyser with the input optics as described in section 3.2. The measurements are performed while the input optics are aligned to the axis of the discharge tube. The radiation in a cylinder of approximately 2 mm diameter through the axis of the discharge tube is captured and focussed onto the fibre. A transparent water jacket is placed around the discharge, as described in section 3.4. The discharge is filled with 1000 Pa neon. It is partly covered with a fluorescent powder coating. The intensity calibration and the correction for the transmission of the water jacket are performed in the same way as described in section 3.4. The electrical system consists of a function generator (Hewlett-Packard 33120A function/arbitrary waveform generator), a power amplifier (Spitzenberger & Spies EV 600/CuG), an additional transformer (Spitzenberger & Spies UT 600/G) a capacitor (0.47 µF), and an inductor (3.1 mH). The capacitor and the inductor are placed in series with the discharge. The inductor suppresses the higher harmonics in the discharge current. The capacitor assures that the current contains no DC component. A current probe (Pearson electronics inc. Wideband current monitor model 150) is used to measure the discharge current. It is connected to an oscilloscope (Tektronix TAS250). The arbitrary waveform generator is used in either the continuous wave (CW) mode or the burst mode. The burst mode can be regarded as amplitude modulation with a square modulation-waveform. The driving frequency is 50 kHz, the waveform is sinusoidal. For the time-resolved measurements, the intensifier of the optical multichannel analyser is used in the gated mode. The intensifier is controlled by a pulse amplifier (EG&G PARC M1304), which is triggered by a gate-pulse interface (EG&G PARC M1303). This interface itself is triggered by a delay generator (Stanford Research Systems DG535). The sync output of the arbitrary waveform generator is used to trigger the delay generator. The delay for the measurements is generated by the gate-pulse interface, which is controlled by the detector interface (EG&G PARC 1461). The Stanford delay generator is used to inhibit the triggering and thus to set the repetition rate of the gating.

We measured the phase-resolved emission spectrum of the discharge in the CW mode without the phosphor coating. The intensifier was gated for 1 microsecond at a repetition rate of 4.5 kHz. Besides this, operating the lamp in the burst mode, we measured the time evolution of the emission spectrum without phosphor during a burst. The intensifier was gated for 200 microseconds at a repetition rate equal to the modulation frequency. Finally, we measured the time-averaged emission spectrum of the lamp with and without the fluorescent coating in the CW and the burst mode. The amplitude of the discharge current was kept constant or it was modulated at a frequency of 50 or 100 Hz.
3.8 Incoherent Thomson scattering

3.8.1 Introduction
Thomson scattering is elastic scattering of photons at free electrons. It has been used extensively for the determination of the electron energy distribution function in high-density plasmas like fusion plasmas. Recently, this technique has also been introduced for plasmas with electron densities down to $10^{17}$ m$^{-3}$. Thomson scattering gives reliable results for the electron density and the electron temperature. In contrast with Langmuir-probe measurements, the interpretation of the measurements is straightforward and the results are unambiguous. However, the main difficulty for performing Thomson scattering in low-density plasmas is the low intensity of the scattered signal. The Thomson scattering intensity is very small compared to the intensities of plasma emission, Rayleigh scattering and stray light. In order to increase the Thomson signal, high power lasers such as Nd:YAG lasers are used. However, using more laser power also increases the Rayleigh signal and the stray light. The wavelength of these two signals is approximately equal to the wavelength of the laser. The spectral width of the Thomson signal, however, is much larger due to the Doppler effect and the movement of the free electrons. This is why normally one can discriminate the stray light (and the Rayleigh scattering) from the Thomson scattering signal by using a grating monochromator. However, due to the instrumental broadening of the monochromator the stray light and the Thomson signal will overlap. Furthermore, the stray light intensity is a few orders of magnitude higher than the Thomson signal, so the Thomson signal will be lost in the noise of the stray light. This is especially the case in a low-density plasma like a fluorescent lamp, where the stray light level is very high due to the proximity of the tube wall.

A notch filter at the laser wavelength will reduce the stray light considerably. However, interference notch filters cannot be used because of the fact that the spectral width of the best interference notch filters available is comparable to the characteristic width of the Thomson signal. One has to look for special notch filters like an atomic absorption cell or a double monochromator. We propose an atomic absorption cell as a notch filter. The biggest problem of using such a filter in a Thomson scattering set-up is finding the laser at the right wavelength, with sufficient spectral purity. We will describe a way to introduce an atomic notch filter in a Thomson scattering set-up.

3.8.2 Overview of the Thomson scattering set-up.
The set-up we built is basically a standard Thomson scattering set-up, except for two components. Firstly, we used an excimer pumped dye laser with an ASE filter instead of a ‘standard’ Nd:YAG laser. Secondly, we added a sodium vapour absorption cell in the detection branch of the set-up. Figure 3.8.1 shows a schematic drawing of the set-up.

The excimer pumped dye laser (Lambda Physik LPX 315, and FL 3002) produces a laser beam with a wavelength of 589.0 nm. The pulse energy is at maximum 60 mJ at a repetition frequency of 150 Hz. The pulse width is 30 ns. The laser beam is focussed in a fluorescent lamp by a lens with a focal distance of 800 mm. The laser wavelength is equal to the wavelength of the Sodium D$_2$ transition. This means that, given the fact that the intensity is not too high and the spectral width of the laser is small enough, the laser radiation will be absorbed by Sodium vapour. On the other hand, the Thomson signal, which is Doppler broadened, will be transmitted through Sodium vapour.
There is one problem with this set-up: The laser not only produces radiation at 589.0 nm, but also a broadband signal called Amplified Spontaneous Emission (ASE). This ASE signal has the same spectral width as the dye-tuning curve. The spectrally integrated ASE level is typically 1‰ to 1% of the laser power. Because of the fact that it is a broadband signal, it will not be absorbed by sodium vapour. Therefore, we need a bandpass filter with a spectral width smaller than the spectral width of the sodium vapour absorption cell. To accomplish this, we will make the spectral width of the ASE filter as small as reasonably possible and we will increase the spectral width of the sodium vapour absorption cell sufficiently. Then, the stray light reduction will be equal to the rejection of the ASE filter multiplied by the ASE level of the laser.

In the next subsections we will discuss the ASE filter, the sodium absorption cell, and the detection branch of the set-up respectively.

### 3.8.3 The Amplified Spontaneous Emission filter

For several applications, the ASE background of a dye laser is unwanted. For example, when using a dye laser for Thomson scattering, the ASE can lead to unwanted fluorescence in the plasma. Because of the small cross-section of Thomson scattering, the magnitude of the fluorescence signal will be some orders of magnitude larger than the Thomson signal. In order to reduce the ASE background, Schlüter already built a spatial filter with one dispersion prism. A dispersion prism is used instead of a grating because in general, prisms are better resistant to high power densities. The 1% width of this filter is in the order of 3 nm. Our goal is to build a filter, with a spectral width much lower than 1 nm. The dispersion of one prism is too small to achieve this goal. Therefore we designed a configuration of 20 prisms in order to have a 20 times larger dispersion.

At first we will discuss the design of the dispersing part of the ASE filter, i.e. the twenty prisms. The monochromator is designed to transmit a certain wavelength; we will define this wavelength as the zero-wavelength $\lambda_0$. This wavelength is of course the central wavelength of the transmission profile of the filter (in our case, 589.6 nm). Along with the zero-wavelength, we define the zero-beam as the beam with wavelength $\lambda_0$. In order to work properly, the configuration of the prisms has to meet three criteria:

- The prisms may not change the cross-section of the transmitted zero-beam.
- The reflection losses for the zero-beam should be minimised.
- The dispersion of the prisms has to be additive.
These three criteria determine the way that the zero-beam will be transmitted through the prisms. The first criterion states that the zero-beam has to be transmitted symmetrically through the prism, i.e. the angle of incidence is the same at both prism surfaces. Then the prisms will not change the cross-section of the beam. The second criterion states that the angle of incidence for the zero-beam on both sides of the prisms has to be near to the Brewster angle. The third criterion determines the relative orientation of two adjacent prisms.

We define $\alpha$ as the angle of incidence on the first prism surface, $\beta$ as the angle of refraction at the first surface, $\gamma$ as the angle of incidence at the second prism surface, and $\varphi$ as the prism angle, as shown in figure 3.8.2 for an isosceles triangle.

We choose equilateral prisms, so $\varphi$ is equal to 60°. The prisms are made of Schott SF10 glass. The expression for the angle of incidence for the zero beam $\alpha_0$ can be obtained using Snell’s law and the first criterion, a symmetrical zero-beam

$$\alpha_0 = \arcsin \left( n_0 \cdot \sin \left( \frac{\varphi}{2} \right) \right) = 59.71^\circ, \quad (3.8.1)$$

where $n_0$ is the index of refraction for the zero-wavelength (with respect to the index of refraction of the surrounding air). The index of refraction of the glass is interpolated from data given by Schott, the value for air is taken from Edlen. The Brewster angle $\alpha_B$ for SF10 glass and this wavelength is

$$\alpha_B = 59.93^\circ, \quad (3.8.2)$$

which is very near to the value of $\alpha_0$. So using these prisms and the zero-beam polarised in the plane of incidence, the first two criteria are met. For the third criterion, the relative orientation of two adjacent prisms, we make a Taylor expansion of Snell’s law around the zero-beam, at both prism surfaces, using

$$n(\lambda) = n_0 + \delta n = n_0 + \left. \frac{dn}{d\lambda} \right|_{\lambda_0} \cdot \delta \lambda, \quad (3.8.3)$$

where $\delta \lambda$ is the deviation with respect to the zero-wavelength. The Taylor expansion results in the following two expressions

$$\frac{\sin(\alpha_0 + \delta \alpha)}{\sin(\beta_0 + \delta \beta)} = n_0 + \left. \frac{dn}{d\lambda} \right|_{\lambda_0} \cdot \delta \lambda, \quad \text{and} \quad \frac{\sin(\gamma_0 + \delta \gamma)}{\sin(\beta_0 - \delta \beta)} = n_0 + \left. \frac{dn}{d\lambda} \right|_{\lambda_0} \cdot \delta \lambda.$$ 

Which can be rewritten as

$$\frac{\sin(\alpha_0) + \cos(\alpha_0) \cdot \delta \alpha}{\sin(\beta_0) + \cos(\beta_0) \cdot \delta \beta} = n_0 + \left. \frac{dn}{d\lambda} \right|_{\lambda_0} \cdot \delta \lambda,$$

and

$$\frac{\sin(\gamma_0) + \cos(\gamma_0) \cdot \delta \gamma}{\sin(\beta_0) - \cos(\beta_0) \cdot \delta \beta} = n_0 + \left. \frac{dn}{d\lambda} \right|_{\lambda_0} \cdot \delta \lambda \quad (3.8.4)$$
where $\delta \alpha$, $\delta \beta$, and $\delta \gamma$ are the deviations with respect to the zero beam for $\alpha$, $\beta$, and $\gamma$, respectively. These deviations are assumed to be small with respect to the corresponding angles for the zero-beam. Note the minus sign in the denominator of the second expression.

We can eliminate $\delta \beta$ from these two expressions, and obtain the following expression for $\delta \gamma$:

$$\delta \gamma = 2\cdot \tan(\alpha_0) \cdot \left. \frac{dn}{d\lambda} \right|_{\lambda=10} \cdot \delta \lambda - \delta \alpha.$$  \hfill (3.8.5)

Note that a positive $\delta \alpha$ results in a negative $\delta \gamma$. The orientation of the prisms has to be chosen in such a way that for the zero-beam, the angle of incidence at the second prism is the same as the angle of incidence at the first prism. This is because of the fact that the beam in the second prism has to be symmetric too. Above this, the dispersion of the two prisms has to be additive. Figure 3.8.3 shows the configuration in which these criteria are met.

The expression for $\theta$ can be obtained from

$$(90^\circ - \alpha_0) + (90^\circ - \alpha_0) + (240^\circ - \theta) = 180^\circ,$$

so we have

$$\theta = 240^\circ - 2\cdot \alpha_0 = 120.57^\circ.$$  \hfill (3.8.7)

With this value for $\theta$, the zero-beam will pass the second prism symmetrically, and the dispersion of the second prism will be added to the dispersion of the first prism. The same construction can be used for the rest of the 20 prisms. The configuration of the 20 prisms is shown schematically in figure 3.8.4.

It can be shown that the expression for $\delta \gamma$ after passing twenty prisms is

$$\delta \gamma_{20} = 20 \cdot \left[ 2\cdot \tan(\alpha_0) \cdot \left. \frac{dn}{d\lambda} \right|_{\lambda=10} \right] \cdot \delta \lambda = 5.0 \cdot 10^{-3} \cdot \delta \lambda \quad [\text{rad}],$$  \hfill (3.8.8)
where we assumed that $\delta \alpha = 0$ at the first prism, and $\delta \lambda$ is expressed in nanometres. It is clear from this expression that the dispersion of the 20 prisms is 20 times higher than the dispersion of one prism. Therefore we can use the 20 prisms in our set-up in order to obtain a filter with a transmission width smaller than 1 nm.

After passing the prisms, the beam will be focussed by a lens with focal length $f$. A pinhole with radius $R$ is placed in the focal plane of this lens. This pinhole selects the wavelength that is transmitted. We assume a Gaussian beam profile, with the exp(-2) laser waist diameter $w$. The position in the focal plane is given by the coordinates $x$ and $y$. The centre of the laser beam is positioned at $x = m$, $y = 0$. The expression for the transmission of the pinhole $T$ is

$$T = \frac{R}{\sqrt{\pi} \cdot 4} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \exp \left( -8 \left( x - f \cdot \tan(\delta \gamma_{20}) \right)^2 + y^2 \right) dy,$$

(3.8.9)

In this expression, we assumed $m = 0$ for the zero-beam.

In order to measure the transmission of the filter as a function of wavelength, a set-up is built that consists of a Ring dye laser (Coherent 899-21 Dye Ring Laser), the ASE filter, a photodiode (EG&G SGD-100A), and a differential amplifier (Tektronix AM 502). The laser power transmitted through the ASE filter is measured with the photodiode. The differential amplifier amplifies the signal of the photodiode. This amplifier is connected to an ADC. The laser we used is a single mode laser with a spectral width smaller than 1 MHz. The spectral impurity of the laser in the region of interest, i.e. from 587.6 to 591.6 nm is negligible. Appendix D describes the estimation of this impurity level. The ASE filter in the test configuration consists of

- A first spatial filter: an $f = 200$ mm lens, a 200 $\mu$m pinhole and an $f = 400$ mm lens
- The twenty prisms.
- A mirror and two beam-guiding prisms.
- A second spatial filter: an $f = 400$ mm lens, a 200 $\mu$m pinhole and an $f = 200$ mm lens.

The first spatial filter cleans the beam from parts that cannot be focussed and expands the beam to a diameter of approximately 10 mm. After passing this first spatial filter, the divergence of the beam is in the order of 0.5 mrad. The 20 prisms with dimensions 30 mm $\times$ 30 mm disperse the beam. They are positioned in four groups of five prisms. These five prisms are at fixed positions with respect to each other. Every group can be rotated, and can be translated in the direction rectangular to the incident beam. The position and the rotation of the four groups is chosen in such a way that the zero-beam enters every first prism and leaves every last prism of a group in the middle of the prism surface. In practice, adjustment of the position and the rotation of the groups did not change the transmission of the filter very much. Without adjusting, just putting all the 20 prisms in their mounts, the transmission was already more than 85%. With minor adjustment a transmission up to 90% could be achieved. The second spatial filter removes radiation with unwanted wavelengths. This is the wavelength selection part of the filter. The second pinhole is positioned by maximising the transmission of the zero-beam.

In order to obtain optimum transmission, the incident beam needs to be polarised parallel to the plane of incidence. The polarisation of the laser beam could be adjusted with a polariser. The proper polarisation of the laser beam is found by adjusting the polariser angle and simultaneously minimising the reflection of the zero-beam at the first prism. Note that the polariser is not needed when the laser polarisation is chosen properly. The laser is scanned from 587.6 to 591.6 nm, and the amount of light passing the second spatial filter is measured. In order to measure a large dynamic range of transmitted light a neutral density
filter (Pomfret Research Optics inc. 3-2220-2 ND 2.0) is used, and for very low intensities the amplification of the differential amplifier is increased. This is possible because of the fact that the linearity of the response of the photodiode is better than 5% over seven decades. The transmission of separate components in the ASE filter is measured with a power meter head (Coherent LD10). The maximum transmission $T$ of a pinhole can be used to estimate the exp(-2) laser waist diameter $w$, using the following expression
\[
T = 1 - \exp\left(-2 \cdot \frac{D^2}{w^2}\right),
\]
where $D$ is the pinhole diameter. This expression is derived from expression (3.8.9), with $\delta \gamma_{20} = 0$. The maximum transmission of the first pinhole was 97%, which results in a waist diameter of 151 $\mu$m. The maximum transmission of the second pinhole was 93%, which results in a waist diameter of 173 $\mu$m. The change in waist diameter can be used as a measure for the monochromatic aberrations introduced by the prisms. The waist diameter before and after the prisms would be the same when there are no aberrations.

The results of the measurements are given in figure 3.8.5. In this figure, the transmission is normalised to the transmission of the zero-beam.

![Graph showing normalized transmission profile](image)

We note that the maxima and the minima in the wings of the profile are reproducible, they result from scattering of the beam on the edges of the mirror and the beam-guiding prisms. The influence of the spectral impurity of the laser beam is negligible, the 1% width of the filter is 0.23 nm, this means that the intensity of the transmitted spectral impurity will be at least 7 orders of magnitude smaller than the transmission for the zero-beam.

With the value of the laser waist diameter in the second pinhole, the theoretical transmission profile can be calculated using expression 3.8.9. The theoretical profile is plotted in figure 3.8.6, along with the measured profile. It is clear that the theoretical profile fits the measured profile very well. This means that the measured dispersion is the very near to the calculated dispersion.

The transmission of the prisms for the zero-beam is measured also. The transmission of twenty prisms is equal to 90%. This means that we lose 5% per prism. This loss contains three components: (1) The reflection loss, (2) The loss due to the fact that the internal transmission is different from unity, and (3) scattering losses at the surfaces. The (Fresnel)
reflection loss per prism is $1.3 \cdot 10^{-5}$, the internal transmission for 15 mm SF10 glass is $0.997^{78}$, so the scattering loss is $1 \%$ per prism surface.

![Transmission vs Wavelength](image)

**Fig. 3.8.6:** The theoretical and the measured transmission profile.

The transmission of the complete ASE filter in the test configuration for the zero-beam is approximately 80 %. The prisms give rise to a loss of 10 %, the first pinhole 3% and the second pinhole 7%. The dispersion of the prisms is as we expected from Snell’s law and the index of refraction of SF10 glass. The 1% width of the filter is 0.23 nm, and the rejection in the region 587.6 nm – 591.6 nm is $10^{-5}$. In the situation where the wavelength is different from the zero-wavelength, the light passing the pinhole is light scattered at the prism and lens surfaces. Inserting baffles did not improve the rejection, except for the case where the baffles blocked the path of the zero-beam. From this, we can conclude that part of the scattered light shines in the direction of the zero-beam, even in the situation where the wavelength is different from the zero-wavelength. So reducing the scattering will result in a better rejection of the unwanted wavelengths. Furthermore, by allowing the beam to pass the prisms for a second time, using a third pinhole, a squared rejection of the unwanted wavelengths can be obtained: the beam that is transmitted through the second pinhole can be transmitted through the prisms for a second time. The prisms are large enough to allow the second beam to pass them independent from the first beam, i.e. without using the same area on the prism surface.

The width of the filter is limited by the laser waist diameter. This waist diameter determines the minimum pinhole diameter that can be used. The ASE filter design can also be used for other wavelengths. If the angle between two adjacent prisms $\theta$ and the angle of incidence on the first prism surface $\alpha_0$ are not changed, then the applicability of the filter is limited by the dimensions of the prisms and the laser beam. The dispersion has to be small enough to allow the beam to pass all the prisms. In our case, wavelengths that deviate a few nanometres from the zero-wavelength could be used. The transmission will change due to the changed Fresnel reflection loss. The Fresnel reflection loss for twenty prisms in the wavelength-range from 585 to 595 nm will be smaller than 5 %. In order to obtain the optimum transmission for wavelengths different from 589.6 nm, the angle between two adjacent prisms $\theta$ has to be changed. This will involve a slight modification of the mechanical mounting of the prisms. When $\theta$ is chosen properly, the maximum Fresnel reflection loss for twenty prisms in the wavelength-range from 400 to 700 nm is 4%.

The ASE filter as used in the Thomson scattering set-up has a slightly different configuration as the one described above. In the Thomson scattering set-up, all the
components are mounted on a vertical breadboard. This vertical orientation has two advantages: At first, we do not need to change the polarisation of the laser beam, and at second a vertical breadboard needs less space in the laboratory. The lenses have a focal distance of 400 mm. The pinholes are produced by focussing the laser on a piece of aluminium tape. In this way we ‘drilled’ a hole which exactly matched the laser focus. The diameter of the pinholes was 320 ± 20 microns. We note that by changing the position of the second pinhole, the central wavelength of the bandpass filter can be changed. We used the filter at 589 and at 589.6 nm.

The next objective is to design an absorption cell with a spectral width that matches the spectral width of the ASE filter. In the next subsection we will discuss the design of our sodium vapour absorption cell.

### 3.8.4 The Sodium vapour absorption cell

This subsection describes the design and the characteristics of the sodium vapour absorption cell. This absorption cell has to absorb all the stray light. On the other hand, the Thomson scattering signal has to be transmitted through the cell without being absorbed. We can define four criteria for the absorption cell:

- The absorption of the stray light has to be very large, even when saturation of the transition is present.
- The excited atoms should lose their energy before the next laser pulse arrives.
- The transmission width has to be larger than the width of the ASE filter.
- The transmission of non-resonant light should be as high as possible.

Note that the transmission width is defined as the spectral width of the transmission profile of the absorption cell. This width is much larger than the width of the absorption lines.

The first two criteria determine the atom density in the cell and the absorption length.

We start with the first criterion. Absorption without saturation can be described by Lambert-Beer’s law. However, due to the high power radiation, we have to consider saturation. A very simple way of treating saturation in the resonant absorption of a nanosecond scale high power laser pulse is considering the saturation length concept. We assume the absorption in the vapour to be high. When a high power laser pulse enters the vapour, it will excite the atoms in the vapour. The upper state of the atoms will be populated. We will divide the laser pulse in $N$ equal parts. The first part of the pulse excites part of the atoms along a certain length in the absorption cell. This part of the cell is called the saturated region. In this region, the excited atom density will be so high that the probability for stimulated emission from the excited state will be equal to the probability for absorption from the ground state. Therefore, when the lifetime of the excited state is much higher than the length of the laser pulse, then the second part of the laser pulse will be transmitted through the part saturated by the first part of the pulse without losing photons. When the second part of the pulse arrives at the end of the saturated region, then it will excite the atoms in the subsequent part of the cell, thereby extending the saturated region. This process will repeat for the $N$ parts of the pulse. After the whole pulse is absorbed, the number of excited atoms in the vapour will be equal to the number of photons in the pulse. In this way we can relate the number of photons $N_p$ in the pulse to the number of excited atoms in the saturated region.

$$N_p = \frac{1}{g_1} \cdot n_a \cdot A \cdot l_s, \quad (3.8.11)$$
where $g_1$ and $g_2$ are the statistical weights of the lower and the upper state of the transition, $n_a$ is the atom density, $A$ is the area of the light beam, and $l_s$ is the saturation depth. We assume that $A$ is equal to the cross-section of the absorption cell. We can rewrite this expression

$$l_s = N_p \cdot \left( 1 + \frac{g_1}{g_2} \right) \cdot \frac{\lambda}{n_a \cdot A} = E_p \cdot \frac{\lambda}{h} \cdot \left( 1 + \frac{g_1}{g_2} \right) \cdot \frac{1}{n_a \cdot A},$$

(3.8.12)

where $E_p$ is the pulse energy, $\lambda$ is the wavelength, $h$ is Planck’s constant and $c$ is the velocity of light. In order to have sufficient absorption, the length of the absorption cell has to be larger than the saturation length.

The second criterion deals with transport of resonance radiation. We will make a very rough approximation of the characteristic time for de-excitation of the atoms in the vapour cell. We assume that the radiation transport can be described as a diffusion process of excited atoms. The diffusion coefficient $D$ for this process is

$$D = \frac{1}{\sigma^2 \cdot \tau \cdot n_a^2},$$

(3.8.13)

where $\sigma$ is the cross-section for absorption and $\tau$ is the lifetime of the upper state. We neglect quenching so the only loss of excited atoms (photons) is at the walls. In this way we estimate the upper limit of the characteristic time for de-excitation of the atoms in the vapour cell. The expression for this characteristic time is

$$\tau_d = \frac{\Lambda^2}{D} = \sigma^2 \cdot \tau \cdot n_a^2 \cdot \Lambda^2,$$

(3.8.14)

where $\Lambda$ is the characteristic length of diffusion. In a long cylinder with a circular cross-section, this length is equal to $R/2.4$, where $R$ is the radius of the cylinder.

The third criterion can be met by adding argon gas to the vapour. We thereby introduce pressure broadening of the resonant absorption lines.

The fourth criterion is met because of the fact that atomic absorption lines have very steep profiles. However, keeping the windows clean is an important issue.

The atomic vapour in the absorption cell is sodium. Sodium is chosen because of its strong D lines at 589.0 and at 589.6 nm. At these wavelengths we can use the Rhodamine 6G dye, which is cheap, relative stable, and efficient. The sodium D$_2$ line at 589.0 nm is more suitable than the D$_1$ line at 589.6 nm. The D$_2$ line is stronger, and its transmission width is larger. We chose the cell radius and the absorption length in such a way that we have met the requirements mentioned above, and in such a way that we could use standard ConFlat (CF) vacuum components.

In table 3.8.1, the values for the absorption length, the cell radius, and the argon pressure are given. This table also shows two different sodium pressures and the values for the saturation length, the characteristic time for de-excitation, and the 1% transmission width. This width is calculated from the sodium vapour pressure$^{71}$ and the collisional broadening cross-section for the sodium D$_2$ line by argon.$^{80}$ For the cold-spot temperature of 250°C, we assumed a gas temperature of 325°C, for the cold-spot temperature of 300°C, we assumed a gas temperature of 330°C. It seems that both sodium pressures can be used: both the saturation length and the characteristic time for de-excitation are small enough. Furthermore, both the transmission widths are large enough. Note that the saturation length is not long enough for the direct absorption of the laser pulse. However, the cell has to absorb the scattered radiation only. The pulse energy of this scattered radiation will be more than two orders of magnitude lower than the laser pulse energy. Due to this, the absorption length for the scattered radiation will be smaller than the length of the absorption cell. Concerning the
de-excitation of the atoms in the cell, we can draw an interesting conclusion. The de-
excitation takes place on a microsecond scale. The Thomson scattering signal is present
during the 30 nanoseconds of the laser pulse. So when we open the detector during the laser
pulse and close it when the pulse is terminated, then the photons emitted during the de-
excitation of the atoms in the vapour cell will not be detected.

Table 3.8.1: The characteristic quantities of the sodium vapour absorption cell.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Value 1</th>
<th>Value 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Absorption length</td>
<td>17 cm</td>
<td>17 cm</td>
</tr>
<tr>
<td>Cell radius</td>
<td>2.5 cm</td>
<td>2.5 cm</td>
</tr>
<tr>
<td>Argon pressure (temperature)</td>
<td>10^5 Pa @20°C</td>
<td>10^5 Pa @20°C</td>
</tr>
<tr>
<td>Sodium pressure (temperature)</td>
<td>0.27 Pa (250°C)</td>
<td>2.12 Pa (300°C)</td>
</tr>
<tr>
<td>Saturation length per pulse energy</td>
<td>70 m/Joule</td>
<td>8.9 m/Joule</td>
</tr>
<tr>
<td>Characteristic time for de-excitation</td>
<td>7 µs</td>
<td>421 µs</td>
</tr>
<tr>
<td>Theoretical 1% transmission width</td>
<td>0.17 nm</td>
<td>0.50 nm</td>
</tr>
</tbody>
</table>

Having performed this theoretical estimation, the design of the sodium cell seems
straightforward. However, it is not. Sodium is very reactive, it is a very strong chemical
reducing agent. Special care has to be taken in choosing materials of the cell that can be in
contact with the sodium vapour. The absorption cell can be divided into six parts: the
windows, the glass to metal seal, metal to metal junction, the vessel, the gaskets, the gas fill.
Note that the metal to metal junction is necessary to match the thermal expansion of the
window material to the vessel. Apart from these six parts, there is also some contamination.
After having studied the literature carefully, we chose suitable materials for every part of the
absorption cell. Table 3.8.2 shows the materials we used, along with their resistance to
sodium as has been found in the literature. We note that we do not claim to give a complete
overview of the resistance data.

Table 3.8.2: The materials used for the sodium absorption cell.

<table>
<thead>
<tr>
<th>Material</th>
<th>Sodium resistance</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Windows</td>
<td>Sapphire</td>
<td>Resistant to Na, at least up to 500°C</td>
</tr>
<tr>
<td>Glass to metal seals</td>
<td>Sealing ceramic: Oxide mixture – also called the sealing frit-</td>
<td>Used for high pressure sodium lamps up to 1500K</td>
</tr>
<tr>
<td>Metal to metal junction</td>
<td>Kovar</td>
<td>Resistant to Na up to 500°C, corrosion resistance better than Fe, worse than stainless steel.</td>
</tr>
<tr>
<td>Vessel</td>
<td>304 Stainless steel</td>
<td>Used with Na up to 1500 K</td>
</tr>
<tr>
<td>Gaskets</td>
<td>Silver and Copper</td>
<td>Silver used up to 300°C, Copper up to 500°C.</td>
</tr>
<tr>
<td>Gas fill</td>
<td>Argon</td>
<td>No reaction</td>
</tr>
<tr>
<td>Contamination</td>
<td>Metal oxides from vessel, and water from Argon impurity.</td>
<td>React with sodium</td>
</tr>
</tbody>
</table>

The purity of the argon gas was 99.999%. This means that we introduce 1 Pa of
impurities, mostly water, in the cell. This ‘contamination pressure’ is higher than the sodium
vapour pressure. So before the cell can be used as an atomic notch filter, it has to be
‘neutralised’, i.e. the contamination has to react with the sodium vapour.
The design of the absorption cell is shown schematically in figure 3.8.7. It consists of a standard CF 63/40 reducer tee (a), with two sapphire windows mounted on CF 63 flanges (b₁ and b₂). On the other flange, a CF 40 full nipple is mounted (c), with a cooling block (d) and an ampule crusher (e). At the end of this nipple, a CF mini valve (f) with a glass pipe (g) is mounted. The valve is covered with heat-isolating ceramic material in order to prevent it from being the coldest part of the set-up. The ampule crusher is a thin stainless steel pipe. In this thin pipe, a closed sodium ampule is placed. By bending the tube, the glass will break, and the sodium is released.

The cell is placed in an oven with four compartments: two compartments containing the sapphire windows (1 and 2), one compartment containing the reducer tee (3), and one compartment for the nipple (4). This part is called the cold part. The oven has two windows to decrease the heat losses (h₁ and h₂). These windows are antireflection-coated. The sapphire windows are heated by heating elements on their flanges, and by air heating in the compartments 1 and 2. The temperature of the cold part is controlled by a heating element, which is placed in between the CF mini valve (f) and the cooling block (d). Furthermore, we can cool compartments 3 and 4 by opening the oven at three positions (i₁, i₂, and i₃). The heating of the four compartments is controlled by a microprocessor. This controller monitors the temperature of the coldest part, the windows, the pipe in between the windows, and the heating elements. These temperatures are measured using $K$-type thermocouples.

The temperature of the coldest part of the set-up determines the sodium vapour pressure. At this cold spot the sodium vapour condenses. In our case the liquid sodium is at the position of the cooling block. The controller sets the cold-spot temperature, and the temperature difference of the windows with respect to the real temperature of the cold spot. Using this scheme, the windows can never get colder than the cold spot.

The glass tube is used to fill the cell with sodium and argon. At first the cell is heated and pumped down in order to remove impurities. After that, the sodium ampule is crushed by bending the thin pipe (e). Then, the cell is flushed a few times with argon, and the valve (f) is closed.

The choice of materials is not the only difficulty with the sodium cell. The dynamic behaviour of the cell is very slow, due to the high argon pressure and the fact that all the transport in the cell is governed by diffusion. The characteristic diffusion time is in the order of 4 minutes, which means that the transport of sodium is very slow. The heating up and cooling down of the cell needs special attention due to this slow transport of sodium.
The transmission of the cell is measured with a single-mode ring dye laser. It is measured in the same way as described in subsection 3.8.3. We measured the transmission at a cold-spot temperature of 250°C with a window temperature of 325°C, and at a cold-spot temperature of 300°C with a window temperature of 330°C. The out of band transmission of the cell is 60%. The normalised transmission profile is shown in figure 3.8.8.

![Graph showing normalised transmission of the sodium absorption cell at two different sodium pressures: 0.27 Pa, 250°C (square), and 2.12 Pa, 300°C (circle).](image)

It is clear in this picture that the 250°C profile is more suitable than the one at 300°C. The wings of the 300°C profile extend much further. This means that a significant part of the Thomson scattering signal will be absorbed. In contrast to this, only a small part of the Thomson signal will be absorbed when we heat the sodium cell to 250°C. We note that the measurements shown in figure 3.8.8 are taken using a detector which is not sensitive to the polarisation of the incoming light. This is relevant, since the sapphire windows are birefringent, which means that they change the polarisation of the transmitted (polarised) beam. This change is wavelength-dependent. The optical axis of the sapphire windows should be parallel with the polarisation of the incoming beam. For our absorption cell, this was not the case. However, the window orientation can only be changed by opening the cell. This necessitates evacuating, passivating, cleaning, refilling and neutralising of the cell. This would take too much time, considering the consequences of the mis-orientation of the windows. These consequences are a discussed in the next subsection.

3.8.5 The detection branch
The detection branch is the last part of the set-up we have to discuss. This detection branch consists of two lenses, the sodium cell, a spectrograph, and an ICCD camera. The two lenses make an image of the Thomson scattering signal onto the entrance slit of the spectrograph. The focal distance of these lenses is 250 mm their diameter is 50 mm. The angle between the laser beam and the optical axis of the detection branch is 90 degrees. This choice makes it easy to make an image of the laser beam onto the entrance slit of the spectrograph. The sodium cell is placed in between the lenses. In front of the first lens, a polariser is placed in order to reduce the plasma emission. We built a 30 cm single optical element spectrograph with a 1500 lines/mm holographic aberration corrected concave grating (type IV, Jobin-Yvon 52200460). The efficiency of this grating is approximately 70% for radiation polarised perpendicular to the slit orientation (the polarisation of the Thomson scattering photons). For the other polarisation, the efficiency is 25%. This difference in the grating-efficiency is
important for the overall transmission of the detection branch. The polarisation of the Thomson scattering photons is changed by the mis-oriented sapphire windows. Compared to the transmission for a well-polarised signal, the transmission can decrease 0 to 30\%, depending on the wavelength of the signal. The width of the input slit of the spectrograph is 500 microns and its length is 15 mm. The slit orientation is parallel to the laser beam, so we have a scattering length of 15 mm. At the exit slit of the spectrograph, an intensified charged coupled device (ICCD) camera (Andor Technology ICCD DH 534) is mounted. This camera is able to detect the spectrum from 580 to 600 nm in one go. The quantum efficiency of the photocathode is in the order of 10 \%. The amplification of the intensifier is at maximum $10^4$ counts/photoelectron. This high gain enables us to use photon counting, which improves the signal to noise ratio. The CCD is cooled to $-30^\circ$C in order to reduce the dark current. A pre-trigger pulse from the excimer laser triggers the intensifier. This trigger pulse is created by the remote control unit of the laser. The intensifier is gated for 300 ns around the laser pulse. This gate width is much larger than the width of the laser pulse, since the jitter on the laser pulse is quite high (approximately 100 ns), and the time between the pre-trigger and the laser pulse is drifting on a time-scale of a few minutes. The necessary electronic delay in order to match the gating of the camera with the laser pulse is created by a delay generator (Stanford Research Systems DG535). The ICCD camera is read out in the full vertical binning mode every second. If necessary, several spectra can be accumulated during the measurements. In order to measure the amount of laser power used for the measurement of a Thomson scattering spectrum, a laser power meter (Ophir 30A-P) is placed behind the fluorescent lamp.

The description of the detection branch completes the experimental set-up section. The next subsection describes the first measurements we performed with the set-up.

3.8.6 Stray light reduction.

The purpose of the sodium cell in combination with the ASE filter is the reduction of stray light (and Rayleigh scattering). At first we will calculate the theoretical stray light reduction from the transmission profile of the ASE filter, and the ASE level of the laser. We define the stray light reduction $\eta$ as

$$\eta = \frac{\eta_{ASE}}{\Delta\lambda_{dye}} \cdot t_{ASE\ filter},$$

(3.8.15)

where $\eta_{ASE}$ is defined as the ratio of the total ASE power to the laser power, $t_{ASE\ filter}$ is the rejection of the ASE filter, and $\Delta\lambda_{dye}$ is the width of the dye tuning curve. In this way, $\eta$ describes the amount of stray light that is measured per nanometre. We can compare this $\eta$ to the normalised apparatus profile of the spectrograph. We assume $\eta_{ASE} = 10^{-2}$, $t_{ASE\ filter} = 10^{-5}$, and $\Delta\lambda_{dye} = 40$ nm. Then the stray light reduction $\eta$ is equal to $3 \cdot 10^{-9}$ nm$^{-1}$. Note that the definition of the stray light reduction assumes a uniform dye tuning curve: this is an approximation.

We also measured the stray light reduction. We did this by comparing the stray light level from a discharge tube at two laser wavelengths: 589 nm and 588 nm. The alignment of the ASE filter was changed in order to let it transmit the 588 nm radiation. At 589 nm, the absorption cell absorbs the stray light, at 588 nm it does not. Both stray light spectra were normalised to the total scattered signal at 588 nm. The results of these measurements are shown in figure 3.8.9.
The curve for $\lambda = 588$ nm is basically the normalised apparatus profile of the spectrograph. The curve for $\lambda = 589$ nm shows no maximum at 589 nm, from this we can conclude that the laser beam is totally absorbed in the absorption cell. The curve is much higher than we would expect from the transmission of the ASE filter and the absorption cell. The most likely explanation for this is that the transmission of the ASE filter for wavelengths other than 589 nm is higher than the $10^{-5}$ shown in figure 3.8.5. This can be due to the fact that the prism surfaces were not as clean as during the measurements described in subsection 3.8.3. A second possible reason is the fact that the detection branch was not sufficiently shielded from the ASE filter and the laser, which results in extra stray radiation. A third possible reason is the fact that the ASE level of the laser is not known: imperfect alignment could cause it to be higher than the specification of the manufacturer.

### 3.8.7 Intensity calibration using Raman scattering.

The electron density in a plasma can be deduced from the total Thomson scattered power. Therefore, we need the sensitivity of the detector and the transmission of the detection branch. The total Thomson scattered power $P_T$ is equal to

$$P_T = P_{in} \cdot \left[ \frac{d\sigma}{d\Omega} \right]_T \cdot n_e \cdot L \cdot \delta\Omega \cdot \theta,$$

where $P_{in}$ is the laser power, the factor in between the square brackets is the differential cross section for Thomson scattering, $n_e$ is the electron density, $L$ is the scattering length, $\delta\Omega$ is the solid angle in which the scattered radiation is captured for the measurements, and $\theta$ is the transmission of the optical system from the incoming laser beam up to the ICCD camera, including the sensitivity of the camera. The product of the last three terms in expression 3.8.16 can be obtained by performing an intensity calibration.

The intensity calibration of the Thomson scattering set-up cannot be performed using the commonly applied Rayleigh scattering. This is because of the fact that the sodium absorption cell absorbs the Rayleigh scattering signal. On the other hand, using Raman scattering in Nitrogen, it is possible to calibrate the set-up. We made a glass tube similar to the discharge tube, filled with 9\cdot10^4 Pa nitrogen. Using this tube, the rotational Raman scattering spectrum in Nitrogen is measured. However, the apparatus profile of our
The spectrograph is too broad to resolve the individual rotational Raman lines. Therefore, we used the wavelength integrated Raman scattering signal for the calibration. The total cross section for this scattering process can be obtained by summing over all the rotational lines, and integrating over the wavelength. The total measured scattered power of all the rotational Raman transitions is equal to

\[
P_{\text{RM}} = P_{\text{in}} \sum_{J=0}^{\infty} \left( \frac{d\sigma_{J \rightarrow J'}}{d\Omega} \right) \frac{n_J}{n_{N_2}} \cdot \int G(\lambda - \lambda_{J \rightarrow J'}) d\lambda \cdot n_{N_2} \cdot L \cdot \delta \Omega \cdot \theta
\]

where \( P_{\text{in}} \) is the laser power, \( \frac{d\sigma_{J \rightarrow J'}}{d\Omega} \) is the differential cross section for the rotational transition \( J \rightarrow J' \), \( n_J \) is the density of the rotational state \( J \), \( n_{N_2} \) is the nitrogen density, \( G(\lambda) \) is the normalised apparatus profile. Note that the product \( L \cdot \delta \Omega \cdot \theta \) has the same value as in expression 3.8.16 when we use the same scattering geometry for the Thomson and the Raman measurements. The expression in between the square brackets can be defined as the total rotational Raman scattering cross section \( \sigma_{\text{RM}} \)

\[
\sigma_{\text{RM}} = \sum_{J=0}^{\infty} \left( \frac{d\sigma_{J \rightarrow J'}}{d\Omega} \right) \frac{n_J}{n_{N_2}} = 4.18 \cdot 10^{-34} \text{ m}^2 \text{ sr}^{-1}
\]

where the numerical value is obtained using the value for the anisotropy of the polarisability tensor measured by Penney et al. We used the right angle scattering geometry with polarisation parallel to the laser polarisation. The gas temperature is assumed to be 295 K.

By measuring the wavelength integrated rotational Raman spectrum with the ICCD camera, along with the laser power needed to take this spectrum, we can calculate the factor \( L \cdot \delta \Omega \cdot \theta \) in expression 3.8.16. Note that we have to correct the value for the total rotational Raman scattering cross section for the transmission of the sodium cell. The value we use is slightly lower than the value in expression 3.8.18.

![Fig. 3.8.10: Raman scattering signal, used for the intensity calibration of the set-up.](image)

Figure 3.8.10 shows an example of a rotational Raman spectrum measured in Nitrogen. Note that we can also calibrate the pixel-sensitivity of our ICCD camera with this spectrum, by comparing the experimental spectrum with the theoretical one. It is also possible to obtain the apparatus profile of the spectrograph, and to calibrate the wavelength...
axis using the individual peaks of the measured Raman spectrum. In conclusion, we can calibrate the wavelength axis, the pixel-sensitivity, and absolute scattering intensity of the set-up by measuring one rotational Raman spectrum.

3.8.8 Measurement and alignment procedure and the measurement series

The components of the experimental set-up for the Thomson scattering measurements are discussed in the preceding sections. In this section, we will discuss the measurement procedure, the data analysis, and the specific details for the various measurement series we performed.

For the analysis of the spectral shape of the scattered radiation, we have to calculate the Salpeter parameter $\alpha$, which describes the coherent effects in the Thomson scattering spectrum. At an electron density of $10^{18} \text{ m}^{-3}$, and an electron temperature of 1 eV, which are both typical for the positive column of a mercury-noble-gas discharges, $\alpha$ is equal to $9 \times 10^{-3}$. This value is much lower than unity, which assures that collective effects can be neglected. Therefore, we can assume that we measure incoherent Thomson scattering spectra. This means that the spectral shape of the scattered signal is equal to the shape of the electron velocity distribution function in the direction of the scattering vector.

A. Measurement and alignment procedure

The two lenses in the detection branch are aligned every time the dye laser alignment has been changed. The laser is focussed in front of the detection branch on an anodised brass surface. The scattering from this surface is blue, so it will be transmitted through the sodium vapour absorption cell. The position of the first lens is chosen in such a way that the captured radiation passes the absorption cell as a parallel beam. This beam is guided through the absorption cell, onto the second lens. The position of this lens is chosen in such a way that we obtain a sharp image of the scattered radiation on the entrance slit of the spectrograph. As mentioned before, this alignment procedure is performed after the alignment of the dye laser has been changed. The alignment of the ASE filter is not that critical, it is sufficient to avoid that the beam falls besides an optical component. The fine-tuning of the alignment is performed before every Thomson scattering measurement.

A Thomson scattering measurement always started with aligning the laser beam through the plasma. This is done by shining the laser through a fixed diaphragm in front of the discharge tube, and through a fixed diaphragm behind the discharge tube. Then the focus of the laser beam is positioned at the projection of the slit of the spectrograph. This is done by minimising the width of the individual peaks of the Raman scattering spectrum. When the alignment of the image of the laser beam on the slit of the spectrograph is not good, then the Raman spectrum will be shifted in the measured wavelength direction. So by adjusting the height of the second lens, the image of the laser beam can be aligned exactly in the middle of the slit. Note that the orientation of the slit is parallel to the laser beam. When this fine-tuning procedure is finished, then the nitrogen filled tube is replaced by the discharge tube, which is similar. Small differences between the angle of the input windows of the discharge and the Raman tube will not lead to significant changes in the laser alignment. Furthermore, we tested the influence of the horizontal position of the Raman tube on the alignment. We did not find any change in the alignment and the intensity calibration when changing the horizontal position of the tube.

The influence of misalignment is tested by changing the position of the second lens in the detection branch, and simultaneously measuring the Raman intensity. It has been found that the Raman intensity changes no more than 10% when the laser beam is translated 100
microns in the vertical direction. Figure 3.8.11 shows the relative intensity as a function of the displacement in the measured wavelength axis $\Delta \lambda$. The displacement in the slit $\Delta y$ can be calculated from $\Delta \lambda$, using the linear dispersion of the spectrograph, since it doesn’t magnify the image of the slit.

![Graph showing relative intensity as a function of displacement](image)

Fig. 3.8.11 The relative Raman intensity as a function of the displacement in the measured wavelength and as a function of the displacement in the slit. The symbols represent two independent measurement series.

We note that the slit width is 500 microns while the diameter of the laser focus is approximately 300 microns. Since the width of the slit has been chosen large than the image of the laser beam in the slit, the position of the image of the laser beam is less critical, and the influence of minor misalignment is reduced. The apparatus profile for the scattered radiation is difficult to determine. This apparatus profile is determined by the dimensions of the image of the laser beam in the slit. On the other hand, the apparatus profile for the plasma emission is determined by the width of the slit, since the whole slit is illuminated.

During the Raman and the Thomson measurements, the laser power is monitored continuously with a laser power monitor (Ophir AN/2). This is necessary since the dye solution degrades very fast; the half-life is approximately one hour. The plasma could be operated stable enough to allow us to measure the plasma emission after the Thomson scattering measurements have been performed. For every Thomson scattering measurement we also measured the stray light. However, in most of the cases we only used this stray light measurement in order to check whether the stray light level was low enough to neglect it.

**B. The argon-mercury series.**

This is the first measurement series we performed. Special attention has been given to the reproducibility of the results. With respect to the set-up described in the preceding sections we made some minor changes. The discharge lamp is positioned in a box with a controllable temperature, heated by a hot air tool (Leister Hot-Jet <S>). The mercury vapour pressure in the lamp is controlled using a thermostat bath (Haake FE). The lamp is a U-shaped Borosilicate glass tube with Brewster windows at the ends. The inner diameter of the lamp is 23.2 mm, its outer diameter is 26 mm. The argon pressure is 500 Pa. Just above the electrodes two water jackets are positioned around the tube. With these jackets the tube is cooled locally. The temperature of these cold spots imposes the mercury vapour pressure in the tube. The water jackets are connected to the thermostat bath. A schematic drawing of the discharge lamp is shown in figure 3.8.12. The discharge is sustained by a standard 35 kHz ballast
(Philips BRC 411/01). The discharge current is monitored using a current probe (Pearson electronics inc. Wideband current monitor model 2877), connected to an oscilloscope (Tektronix TDS 210 real-time digital oscilloscope).

![Schematic drawing of the discharge lamp. The grey rectangles represent the water jackets.](image)

The dye laser is operated at the wavelength of 589.0 nm. The laser power used for the Thomson scattering spectra was approximately 1 Watt at a repetition rate of 150 Hz. We used a one-litre dye circulator for the oscillator stage and a two-litre circulator for the amplifier stage of the dye laser. Even though we used the relatively stable Rhodamine 6G dye, we had to change dye at least every hour. The lifetime of the dye is usually expressed in the amount of pump-laser energy needed to decrease the efficiency with a factor two. For our laser combination, this takes 90 Watts×hour. The excimer laser power is approximately 90 Watts, therefore it is necessary to replace the dye every hour. Besides this, the gas fill of the excimer laser had to be replaced daily (appr. 5 hours laser on), and the windows of the excimer laser had to be cleaned every two weeks (appr. 50 hours laser on).

We integrated the Thomson scattering spectrum for 30 minutes in order to improve the signal to noise ratio. Along the horizontal (wavelength) axis the camera is read out per three pixels. The width of the response of the camera to a single photoelectron event is also three pixels. There is no correlation between the phase of the electric field and the timing of the Thomson scattering measurement, since the laser is used as a master for the triggering of the experiment. The influence of the averaging over the phase of the electric field is estimated in Appendix E. The main conclusion of the calculations in that appendix is that the temperature of the averaged Thomson scattering spectrum corresponds to the Thomson spectrum using the averaged temperature. Concluding, we measured the time-averaged electron temperature and density.

The relative transmission of the detection branch as a function of the wavelength is measured with a calibrated tungsten filament lamp (Osram Wi 17/G). All the measured spectra are corrected with this measured transmission profile. The net Thomson scattering spectrum is obtained by subtracting the plasma emission and the stray light from the raw Thomson scattering spectrum. The absolute intensity is obtained using the Raman scattering spectrum, which is performed before every Thomson scattering measurement. We note that with the Raman scattering spectrum, the relative transmission of the detection branch can also be obtained, we used it as a check for the tungsten filament lamp measurement. The Thomson scattering spectrum is then fitted using a Gaussian profile for the electron velocity distribution function.

\[
f_\varepsilon(\lambda) = \frac{n_e}{\sqrt{\pi} \Delta\lambda_{th}} \exp\left(-\frac{(\lambda - \lambda_0)^2}{\Delta\lambda_{th}}\right),
\]

(3.8.19)

where \(\lambda_0\) is the laser wavelength, and the expression for \(\Delta\lambda_{th}\) is given by
\[
\Delta \lambda_{th} = \frac{2\lambda_e}{c} \sqrt{\frac{kT_e}{m_e}},
\]
(3.8.20)

where \(m_e\) is the electron mass. Note that we used the right angle scattering geometry. The fit is obtained using a least-squares method. The sum of the squares of the differences divided by the standard deviation is minimised. In the fitting procedure we only used the wings of the profile. The profile is fitted on a logarithmic scale as a function of the square of the wavelength difference. This is convenient, since the profile transforms into a straight line when we plot it in this way. From the fit, we obtain the electron density and the electron temperature. We note that the wavelength regions where spectral lines of either mercury or argon are situated are discarded in the fitting procedure.

**C. The neon-mercury series**

This series is similar to the argon-mercury series. However, some important changes have been made to the set-up. At first, the ICCD camera is partly covered in order to suppress the emission of the strongest neon lines. We used the spectral region in between the 588.2 and the 594.5 nm lines of neon. Only the wings of these lines could be detected. Assuming a Maxwellian energy distribution function, the distribution function of electrons with energies ranging from 0.5 to 5 eV could be measured. Although we suppressed the strongest lines in this way, ten weak neon lines remain, overlapping the Thomson scattering spectrum. We cannot discard the spectral regions where the Thomson scattering and the plasma emission overlap, since then there will not be enough data points to get a reasonable fit. Other differences with respect to the argon-mercury series are the laser wavelength, the integration time and the gate width. The dye laser is operated at the wavelength of 589.6 nm. We integrated the Thomson scattering spectrum for 60 minutes in order to improve the signal to noise ratio. The ICCD camera is used in the gated mode, the gate width was 200 ns. Finally, the intensity calibration combined with the relative transmission of the detection branch as a function of the wavelength is carried out somewhat different than during the argon-mercury series. Like in the argon-mercury series, we measured this transmission with a tungsten filament lamp. However, since the alignment of the spectrograph is sensitive to mechanical and thermal effects, we always used the measured Raman scattering profile for the determination of the transmission. This profile is measured before every Thomson scattering measurement. In practice, the tungsten filament lamp measurements could also be used. The transmission profile of the detection branch did not change significantly.

**D. The eedf series**

This series is performed in an argon-mercury discharge with an argon pressure of 300 Pa, and a cold-spot temperature of 40°C. These conditions are chosen in order to enhance the influence of the depletion of energetic electrons on the Thomson scattering spectrum. The low argon pressure results in a relatively high electron temperature. The relatively high mercury pressure ensures the suppression of argon emission by the discharge.

We used the same measurement procedure as in the argon-mercury series. However, here the laser wavelength is 589.6 nm and the gate width of the ICCD camera is 200 ns. Furthermore, the Thomson scattering set-up is modified in order to reduce ambiguities as much as possible. First, we inserted an interference band-pass filter (Andover corporation optical filter 589FS10-50) in the detection branch in between the sodium cell and the first lens. The central wavelength of this filter is 591.03 nm. Its 50% transmission bandwidth is 9.54 nm. This filter is positioned in the detection branch in order to suppress the emission of the discharge. Although the suppressed emission is not overlapping the Thomson scattering spectrum, it is influencing the measurements. This is because of the fact that it produces stray
radiation in the spectrograph. Inserting the filter has one disadvantage. The transmission of the filter is approximately 60%, so we lose 40% of the Thomson scattering photons.

The second change in the set-up is related to the intensifier of the ICCD camera. The ICCD camera can be used in a photon counting mode. This mode can be used when extremely low signal levels are measured. The camera is able to detect single photon events. The amplification of the intensifier is very high, for every photoelectron, the CCD camera records more than $10^4$ counts. So every photoelectron results in a ‘spike’ recorded by the CCD camera. However, the height of this spike is not constant. Every multiplication stage of the micro-channel plate (MCP) in the intensifier causes Poisson distributed noise on the amplification. Therefore, although the average amplification is $10^4$ counts per photoelectron, one photoelectron can result in a peak of a few times $10^3$ counts to more than $4\cdot10^4$ counts. Figure 3.8.13 shows the distribution of the number of counts per pixel per photoelectron along with the distribution of the width of the spikes. These distributions are obtained by analysing the full resolution, full vertical binning response of the camera to a very low signal. We assumed that we detect a photoelectron when the number of counts per pixel is higher than 3000 counts.

\[
\begin{align*}
\text{Fig. 3.8.13: The distribution of the amount of counts per pixel per photoelectron (left) and the distribution of the width of the peaks (right).}
\end{align*}
\]

Obviously, this distribution creates additional noise on the CCD read-out. This noise can be reduced by using a photon-counting method. At first we assume that one spike corresponds to one photoelectron, so the signal level should be very low. Second, we define a threshold and assume that we detect one photon when the signal level is above this level. Then every photoelectron corresponds to one count and the noise caused by the intensifier is eliminated. Another advantage of the photon-counting method is that the dark-current and the non-uniformity of the response of the CCD chip are not influencing the measurements anymore. Using a Monte-Carlo type of method, we can calculate the error we introduce by assuming every spike to correspond to one photoelectron. This error can be approximated by the following expression for the amount of ‘missed’ photoelectrons $n_m$

\[
\begin{align*}
n_m = \frac{1}{N} \sum_{i=1}^{N} i = \frac{1}{2N} \cdot n \cdot (n - 1),
\end{align*}
\]

(3.8.22)

where $N$ is the number of superpixels, and $n$ is the amount of photoelectron falling on the CCD camera, i.e. the signal level. For the derivation of this expression, we assume $n$
photoelectrons to fall subsequently on the detector. We neglected the effect of overlapping photoelectrons for the photoelectrons that are still to come.

Theoretically, the intensifier noise is completely reduced. However, there is one complication with real-life intensifiers. The response of one photoelectron is spread out over more than one pixel of the CCD. In order to reduce the influence of this spreading out, we used superpixels of 3×3 pixels of the CCD. For every superpixel, the photon-counting procedure as described above is used. The size of the superpixel corresponds to the size of one photoelectron event. Special care has to be taken for determining the threshold, since a photoelectron can fall on the border between two superpixels. Then one photoelectron is partly detected in two, three, or four superpixels. So one photoelectron results in one, two, three, or four detected counts. This spreading of the response of the camera to one photoelectron over more than one pixel results in the dip in the distribution as shown in figure 3.8.13 at 6000 counts. A more sophisticated data processing routine, in which the full resolution image is scanned for photoelectrons can also eliminate this resulting noise. However, the software for our camera could not be changed in order to incorporate such a data processing routine. At the end of the measurement, the camera image is binned vertically. So the result of the measurement is a single spectrum.

A third change in the data acquisition and processing procedure is the measurement of the plasma emission. In the eedf series, the Thomson scattering and the plasma emission are measured alternately in measurements of 5 minutes each. The measurements are divided into exposure times of 0.5 to 5 seconds, depending on the signal level. We measured the plasma emission and the Thomson scattering alternately in order to eliminate the influence of instabilities of the discharge on the measurements.

The electron energy distribution function can be obtained from the Thomson scattering spectrum. However, this is not as straightforward as it seems. With Thomson scattering, we measure the electron velocity distribution function \( f_v(v_z) \) in the direction of the scattering vector \( (z\text{-axis}) \). This distribution function is different from the distribution function of the absolute value of the velocity \( f(|v|) \). When we integrate this function \( f(|v|) \) over the plane perpendicular to the \( v_z \)-axis, we obtain the velocity distribution function in the direction of the \( z \)-axis. The result can be rewritten in the electron energy distribution function

\[
 f(E)dE = \frac{2}{m} \left. \frac{df_v(v_z)}{dv_z} \right|_{v_z=0} dE
\]

(3.8.23)

Appendix C describes the derivation of this expression. Due to the differentiation of the velocity distribution function \( f_v(v_z) \), the noise on the measurements increases. On the other hand, the influence of a constant background signal under the Thomson scattering spectrum is cancelled out by the same differentiation. We note that the expression for the Maxwellian eedf is

\[
 f_m(E) = \frac{2}{\sqrt{\pi}} \frac{\sqrt{E}}{kT_e^{3/2}} \exp \left( -\frac{E}{kT_e} \right)
\]

(3.8.24)

as can be obtained using expression 3.8.19 and 3.8.23. Finally, the distribution function \( f_0(E) \) is defined as

\[
 f_0(E) = \frac{f(E)}{\sqrt{E}}
\]

(3.8.25)

which is a straight line on a logarithmic scale when the electron energy distribution function \( f(E) \) is Maxwellian.
E. The phase-resolved series

The measurement series, as described in the preceding three subsections are all time-averaged Thomson scattering measurements. The measurements are taken in a stable discharge over a time in the order of an hour. Since the driving frequency is approximately 35 kHz, there is some time-dependence in the electron density and the electron temperature. However, the time-dependency in the Thomson scattering measurements is averaged out. There is no correlation in time between the phase of the discharge voltage and current and the laser pulse. This correlation is absent since the laser is used as the master for the triggering. The measurements described in this subsection are performed using the discharge current as the master for the triggering of the experiment. The discharge current is measured with an oscilloscope (Tektronix 465B). This oscilloscope is triggered on the positive slope at the zero volt level, i.e. at phase zero. The triggering output of the oscilloscope is used to trigger the delay generator. The delay generator triggers the excimer laser and the camera. By setting the delay, the phase-resolved Thomson scattering experiments can be performed. The triggering of the delay generator is inhibited for 7 ms after every trigger pulse in order to limit the trigger repetition rate to 143 Hz. The maximum repetition rate of the laser is 150 Hz. We measured the phase-resolved electron density and electron density in a 1000 Pa neon discharge, at a cold-spot temperature of 40°C, and a current of 400 mA. The driving frequency of the discharge is 36.2 kHz. This series is performed in the same way as the argon-mercury series. However, here the laser wavelength is 589.6 nm, the gate width is 250 ns. The discharge current is measured using a current probe (Pearson electronics inc. Wideband current monitor model 2877). The discharge voltage is measured using two voltage probes (Tektronix Tek P5100 100x), connected to an oscilloscope (Tektronix TDS 520A).

3.8.9 Conclusions Thomson scattering

The main conclusion that can be drawn from the preliminary measurements we performed is that the stray light level can be reduced considerably by introducing a sodium vapour absorption cell in a Thomson scattering set-up. The theoretical stray light reduction has not yet been achieved. However, the results we obtained are promising.

The sodium absorption cell reduces the 589 nm radiation to a level below our detection limit. The remaining stray light is the scattered spectral impurity of the dye laser. Therefore, the stray light reduction is limited by the rejection of ASE by the ASE filter. The ASE that passes the ASE filter is mainly due to scattering of the laser beam at the surface of the prisms, and at dust on the surface of the prisms.
4. Results

4.1 Introduction
This chapter describes the results of the measurements. We start with the discussion of the optical and the electrical measurements performed on the hybrid discharge (section 4.2 and 4.3). The conclusions drawn from these measurements naturally lead to the investigation of radial cataphoresis in the neon-mercury discharge. In order to find the right discharge conditions, we first performed a parameter scan, using emission spectroscopy and electric field measurements (section 4.4). Direct proof of mercury depletion is given in section 4.5 where the mercury ground state density measurements are discussed. In section 4.6, we discuss the emission and excited state density profiles for a series of discharge conditions. Section 4.7 discusses the time-evolution of the emission. Section 4.8 describes the results of the incoherent Thomson scattering experiments. Subsection 4.8.1 describes the electron density and temperature in an argon-mercury discharge. These measurements are the first step to the measurements described in subsection 4.8.2: Thomson scattering measurements in a neon-mercury discharge. Subsection 4.8.3 describes the measurements we performed to obtain the electron energy distribution function in an argon-mercury discharge. Subsection 4.8.4 describes the phase-resolved Thomson scattering measurements. We note that in chapter 5, the results of the experiments will be combined to obtain a more complete view of the two discharges.

4.2 The emission spectrum of the hybrid discharge
The influence of the ccrf discharge on the emission spectrum of the hybrid lamp is investigated as described in the preceding chapter. We used 26 mm diameter lamps, coated with a 4000 K fluorescent powder containing the phosphors BAM, CBT, and YOX. The lamps were filled with neon and mercury. The neon pressure was either 500, 1000, 1500, 2000, or 3000 Pa. The experiments are divided into two series; the ccrf discharge without the If discharge, and the combined discharge. We will start with the discussion of the results of the ccrf discharge measurements.

4.2.1 The ccrf discharge
In this section, we will present the results of the optical measurements for the ccrf discharge. Figure 4.2.1 shows the spectrum of the ccrf discharge with a neon pressure of 3000 Pa at high and low rf input power. The spectrum is taken in the middle of the tube by imaging a small area of the fluorescent layer. In these spectra, we can see the 404.66, the 435.83, and the 546.07 nm spectral lines of mercury. We can also identify a lot of spectral lines of neon in the $\lambda > 575$ nm region. The strongest of these lines is the 585.2 nm line. Furthermore, we observe the radiation of the phosphors BAM, CBT and YOX as relatively broadband spectra around 450, 550 and 600 nm, respectively. The strength of the 436 nm spectral line is approximately the same at low and high rf power. However, the neon peaks in the $\lambda > 575$ nm region change a lot in size.
Fig. 4.2.1: The lamp spectrum for two different rf lamp powers: 16.2 Watt (solid), and 2.7 Watt (dot).

We will use the 436 nm spectral line of mercury as a measure for the mercury emission, and the 724 nm spectral line of neon as a measure for the neon emission. Both lines can be easily separated from the background. The intensity of these lines is plotted in figure 4.2.2 for several neon pressures. The intensity of the neon line is approximately proportional to the rf lamp power. The mercury emission however decreases at a certain rf power. We note that $P_{\text{lamp}}$ is the power dissipated in the lamp, including the glass tube. This lamp power is obtained using the power meter and the results of the impedance measurements.

Fig. 4.2.2: The intensity of the 436 nm spectral line of mercury (left) and the 724 nm line of neon (right) as a function of the rf lamp power for several neon pressures: 500 (square), 1000 (circle), 1500 (up-triangle), 2000 (down-triangle), and 3000 Pa (diamond).

The observed spatial emission pattern reveals some important information. The red light is produced in a very small region near the ccrf electrodes. The glow of the ccrf discharge is always blue. It seems that the electrode regions produce the main part of the emitted light.
4.2.2 The combined rf and lf discharge

The mutual influence of the two discharges can be investigated by looking at the lamp spectrum. We used a lamp with a neon pressure of 3000 Pa. We will again start with the intensity of the spectral lines of mercury and neon. Figure 4.2.3 shows the intensity of the 436 nm and the 724 nm spectral lines.

![Intensity graphs](image)

Fig. 4.2.3: The intensity of the 436 nm (left) and the 724 nm (right) spectral lines as a function of the lf power input, for several forward rf powers: 4.6 (square), 8.0 (circle), 13 (up-triangle), 18 (down-triangle), and 24 Watt (diamond).

The intensity of the mercury line increases sharply at a certain lf power input. This increase can be attributed to the transition of the lf discharge from a cold-cathode to a hot-cathode discharge. Apart from a slightly negative trend, the intensity of the neon lines can be considered to be independent of the lf power input. This means that there is only little influence of the lf discharge on the rf discharge. Figure 4.2.4 shows the correlated colour temperature of the lamp as a function of the lf power input. In this figure, the rf power input is used as a parameter. We note the change in $T_c$ in the transition region.

![Temperature graph](image)

Fig. 4.2.4: The correlated colour temperature as a function of the lf power input, at several forward rf powers: 0 (square), 4.6 (circle), 8.0 (up-triangle), 13 (down-triangle), 18 (diamond), and 24 Watt (cross).
The sudden raise of the intensity at a certain If power input coincides with the sudden change of the lamp colour. This change in colour is caused by the huge increment in the production of white light from the positive column of the If discharge. The change in colour is not caused by a decrease of red light from the rf discharge.

We measured the lamp spectrum at different positions. At first, we will look at the axial dependence of the colour. The colour of the lamp is measured at several axial positions \( x \), at a fixed forward rf power input of 18 Watt and at four different values for the If power input. Figure 4.2.5 shows the results of these measurements in the direct colour representation.

![Fig. 4.2.5: The axial dependence of the colour for four values of the If power input](image)

The colour of the rf discharge alone is not uniform over the length of the tube. If the If power input is not zero, then the axial colour pattern becomes even more non-uniform. At the ends of the tube, the lamp colour becomes whiter, until at a certain value of the If power input, the red spot disappears completely. At the If electrodes, the colour stays reddish, because of the fact that at the electrodes the electric field is high enough for the electrons to excite the neon atoms. For the rf discharge, the non-uniformity can be explained from the fact that the power is coupled to the electrodes from the right-hand side (\( x = 56 \) cm). Besides this, the experimental set-up for the If discharge is not symmetric also. The right-hand side If electrode is grounded, and the left-hand electrode is not grounded.

The radial dependence of the correlated colour temperature of the lamp is also measured. It is measured at different heights at \( x = 28 \) cm. The colour temperature is uniform within 4%, despite the fact that the red light is produced in the sheath region of the ccrf discharge only. This is due to the fluorescent layer that serves as a diffuse reflector inside the tube.

The time dependence of the colour is studied by turning on the rf discharge at a certain point in time and measuring the correlated colour temperature of the lamp at fixed intervals. The discharge will heat the glass tube, resulting in a higher mercury pressure. The excitation rate for mercury atoms will also increase, so the colour temperature will increase. Figure 4.2.6 shows the results for the time-dependence measurements; the correlated colour temperature as a function of the time after the discharge is turned on. The forward rf power was 24 Watt.

It is clear that the correlated colour temperature changes a lot due to this temperature rise. It is this result that changed the research focus into the investigation of the mercury density as a parameter for the control of the colour of a fluorescent lamp. Furthermore, we can conclude that the addition of the ccrf discharge is dramatically decreasing the efficiency of the lamp. The addition of the ccrf discharge at least doubles the dissipated electrical power, while the emitted power stays approximately constant. A lot of rf power is needed to change the colour of the lamp. This also forced us to look at alternative ways to change the colour of fluorescent lamps.
4.2.3 Conclusions
The most important conclusion that can be drawn from the optical measurements is that the efficiency of the production of additional red light by the ccrf discharge is very low. The efficiency of the hybrid lamp drops when the ccrf discharge is turned on. Furthermore, the change in the correlated colour temperature is much too small.

Some interesting features have been found. The neon radiation is produced in the sheath regions only. The glow only produces mercury radiation. From this we can conclude that the amount of high-energetic electrons is much higher in the sheath than in the glow region. In the sheath region, the ionic bombardment of the wall results in significant heating of the discharge tube. This heating has a negative impact on the efficiency of the discharge. Furthermore, the decrease in the intensity of the mercury lines at increasing ccrf power input is peculiar. It has been suggested that this effect can be attributed to mercury depletion. Finally, we can conclude that the influence of the mercury density on the emission of the discharge deserves further investigation.

4.3 Electrical characterisation of the hybrid discharge
In this section, we will discuss the results of the electrical measurements performed in the hybrid discharge. Here, we also divided the measurements into two series: the ccrf discharge without the lf discharge, and the combined discharge. We will start with the characterisation of the ccrf discharge. This characterisation is based on impedance measurements. For these impedance measurements we developed a new way of characterising the stray impedances in the network. In the second subsection, we will discuss the mutual influence of the lf and the ccrf discharge. We used the same lamps as described in the preceding section.

4.3.1 The ccrf discharge
The impedance of the discharge at several neon pressures and input powers is measured using the method described in the preceding chapter. First, the stray impedances are determined. Then the ccrf discharge is struck in the tube. For different input powers, the lamp impedance is measured by recording the settings of the capacitances $C_1$ and $C_2$. In figure 4.3.1, the real and the imaginary part of the lamp impedance are plotted, as a function of the power.
dissipated in the lamp $P_{\text{lamp}}$. The temperature of the tube is around 22°C, the ambient temperature. The heating of the tube by the discharge can be neglected since the discharge is on for a few seconds before the impedance measurements are taken. The measurements are performed for five neon pressures. Note that the power dissipated in the lamp is not equal to the reading of the power meter because of the fact that the resistance $R$ in the matching network dissipates power too. The discharge impedance is not the same as the lamp impedance. The lamp impedance consists of the impedance of the glass walls in series with the discharge impedance. We assumed the impedance of the glass to be a capacitor with a shunt resistance. The tube is made of soda-lime glass doped with iron oxide. The dielectric constant ($\varepsilon_R$) and the dielectric loss factor ($\tan \delta$) of this type of glass are not known, we used the values for soda-lime glass as given in Landolt-Börnstein,$^{93} \varepsilon_R = 7$, and $\tan \delta = 100 \cdot 10^{-4}$.

Using these values, the impedance of the glass is: $Z_{\text{glass}} = 1 - 100j \, \Omega$. We did not measure the impedance of the glass tube without the discharge.

![Graph](image)

\textbf{Fig. 4.3.1:} The real and the imaginary part of the lamp impedance for several neon pressures: 500 (square), 1000 (circle), 1500 (up-triangle), 2000 (down-triangle), and 3000 Pa (diamond).

The electron density can be estimated from the imaginary part of the discharge impedance. In order to make this estimation, a uniform, symmetric model for the ccrf discharge is used, as developed by V.A. Godyak \textit{et al.}$^{17,94}$ In this one-dimensional model, the ion density and the electron density in the glow are assumed to be uniform, the electron density in the sheaths is assumed to be zero. The electron density in the glow is equal to the ion density. Assuming a sinusoidal discharge voltage and current, the sheath impedance will be a capacitor and the glow impedance a resistor with a shunt capacitor. The capacitance of the glow is large enough to represent the glow as a pure resistor. The expression for the sheath capacitance $C_s$ is:

$$C_s = \frac{\varepsilon_0 A}{\bar{d}}, \quad (4.3.1)$$

where $A$ is the electrode area, and $\bar{d}$ is the time-averaged sheath thickness. The expression for the discharge voltage is:

$$|V|^2 = \left(2 \frac{n_e e \bar{d}^2}{\varepsilon_0} \right)^2 + \left(\frac{\omega \bar{d} L}{\mu_e} \right)^2, \quad (4.3.2)$$
where $V$ is the discharge voltage, $\omega$ is the angular rf frequency, $L$ is the gap length ($L = 24$ mm), and $\mu_e$ is the electron mobility in the glow. In expression 4.3.2, the first term on the right-hand side represents the voltage across both sheaths, and the second term the voltage across the glow region. Expression 4.3.2 can be rewritten in:

$$n_e = \frac{\epsilon_0}{2ed^2} \sqrt{|V|^2 - \left(\frac{\omega dL}{\mu_e}\right)^2}, \quad (4.3.3)$$

The discharge voltage is known from the impedance measurements. The electron mobility is calculated from the cross-section for momentum transfer for mercury and neon. It is averaged over the electron temperature range from 1 to 3 eV. The electron density, as calculated using expression 4.3.3, is not at all sensitive to variations in the electron mobility. Therefore the averaged value is good enough for our calculations.

Using expressions 4.3.1 and 4.3.3, the electron density can be estimated from the imaginary part of the discharge impedance. We note that the calculated electron density is an estimate, the geometry is not a parallel-plate one, the electron density is not uniform, the sheath edge is not as steep as we assumed, the ion density is not uniform, the voltages and currents are not sinusoidal, and so on. Therefore, better values for the dielectric constant and the loss tangent of the glass were not necessary. When the dielectric constant was varied from 6 to 8, the electron density did not change more than 30% with respect to the calculation using the dielectric constant 7. Figure 4.3.2 shows the result of the estimation, in this plot, the electron density is plotted as a function of the power dissipated in the discharge $P_{\text{dis}}$. This $P_{\text{dis}}$ is different from $P_{\text{lamp}}$ due to the resistive component of the glass.

Fig. 4.3.2: The estimate for the electron density in the ccrf discharge for several neon pressures: 500 (square), 1000 (circle), 1500 (up-triangle), 2000 (down-triangle), and 3000 Pa (diamond).

The glow conductance cannot be used for the estimation of the electron density for two reasons: (1) the glow dimensions are not known. (2) The calculation is very sensitive to the electron mobility, which changes with changing electron temperature.

When, apart from the glow, the sheath dissipates power too, the sheath impedance is not purely capacitive. We assume that the sheath impedance $Z_s$ can be regarded as a capacitor $C_s$ with a shunt resistance $R_s$. We note that this shunt resistance is very high, resulting in a low resistive part of the sheath impedance:

$$Z_s = \frac{R_s}{1 + j\omega C_s R_s} = \frac{R_s}{1 + \omega^2 C_s^2 R_s^2} + \frac{1}{j\omega C_s} \cdot \frac{\omega^2 C_s^2 R_s^2}{1 + \omega^2 C_s^2 R_s^2} = \frac{1}{\omega^2 C_s R_s} + \frac{1}{j\omega C_s} \quad (4.3.4)$$
It is clear that the shunt resistance does not affect the imaginary part of the sheath impedance. Therefore, the imaginary part of the sheath impedance is equal to the impedance of the sheath capacitance, and the estimation of the electron density will not be changed.

Comparing the figures 4.3.1 and 4.3.2, leads to some interesting results. According to the trend of the electron density, the glow conductance increases with increasing dissipated power. This results in a decreasing glow resistance when we neglect the influence of the changing electron mobility. Figure 4.3.1 indeed shows a decreasing resistive part of the discharge at powers lower than 7.5 Watt. However, above this threshold, the resistive part increases. This cannot be attributed to the glow resistance since it decreases with increasing power. Therefore, we conclude that the resistive part of the sheath impedance increases at high powers.

4.3.2 Mutual influence of the lf and the ccrf discharge.

The influence of the lf discharge on the ccrf discharge can be seen in figure 4.3.3, where the real and the imaginary part of the ccrf discharge impedance are plotted for four different lf currents. It is clear that the influence of the lf discharge on the rf discharge is most pronounced in the low rf power input region. This is due to the fact that the lf discharge supplies the rf discharge with electrons. The ionisation caused by the rf discharge is smaller than the ‘flux’ of electrons from the positive column of the lf discharge. The real part of $Z_L$ becomes smaller and the absolute value of the imaginary part of $Z_L$ gets smaller due to the higher electron density. If the rf input power gets higher, then the influence of these extra electrons and ions is weaker, although they are still influencing the rf discharge.

![Fig. 4.3.3: The influence of the lf discharge on the real (left) and the imaginary (right) part of the ccrf discharge impedance. The lf currents are 0 (square), 50 (circle), 70 (up-triangle), and 90 mA (down-triangle).](image)

The influence of the ccrf discharge on the lf discharge can be investigated by measuring the lf discharge voltage and current. The root mean square ($V, I$) characteristic of the lf discharge with a neon pressure of 1500 Pa is measured at different levels of the rf input power. Figure 4.3.4 shows the results of these measurements. From this plot, it is clear that the presence of the rf discharge has a profound impact on the primary discharge. The ($V, I$) characteristic changes from a negative one (positive column) into a positive one. The current through the discharge did not change significantly when the rf power input changed, at a fixed lf power input. This why the influence of the ccrf discharge on the lf discharge can be
regarded as a change in the conductivity of the positive column of the If discharge. The conductivity of the If discharge increases with increasing power input to the ccrf discharge.

As a first order approximation, the explanation for these curves is that at high rf input powers, the rf discharge generates a lot of electron-ion pairs in comparison with the lf discharge. Because of this, the conductivity for the lf discharge will be much higher. Due to this, the lf discharge voltage will decrease. This effect is more pronounced at lower lf currents, because there the electron density in the lf discharge is lower.

4.3.3 Conclusions
The electron density in the ccrf discharge is estimated from the discharge impedance. This electron density, combined with the electrical measurements show that a significant fraction of the ccrf power is dissipated in the sheath regions, the glass, and the matching network. It is believed that the power dissipated in the sheath is mainly dissipated by the ions bombarding the discharge tube. By optimising the matching network and the electrodes, some power loss can be eliminated. However, the efficiency for the production of neon radiation will stay very low. It seems that the electrons need to be heated excessively to be able to excite neon atoms in a neon-mercury discharge. The presence of mercury cools the electrons very efficiently. It is therefore desirable to look for ways of getting rid of the mercury. Then the cooling is absent and the heating does not have to be this intense. Radial cataphoresis in the positive column of the lf discharge seems a good candidate as a tool for changing the mercury density and for adding neon lines to the emission spectrum. The results of the investigation of the influence of radial cataphoresis and the mercury pressure are discussed in the next section.

4.4 The influence of mercury depletion on the emission spectrum of a fluorescent lamp

4.4.1 Emission spectrum
In order to investigate the influence of mercury depletion on the emission spectrum of the neon-mercury discharge, we performed a parameter scan, looking at the influence of the neon pressure, the mercury pressure, the lamp diameter and the discharge current. The neon
pressure in the investigated lamps was either 300, 500, 1000, or 1500 Pa. The cold-spot
temperature varied from 18 to 37°C. We used lamps with an inner diameter of either 11, 14.1,
or 24 mm. The current ranged from 100 mA up to 2000 mA. The measurements can be
categorised in three series. In series A, the mercury pressure is varied. The lamp diameter was
26 mm. In series B, we measured the spectrum of lamps with several diameters. The neon
pressure in these lamps was 500 Pa. Finally, in series C we measured the emission of lamps
containing neon at several filling pressures, the diameter of these lamps was 26 mm.

As in the case of the hybrid lamp, we will again start with the discussion of the
emission spectrum. The influence of mercury depletion and the subsequent addition of
mercury radiation is obvious in the emission spectrum of the lamp. Figure 4.4.1 shows this
emission spectrum for a lamp with a cold-spot temperature of 18°C, a diameter of 26 mm,
filled with 1000 Pa neon at two different currents, 200 and 1500 mA. In the spectrum at a
current of 200 mA, we can identify the 404.66, the 435.83, and the 546.07 nm lines of
mercury. Furthermore, we observe the radiation of the phosphors BAM, CBT and YOX as
relatively broadband spectra around 450, 550 and 600 nm, respectively. Apart from the fact
that all these contributions are stronger in the spectrum at a current of 1500 mA, we see the
addition of a number of lines in the wavelength region from 580 to 750 nm. These lines are
all neon lines; the strongest is the 640.22 nm line.

Fig. 4.4.1: The emission spectrum of a 1000 Pa neon, 26 mm diameter lamp with a cold-spot temperature of
18°C and the electric currents 200 mA (dot) and 1500 mA (solid).

The correlated colour temperature $T_c$ and the chromaticity coordinates of the 1000 Pa
neon, 24 mm inner diameter lamp are shown in figure 4.4.2 for several cold-spot
temperatures and currents. At low currents, the colour temperature is 4000 K, this is the
colour temperature of the phosphor/mercury spectrum. It is clear that the colour temperature
decreases for higher currents and lower cold-spot temperatures. This decrease is caused by
the addition of neon radiation to the spectrum. From figure 4.4.2 we can conclude that at
certain conditions, we will have radial cataphoresis leading to significant mercury depletion
and moreover, the production of neon radiation. The luminous efficacy of the positive
column with a neon pressure of 1000 Pa ranges from 20 to 80 Lumen/Watt, depending on the
amount of neon radiation emitted by the discharge. Mercury is much more efficient in
producing radiation than neon. Therefore, at low mercury pressures, where a significant
amount of neon radiation is produced, the efficacy is low.
Results

Fig. 4.4.2: Left: The colour temperature for the 1000 Pa neon, 26 mm diameter lamp as a function of the current, for the cold-spot temperatures 18 (square), 20 (circle), 22 (up-triangle), 24 (down-triangle), 27 (diamond), and 30 °C (cross). Right: The colour of the lamp with a cold-spot temperature of 18 and 30 °C in the CIE diagram.

Fig. 4.4.3: The colour of the discharge as a function of the cold-spot temperature and the current. From upper left to lower right (inner diameter in mm, neon pressure in Pa): (24, 300), (24, 500), (24, 1000), (24, 1500), (14.1, 500), (11.0, 500).
Figure 4.4.3 shows the colour of the lamps as a function of cold-spot temperature and current, for six different lamps. Note the small nuances in colour between the different graphs. It is clear that the colour of the lamps is too red, the deviation from the blackbody locus in the CIE diagram is high, as can also be observed in figure 4.4.2.

The intensity of the spectral lines of mercury and neon can be determined by deconvoluting the spectrum with the apparatus profile of the spectrograph, taking into account the possible overlap of spectral lines and the phosphor emission. Figure 4.4.4 shows the intensity of the 404.66, the 435.83, and the 546.07 nm spectral lines of mercury, along with the intensity of the 724.52 nm line of neon as a function of the current through the lamp. The cold-spot temperature was 18°C, the neon pressure was 1000 Pa, and the diameter of the tube was 26 mm. It is clear that the production of neon radiation starts to become significant at a certain current. The current at which the tangent line through the point of inflection of the neon intensity curve coincides with the current axis is defined as the critical current. For the conditions in figure 4.4.4, this critical current is 200 mA.

![Graph showing the intensity of spectral lines of mercury and neon](image)

**Fig. 4.4.4:** The intensity of the 404.66 nm (square), the 435.83 nm (circle), and the 546.07 nm (up triangle) lines of mercury, along with the 724.52 nm line of neon (down triangle).

In the same way, we can determine the critical current for the other experimental conditions. In figure 4.4.5a the critical current is given as a function of the mercury pressure for series A. In figure 4.4.5b, the critical current is given as a function of the square of the inner radius $R$ of the tube for series B. In figure 4.4.5c the critical current is given as a function of the neon pressure for series C.

We can conclude on the one hand that the critical current is approximately proportional to the mercury pressure and the square of the radius of the tube. On the other hand, the critical current is not proportional to the reciprocal neon pressure as can be concluded from figure 4.4.5c. These conclusions were already predicted in section 2.4.
Fig. 4.4.5a: The critical current as a function of (a) the mercury pressure for the neon pressures 300 (square), 500 (circle), 1000 (up triangle), and 1500 Pa (down triangle).

Fig. 4.4.5b: The critical current as a function of the squared radius for the cold-spot temperatures 18°C (square), 20°C (circle), 22°C (up triangle), 24°C (down triangle), 27°C (diamond), and 30°C (cross).

Fig. 4.4.5c: The critical current as a function of the neon pressure, same legend as figure 4.4.4b.
4.4.2 Electric field

The results of the electric field measurements are shown in figure 4.4.6. Note that figure 4.4.6 shows the root mean square (rms) values of the electric field. The slope of the $E$-$I$ characteristic of a fluorescent lamp is negative under normal conditions. However, in figure 4.4.6, we can see that this slope can also be positive. This is due to the fact that a neon discharge requires a higher electric field to be able to sustain itself than a neon-mercury discharge that is dominated by mercury.

![Electric field measurements](image)

Fig. 4.4.5: The electric field (rms) as a function of the current for the 1000 Pa neon, 26 mm diameter lamp for several cold-spot temperatures (left), and the electric field (rms) as a function of the current for a cold-spot temperature of 18 °C, and several neon pressures (right).

4.4.3 Conclusions

The results described in this section show the effect of the radial cataphoresis process on the emission spectrum and the electric field. The knowledge where the effect of the radial cataphoresis process is significant is very useful for the choice of the discharge parameters for further experiments, and for the application of the discharge in a variable colour fluorescent lamp. Especially the fact that the noble-gas emission can be controlled by the discharge current is important. Furthermore, the trends in the critical current as predicted in section 2.4 using the mass balance for mercury are similar to the measured trends.

4.5 The mercury density profile in a Ne-Hg discharge

In the preceding section, mercury depletion is observed indirectly in the addition of neon lines to the emission spectrum of the discharge. To give direct proof of the depletion, we measured the transmission of the discharge for 254 nm ultraviolet radiation. The UV transmission measurements are performed in a 24 mm inner diameter tube, filled with 1000 Pa neon and 5 mg mercury. The cold-spot temperature was 18°C. The tube was made of quartz. The lateral profile of the mercury density is measured for the discharge currents 100, 200, and 400 mA. We note that the colour temperature of the lamp decreases significantly with the discharge current under these circumstances, as can be seen in figure 4.4.2. Figure 4.5.1 shows the measured lateral profiles of the line-integrated mercury density, along with the fits.
As discussed in the preceding chapter, we used the following expression for the fit of the radial profiles:

\[ n_{Hg}(r) = a + b \cdot r^n \]  

(4.5.1)

The deviation from the fit gives an idea of the magnitude of the experimental noise. Figure 4.5.1 also shows the density profiles resulting from the lateral profiles. The symbols represent the directly transformed radial profiles and the lines represent the fits. The colour temperatures of the lamp at 100, 200, and 400 mA are 4000, 3800, and 3000 K respectively, as can be seen in figure 4.4.2. It is clear that a lot of mercury has to be removed for a significant change in the colour temperature. We note that the observed depletion cannot be attributed to gas heating. Based on temperature measurements by Kenty et al.,\textsuperscript{97} it can be estimated that gas heating results in a decrease in the atom density on the axis of at maximum 10, 20, and 40% for the currents 100, 200, and 400 mA, respectively.

We can conclude that the influence of the radial cataphoresis process on the mercury density profile is significant. Especially the fact that the mercury density profile can be controlled by the discharge current is important.

4.6 The emission profiles, and the excited state density profiles in a Ne-Hg discharge

The emission of a lamp coated with a fluorescent powder gave us an idea of the colour change that can be obtained by using the process of radial cataphoresis. From the measured mercury density profile, we can conclude that the mercury density has to be very low before the discharge starts producing a significant amount of neon radiation. In order to see where the mercury radiation and the neon radiation are produced in the lamp, and to verify whether the emission profile correlates with the mercury ground state density, we performed spatially resolved emission measurements. These measurements are done as described in the preceding chapter. We measured the emission from lamps filled with mercury and either 300, 1000, or 1500 Pa neon. The cold-spot temperature was either 18, 22, 27, or 30°C. The current was either 100, 150, 200, 300, or 400 mA. First, we will discuss the emission profile. For two
specific spectral lines of mercury and neon, we will then discuss the current, mercury pressure, and neon pressure dependency.

### 4.6.1 Spatially resolved emission; cross section of the discharge for several currents.

We will start with the emission of the lamp with 1000 Pa neon, a cold-spot temperature of 18°C, and the electric currents 100, 150, 200 and 400 mA. The radial profile of this emission is converted into a bitmap containing the colour distribution cross-section of the discharge, as described in section 3.6. The radial profile of the mercury emission spectrum is constructed using the emission coefficients for the spectral lines of mercury. The neon spectrum is constructed by using the \( y=0 \) spectrum and scale it to the 588.2 nm spectral line. This construction saves a lot of calculation time. It can be used since the shape of the normalised neon spectrum stays constant within 10%. Note that the influence of self-absorption on the intensity is also in the order of 10%. These errors are negligible for the construction of the cross-sections of the discharge. There are two reasons for this. At first, we do not give intensity information in the cross-section. Furthermore, the colour of monochromatic radiation with a wavelength in the range of 620 nm to 775 nm is virtually the same. Small changes in the shape of the neon spectrum will therefore not result in a different colour, as can be seen in the CIE diagram (fig. 2.2.1).

![Fig. 4.6.1: The cross section of the emission at 100, 150, 200, and 400 mA.](image)

The cross-sections are given in figure 4.6.1. The cross-sections show the development of a red area in the middle of the discharge. The size of this area increases with increasing current. This is due to the fact that at higher currents, the mercury density profile gets more depleted in the middle part of the discharge. The development of the red area causes the overall colour of the lamp to shift towards red.

### 4.6.2 The atomic state density profiles.

We can obtain the density of the upper state of the transitions from the emission coefficient \( \varepsilon_{ki} \), using expression 3.6.2. For the discharge with a neon pressure of 1000 Pa, a cold-spot temperature of 18°C and a current of 400 mA, we can use the 435.83, the 576.96, and the 491.60 nm spectral lines to obtain the density of the \( 7^3S_1 \), the \( 6^3D_2 \), and the \( 8^1S_0 \) state, respectively. For the optically thin lines of neon we can do the same calculation. All the neon lines show the same behaviour. Figure 4.6.2 shows the density of the \( 7^3S_1 \), the \( 6^3D_2 \), and the \( 8^1S_0 \) states of mercury, and the \( 2p_1 \), the \( 2p_2 \), the \( 2p_{10} \), the \( 3d_3 \), and the \( 4d_5 \) states of neon, as obtained from the emission measurements. The lines represent the first lateral-to-radial transformation method, the symbols represent the second method.
The electron density in the discharge is expected to have a somewhat flattened Bessel profile. This profile and the mercury ground state density (fig. 4.5.1) explain the shape of the excited state density profiles of mercury. For the production of excited mercury atoms, ground state mercury and electrons are needed. Since the ground state Hg density profile is hollow, the excited state density profiles will also be hollow. Furthermore, since the electron density is approximately zero at the walls, the excited state density profiles will also be approximately zero at the wall.

The excited state density profiles for neon are bell shaped. The main reason for this is the fact that the $2p$ states are mainly populated from the $1s$ states. Therefore, for the production of neon atoms excited to the $2p$ states, electrons and neon in one of the $1s$ states are needed. The density profile of both of these particles is approximately Bessel shaped, so the $2p$ state density will be bell shaped. A second reason for the bell shaped profile is the fact that the resonant states will also have a bell shaped density profile due to radiation trapping. The third reason for the bell shaped profile can be the quenching of neon metastable atoms due to Penning ionisation of mercury, which results in a bell shaped density profile of metastable neon.

In the remainder of this section, we will only give the $6^3D_2$ density of mercury, along with the $2p_2$ or the $3d_3$ density of neon, since they can be obtained for all the experimental conditions.

**4.6.3 The Hg $6^3D_2$ density and the Ne $2p_2$ density: current dependency.**

The mercury $6^3D_2$ density and the neon $2p_2$ density can be obtained from the emission measurements. We measured this emission using a discharge with 1000 Pa neon, and a cold-spot temperature of 18 °C, at currents 100, 150, 200, and 400 mA. The results of these measurements are given in figure 4.6.3, where the two excited state densities are given. It is clear that the mercury $6^3D_2$ density profile becomes more hollow when the current increases. This is due to the mercury depletion, which is more pronounced at higher currents. The $6^3D_2$ density gradient at the wall increases with increasing current. This means that the mercury ground state density gradient and the mercury ion density gradient also increase with increasing current. The neon $2p_2$ density increases with the discharge current.
Fig. 4.6.3: The Hg $6^3D_2$ density profile (left), and the Ne $2p_2$ density profile (right) for four currents: 100 (square), 150 (circle), 200 (up-triangle), and 400 mA (down-triangle).

4.6.4 The Hg $6^3D_2$ density and the Ne $2p_2$ density: mercury pressure dependency.

We measured the emission spectrum of the discharge at four different cold-spot temperatures: 18, 22, 27, and 30 °C. The neon pressure was 1000 Pa, the current was 400 mA. The results of these measurements are shown in figure 4.6.4, where the Hg $6^3D_2$ density profile and the Ne $2p_2$ density profile are given.

Fig. 4.6.4 Left: The Hg $6^3D_2$ density profile (left) for four cold-spot temperatures: 18 (square), 22 (circle), 27 (up-triangle), and 30 °C (down-triangle). Right: The Ne $2p_2$ density profile for three cold-spot temperatures: 18 (square), 22 (circle), and 27 °C (up-triangle).

It is clear that the mercury depletion is more pronounced at lower mercury pressures. This is due to the fact that a decrease of the mercury density has more effect when the mercury density is low than at a high mercury density. Note that the gradient in the $6^3D_2$ density is approximately the same for all the mercury pressures. This means that the gradient in the mercury ground state density and the mercury ion density are approximately constant at changing mercury pressures. The neon $2p_2$ density decreases with increasing mercury pressure.
4.6.5 The Hg $^3D_2$ density and the Ne $3d_3$ density: neon pressure dependency.

We have measured the emission spectrum of three different lamps. The neon pressure in these lamps was 300, 1000, and 1500 Pa. The current through the lamp was 400 mA, the cold-spot temperature was 18°C. The Hg $^3D_2$ density profile and the Ne $3d_3$ density profiles are given in figure 4.6.5 for the three neon pressures 300, 1000, and 1500 Pa.

It is clear that the production of neon radiation is much more efficient at high neon pressures. This is probably due to a higher electron density in the discharge at higher neon pressures, resulting in a higher degree of depletion. For mercury radiation the opposite holds. The higher the neon pressure, the less mercury radiation. This is probably due to the increase of the energy loss in elastic collisions of electrons with neon atoms, and the higher degree of depletion.

![Graphs showing density profiles](image)

Fig. 4.6.5: The Hg $^3D_2$ density profile (left), and the Ne $3d_3$ density profile (right) for three neon pressures: 300 (square), 1000 (circle), and 1500 Pa (up-triangle).

4.6.6 Conclusions

The results of the spatially resolved emission measurements show where the neon and the mercury radiation is produced. The neon emission profiles are bell shaped, while the mercury emission profiles are hollow. Both profiles show the influence of the mercury density on the emission of the discharge. The current and pressure dependency of the profiles gives new information on the radial cataphoresis process. The trends are consistent with the trends in the mercury ground state, the critical current, and the electric field.

4.7 Time evolution of the emission of a Ne-Hg discharge and the influence of amplitude modulation of the discharge current

Section 4.4 and 4.6 describe the time-averaged emission measurements we performed using a neon-mercury discharge at a driving frequency in the kilohertz range. The modulation of this emission is averaged out during these measurements. This section describes the results of the phase-resolved emission measurements. Furthermore, we will also discuss the influence of amplitude modulation of the discharge current on the emission spectrum.
4.7.1 Results phase-resolved emission measurements

The emission spectrum of a 50 kHz sinusoidal driven neon-mercury discharge is measured phase-resolved at a cold-spot temperature of 18°C, and a current of 400 mA. The neon pressure was 1000 Pa. Figure 4.7.1 shows the intensity of some spectral lines of mercury and neon. The corresponding waveform of the current is sinusoidal.

![Figure 4.7.1: The intensity of the 404.66 nm (square), the 435.83 nm (circle), and the 546.07 nm (up triangle) spectral lines of mercury, along with the 724.52 nm (down triangle) and the 640.22 nm (diamond) spectral lines of neon as a function of time. The drawn line represents the absolute value of the current as a function of time.](image)

The emission is shifted approximately 1 µs with respect to the current. This is attributed to the phase shift between the modulation of the electron temperature and the discharge current as found by Drop et al.⁹⁸

4.7.2 Amplitude modulation of the discharge current

In order to de-couple the discharge current from the dissipated power, we modulated the amplitude of the discharge current. The modulation frequency was either 50 or 100 Hz. We used a square waveform: the discharge is on for a time $\tau$, and off for the remaining part of the modulation period. The discharge current during the burst was 400 mA, sinusoidal at 50 kHz. The cold-spot temperature was 18°C and the neon pressure was 1000 Pa. We first discuss the time-resolved measurements. The discharge was on for 50% and off for the remaining 50% of the modulation period. Figure 4.7.2 shows the intensity of two spectral lines as a function of time. At $t=0$, the discharge is switched on. Figure 4.7.2 also shows the voltage and the current at the start of the burst. The large voltage in the first 50 kHz period is due to the re-ignition of the discharge. Breakdown requires a higher voltage than sustaining a discharge. The high electric field results in the enhancement of the production of neon radiation, as can be seen in the left-hand side plot in figure 4.7.2.

The increase of the neon line and the decrease of the mercury line with time can be attributed to the radial cataphoresis process, which depletes the discharge from mercury. A lower mercury density results in more neon radiation and less mercury radiation. The difference between the intensity of the spectral lines in the CW mode and the AM mode can be fitted using an exponentially decreasing function. The resulting characteristic time for mercury transport is 10 ms for 100 Hz modulation and 8 ms for 50 Hz modulation. These times are of the same order of magnitude as the characteristic time for ‘free’ diffusion, which is 13 ms, as calculated using expression 2.4.11. We used the value for the diffusion
coefficient for mercury atoms measured by Redko et al.⁹⁹ The characteristic length for diffusion is $R/2.4$, this is the characteristic length for diffusion for the lowest-order Bessel function $J_0(r)$. We note that the diffusion equation is a Bessel equation.

The deviation between the two values for the characteristic time for radial cataphoresis is probably due to the fact that the difference between the intensity of the spectral lines in the CW mode and the AM mode is not exactly decaying exponentially. Furthermore, the time the discharge is off differs. It is 5 ms for the 100 Hz, and 10 ms for the 50 Hz modulation. At 50 Hz modulation, the time the discharge is off is comparable to the characteristic time for diffusion, while at 100 Hz modulation the time that the discharge is off is much lower. Therefore, at the start of the burst, the mercury density profile will be more uniform in the 50 Hz case than in the 100 Hz case. A result of this more uniform density profile in the 50 Hz case is a lower intensity of the neon lines at the start of the burst, as can be seen in figure 4.7.2. It is clear from figure 4.7.2 that the average intensity of the neon lines during the burst is lower than the intensity in the CW mode with the same current.

We also measured the time-averaged spectrum of the 1000 Pa neon lamp with phosphor coating in the CW and the AM mode. From these spectra, we calculated the colour temperature. In the AM mode, the discharge was on for a time $\tau$. The current during the burst was chosen in such a way that the current averaged over one modulation period, was constant. Since the discharge voltage didn’t change that much, the power dissipated in one modulation period remained constant. Therefore, also the luminous flux of the discharge was approximately constant. Figure 4.7.3 shows the colour temperature and the intensity (radiometric) as a function of the current. For the AM mode this is the current during a burst. We note that for the AM mode the average current during one modulation period is kept constant at 200 mA (rms). The cold-spot temperature was 18°C, the driving waveform is sinusoidal, at 50 kHz. The influence of the modulation is very clear in both plots. The colour temperature decreases less with the current for the AM mode than for the CW mode. Besides this, the colour temperature decreases less with the current for the 100 Hz AM mode than for the 50 Hz AM mode. Both effects are caused by the fact that during a fraction of the time needed for the transport of mercury the discharge is not producing neon radiation. The intensity of the lamp is approximately constant when we use the AM mode, while it increases significantly for the CW mode.

Fig. 4.7.2: The intensity of the 546.07 nm line (square) of mercury and the 640.22 nm line of neon (circle). The full symbols represent the 50 Hz modulation, the open symbols represent the 100 Hz modulation. The crossed symbols represent the intensity of the lines in the continuous wave mode.
4.7.3 Conclusions

The results of the phase-resolved emission measurements show that the intensity of the spectral lines of both neon and mercury is modulated. This modulation suggests that the electron energy distribution function is also modulated. Furthermore, we can conclude that it is possible to vary the colour of the lamp independently from the luminous flux by modulating the amplitude of the discharge current. The colour temperature variation is quite low for the discharge conditions we studied. However, decreasing the characteristic times for radial cataphoresis and diffusion will have a positive influence on this colour temperature variation. Decreasing the neon pressure in the tube and/or decreasing the tube diameter will result in a decrease of the characteristic times.

4.8 Results of the Thomson scattering experiments

The emission measurements have shown for which discharge conditions the amount of high-energetic electrons ($E>16.5$ eV) is sufficient to have a significant amount of neon radiation produced in the discharge. Furthermore, the time-resolved emission measurements have shown that the electron energy distribution function is modulated. In other words, the emission measurements have been used as a qualitative probe for the high-energetic electrons.

The distribution of the bulk of the electrons ($E<4$ eV) has been measured using Thomson scattering. This section describes the results of these Thomson scattering measurements.

4.8.1 The electron density and temperature in an Ar-Hg discharge

Thomson scattering is very weak and the plasma emission from a neon-mercury discharge in the wavelength region where we try to measure the Thomson scattering spectrum is very high. Therefore, we first performed the Thomson scattering experiments in an argon-mercury discharge. The plasma emission is much lower for this argon-mercury discharge. We can use the results of these Thomson scattering experiments to find the trends in the electron density.
and temperature in the neon-mercury discharge, since the behaviour of the argon-mercury and the neon-mercury discharge will be similar. We are especially interested in the process of radial cataphoresis and its influence on or cause in the electron energy distribution function. Even though the discharge conditions where depletion takes place in an Ar-Hg discharge will be different from the Ne-Hg case, as a first order approximation, we can use the same discharge conditions in the argon-mercury and the neon-mercury discharge.

The Thomson scattering measurements have been performed in three series. Table 4.8.1 shows the experimental conditions for each series. Series A is performed at a cold-spot temperature of 18°C. This temperature corresponds to a mercury vapour pressure of 0.14 Pa. The electrical current through the lamp was 200, 300, and 400 mA. In this series, we measured the radial profiles of the electron density and the electron temperature. Series B is performed at the cold-spot temperatures of 18°C and 40°C. The cold-spot temperature of 40°C corresponds to a mercury vapour pressure of 0.85 Pa. We measured the electron density and the electron temperature at the tube axis for discharge currents between 200 and 400 mA. Series C is performed at cold-spot temperatures between 18°C and 50°C. The cold-spot temperature of 50°C corresponds to a mercury vapour pressure of 1.76 Pa. The discharge current was 400 mA. We measured the electron density and the electron temperature at the tube axis.

<table>
<thead>
<tr>
<th>Series</th>
<th>A</th>
<th>B</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cold-spot temperature (°C)</td>
<td>18</td>
<td>18, 40</td>
<td>18 - 50</td>
</tr>
<tr>
<td>Mercury pressure (Pa)</td>
<td>0.14</td>
<td>0.14, 0.85</td>
<td>0.14 - 1.76</td>
</tr>
<tr>
<td>Current (mA)</td>
<td>200, 300, 400</td>
<td>200 - 400</td>
<td>400</td>
</tr>
<tr>
<td>Measured quantities</td>
<td>$n_e(r), T_e(r)$</td>
<td>$n_e(0), T_e(0)$</td>
<td>$n_e(0), T_e(0)$</td>
</tr>
</tbody>
</table>

The measurements are performed as described in the preceding chapter. We fitted the measured spectra using a Maxwellian electron velocity distribution function. For the fit, we used the wavelength regions from 586.5 to 587.8 nm and from 590.9 to 591.4 nm. These regions correspond to the energy ranges from 0.5 to 2.3 eV and from 1.4 to 2.2 eV, respectively. Therefore, the electron temperatures we report correspond to electrons with an energy lower than 2.3 eV. We note that the experimental noise is too high to verify whether or not the electron velocity distribution function is Maxwellian outside the fitted region.

An example of a measured Thomson scattering spectrum is shown in figure 4.8.1. The fit of the spectrum is also shown in this figure. For the fit, we used a Maxwellian electron velocity distribution function, the measured transmission of the sodium vapour absorption cell, and the apparatus profile of the spectrograph. This apparatus profile is not exactly known, as discussed in subsection 3.8.8. Therefore, the fit deviates from the measurement in the vicinity of the absorption lines of sodium at 589 and 589.6 nm. Furthermore, it is clear that the fit is good in the wings of the Thomson scattering profile. The electron density and temperature resulting from the fit in figure 4.8.1 are $1.0 \times 10^{18}$ m$^{-3}$, and 0.94 eV, respectively.
Fig. 4.8.1: Example of a Thomson scattering spectrum. The symbols represent the measurements, the dashed curve is the Maxwellian velocity distribution function, and the full curve is the velocity distribution multiplied by the transmission of the sodium cell, convoluted with the apparatus profile of the spectrograph.

Figure 4.8.2 shows the results of the measurement series A. The electron density and the electron temperature are plotted as a function of the radial position $r$. The drawn lines are Bessel profiles, fitted to the electron density measurements. The zero-order Bessel function is the diffusion solution according to the Schottky theory. The agreement is good.

Figure 4.8.2 shows that, within the experimental uncertainty, the measured electron temperature is not dependent on the radial position. We note that the measurements at the radial position of 10 mm are very difficult to obtain. The electron density is very low, and the stray light level is extremely high. On the one hand, the scattering of the laser beam at the tube wall is very high, the laser beam could be seen very clearly with the naked eye. On the other hand, the Thomson scattering signal contains on the average only one photon per laser pulse.

For series B we only measured the electron properties at the tube axis, i.e. at $r=0$. The dependency on the current $I$ is investigated. Figure 4.8.3 shows the results of these
measurements. The electron density is proportional to the current through the lamp. The electron temperature decreases with increasing current. This decreasing electron temperature is due to the multistep ionisation of mercury, which starts becoming important at higher electron densities. The multistep processes allow the electron energy to decrease since the energy difference to overcome by the electrons decreases.

![Graph](image)

Fig. 4.8.3 The electron density (left), and the electron temperature (right) as a function of the current (series B).

The results of measurement series C are shown in figure 4.8.4. The electron density and the electron temperature both decrease with increasing cold-spot temperature $T_{\text{Hg}}$. This is due to the increasing influence of mercury in the ionisation and excitation processes. This increasing influence is clear in the emission spectrum of the discharge, as can be seen in figure 4.8.5, where the emission of the 585.925 nm line of mercury and the emission of the 603.213 nm line of argon is plotted as a function of the cold-spot temperature.

![Graph](image)

Fig. 4.8.4: The electron density (left), and the electron temperature (right) as a function of the cold-spot temperature (series C).
Fig. 4.8.5: The emission of the 585.925 nm line of mercury and the emission of the 603.213 line of argon as a function of the cold-spot temperature.

The outer wall temperature of the discharge tube at the position where the Thomson scattering spectra are obtained is plotted in figure 4.8.6, for the series A, B, and C. Note that series A is measured without increasing the ambient temperature, this series is taken at an ambient temperature of 27°C. Series B is taken at an ambient temperature of 48°C. Series C is taken at an ambient temperature of 48°C, except for the measurements at 45 and 49.5°C, these were taken at an ambient temperature of 69°C.

Fig. 4.8.6: The outer wall temperature of the tube for series A, and B at \( T_{\text{Hg}} = 18°C \) (square), B at \( T_{\text{Hg}} = 40°C \) (circle), and C (cross).

The accuracy in the measurements is limited by systematic and a statistical errors. Systematic errors reduce the accuracy of the absolute value of the measured quantities, while the statistical errors reduce the reproducibility of the results. We will start with the discussion of the error in the electron density followed by an analysis of the error in the electron temperature. We note that the reproducibility of the measurements is quite good, as can be seen in figure 4.8.3 and 4.8.4, where several measurements are performed for the same experimental condition.
**Electron density.** The absolute value of the electron density is dependent on the theoretical cross-section used for rotational Raman scattering. The error in the cross-section is 8%. The uncertainty in the nitrogen pressure in the tube is approximately 10%. The error this uncertainty introduces is however reduced since we compared the nitrogen tube measurement to rotational Raman measurements in air. These errors result in a total error in the absolute value of the electron density of approximately 15%. In addition to the systematic error, we introduce a statistical error, i.e. noise in the experiment itself. The first source is the power monitor reading in the Raman and Thomson scattering experiments. The inaccuracy of the power measurements is estimated to be 5 percent. The second source is the noise on the plasma emission, which causes an additional error, since it determines the baseline of the Thomson spectra. This inaccuracy is estimated to be a few percent. The last source is the alignment of the set-up. Misalignment introduces an error, since small changes in the position of the laser beam result in a different transmission of the detection branch. This inaccuracy is estimated to be 15 percent at maximum. The misalignment is checked after each measurement by performing a second Raman scattering experiment. The total statistical error in the electron density measurements adds up to roughly 25 percent. This relative inaccuracy is however reduced by measuring more than one spectrum at the same experimental conditions.

**Electron temperature.** The systematic error in the absolute value of the electron temperature is negligible compared to the experimental noise. The influence of the apparatus profile is in the order of 1%. Furthermore, the wavelength axis could be calibrated very precisely. The experimental noise in the electron temperature is caused by two factors. At first, the low amount of Thomson scattering photons results in a low signal to noise ratio per wavelength interval. Second, the influence of the plasma emission is not negligible in the structure of the Thomson scattering spectra. Especially the presence of spectral lines of argon and mercury that overlap the Thomson spectra is important. The total error in the electron temperature measurements is in the order of 10 to 20%.

Comparison of the present results with the Langmuir probe measurements performed in the past is hard since the discharge conditions are not the same. Verweij performed an extensive survey of an argon mercury discharge at several currents, argon pressures, and mercury pressures. The inner diameter of his discharge lamps was 36 mm, which is significantly different from the 23.2 mm we have. Denneman *et al.* investigated the argon mercury discharge with an inner diameter of 24 mm. However, the argon pressure in their discharge tubes was 400 Pa, which is different from the 500 Pa we have. Wani determined the electron energy distribution function in an argon mercury discharge with an inner diameter of 24 mm. The argon pressure in Wani’s discharge tubes was 333 Pa, which is also different from the 500 Pa we have. Table 4.8.2 gives a summary of the results of the measurements of Denneman and Wani, and compares them to our results.

Table 4.8.2: Comparison between the present values for the electron density and the electron temperature and the values found in literature.

<table>
<thead>
<tr>
<th>$T_{e}$ (°C)</th>
<th>$p_{Ar}$ (Pa)</th>
<th>$I$ (mA)</th>
<th>$n_e$ ($m^{-3}$)</th>
<th>$T_e$ (eV)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>42</td>
<td>333</td>
<td>400 (dc)</td>
<td>$6.2 \cdot 10^{17}$</td>
<td>1.5</td>
<td>28</td>
</tr>
<tr>
<td>42</td>
<td>400</td>
<td>400 (dc)</td>
<td>$8.0 \cdot 10^{17}$</td>
<td>1.1</td>
<td>27</td>
</tr>
<tr>
<td>40</td>
<td>500</td>
<td>400 (rms 35 kHz)</td>
<td>$1.0 \cdot 10^{18}$</td>
<td>0.9</td>
<td>This work</td>
</tr>
</tbody>
</table>
It is clear that the difference between our electron density and temperature and the values obtained by Denneman and Wani is mainly caused by the higher argon pressure in our discharge. The same trend as a function of the argon pressure is already observed by Verweij.  

The results of the measurements show that it is possible to measure the electron density and temperature in an Ar-Hg positive column using Thomson scattering. The reported measurements are the first non-intrusive, spatially resolved electron density and temperature measurements performed in an Ar-Hg positive column. The measurements give valuable information on the electron density and the electron temperature in the Ar-Hg discharge. The next challenge is performing Thomson scattering in a Ne-Hg discharge, where the plasma emission is very high.

4.8.2 The electron density and temperature in a Ne-Hg discharge

The Thomson scattering measurements in the argon-mercury discharge do not show an increase in the electron temperature at higher currents at the cold-spot temperature of 18°C. However, the increase of the electron temperature when the cold-spot temperature is lowered is well pronounced. This is most likely caused by the fact that the electron impact ionisation and excitation of argon requires electrons with a higher energy than the ionisation and excitation of mercury. The increase of the intensity of the 585.925 spectral line of argon when the mercury pressure is lowered is a proof of the existence of excitation of argon. Since we do not have more data on the emission and density profiles in the argon-mercury discharge, returning to the neon-mercury discharge seems logical in order to investigate the radial cataphoresis process.

As described in the preceding chapter, after taking the necessary arrangements, the Thomson scattering experiments can also be performed in a neon-mercury discharge. These measurements are performed in a U-shaped tube with an inner diameter of 23.2 mm. The neon pressure in the tube was 1000 Pa. We performed five measurement series. Series A is performed at a cold-spot temperature of 18°C. This temperature corresponds to a mercury vapour pressure of 0.14 Pa. The electrical current through the lamp was 100, 200, and 400 mA. In this series, the radial profiles of the electron density and the electron temperature are measured. The series B, C, D, and E are performed at the cold-spot temperatures of 18, 24, 30, and 40°C, respectively. The corresponding mercury vapour pressures are 0.14, 0.24, 0.39, and 0.85 Pa. The dependency on the electrical current through the lamp is obtained between the currents 100 and 400 mA. The electron density and the electron temperature are measured at the tube axis. Table 4.8.3 tabulates the experimental conditions.

<table>
<thead>
<tr>
<th>Series</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cold-spot temp. (°C)</td>
<td>18</td>
<td>18</td>
<td>24</td>
<td>30</td>
<td>40</td>
</tr>
<tr>
<td>Mercury pressure (Pa)</td>
<td>0.14</td>
<td>0.14</td>
<td>0.24</td>
<td>0.39</td>
<td>0.85</td>
</tr>
<tr>
<td>Current (mA)</td>
<td>100, 200, 400</td>
<td>100 - 400</td>
<td>100 - 400</td>
<td>100 - 400</td>
<td>100 - 400</td>
</tr>
<tr>
<td>Measured quantities</td>
<td>$n_e(r), T_e(r)$</td>
<td>$n_e(0), T_e(0)$</td>
<td>$n_e(0), T_e(0)$</td>
<td>$n_e(0), T_e(0)$</td>
<td>$n_e(0), T_e(0)$</td>
</tr>
</tbody>
</table>

The ICCD camera had to be covered partly, suppressing the strong emission lines of neon. We used the spectral region in between the 588.2 and the 594.5 nm lines of neon. Only the wings of these lines could be detected. Assuming a Maxwellian energy distribution function, the distribution function of electrons with energies ranging from 0.5 to 5 eV could...
be measured. Although we suppressed the strongest lines in this way, there are ten weak neon lines remaining, overlapping the Thomson scattering spectrum. The lower levels of the transitions corresponding to these weak lines are in the $2p$ group. The weak lines resulted in an unexpected problem. The broadband ASE signal, which is suppressed by means of the ASE filter can be absorbed by neon atoms in the lower state of the weak lines. This absorption is succeeded by emission. This additional emission is not present in the spectrum of the plasma without the laser. So when we subtract the plasma emission from the combined spectrum of the plasma and the laser, we obtain the Thomson scattering spectrum and some additional emission from the weak neon lines, which for convenience we will call laser induced fluorescence (LIF). Normally, this additional emission can be detected in the spectrum by identifying the emission at a certain wavelength as a certain spectral line. This emission region can then be discarded in performing the fit of the Thomson scattering spectrum. In our case however, the spectral profile of the additional emission is similar to the spectral profile of the Thomson scattering signal, except for the 588.2 and the 594.5 nm lines, as can be seen in figure 4.8.7.

![Figure 4.8.7: The plasma emission for the cold-spot temperature of 18°C, and a current of 400 mA.](image)

This makes it very hard to discriminate between the Thomson scattering photons and the LIF photons. Based on the ASE level as reported in subsection 3.8.3 and the density of the $2p$ states as given in section 4.6, we can conclude that the LIF signal is of the same order of magnitude as the Thomson scattering signal for the measurements in series A and B. For series C, the LIF signal is much lower, and for series D and E, the LIF signal is negligible. We used the following procedure to approximate the correction for the LIF photons. The ratio of the LIF intensity to the intensity of the plasma emission for one spectral line is given by the following expression:

$$\frac{\Psi_{LIF}}{\Psi_{plasma}} = \frac{g_k}{g_i} \cdot \frac{n_i}{n_k} \left[ 1 - \exp \left( \frac{\tau_{iccd}}{\tau_k} \right) \right] a,$$

where $g_k$ and $g_i$ are the statistical weights of the upper and the lower state, $n_k$ and $n_i$ are the densities of the upper and the lower state, $\tau_{iccd}$ is the gate width of the camera, $\tau_k$ is the lifetime of the upper state, and $a$ is a constant depending on the geometry, the wavelength of the transition, and the ASE level of the laser.

The combined LIF and Thomson spectrum is now corrected by subtracting a fraction $\xi$ of the plasma emission. The fraction $\xi$ is chosen in such a way that the LIF signal of the 594.5 nm line vanishes. It can be shown using expression 4.8.1 that the ratio of the LIF
intensity to the intensity of the plasma emission is the highest for this line. So this correction is too high for the weak neon lines. However for the fit, we only use the wavelength range where there is only influence of two of the weak neon lines at approximately the same wavelength (591.89 and 591.90 nm) and where the correction is less than 30% of the resulting Thomson scattering profile. This means that the measured electron density will be at maximum 30% too low. On the other hand, the spectral profile of the plasma emission in this region has a similar form as a Thomson scattering profile with the temperature of approximately 1.5 eV. Therefore, the influence of the LIF correction on the measured electron temperature is low, since the electron temperature of the plasma is also approximately 1.5 eV. This is also apparent in the correction for the LIF signal, which is approximately a constant fraction of the Thomson scattering spectrum over the region of interest for Thomson scattering.

An example of a measured Thomson scattering spectrum is shown in figure 4.8.8, along with a Gaussian fit of the data, representing the Maxwellian velocity distribution function.

![Thomson Scattering Spectrum](image)

Fig. 4.8.8: Left-hand side: Example of a Thomson scattering spectrum in series E, with the fitted profile. Right-hand side: Example of a Thomson scattering spectrum in series B and one in series E, with the fitted profiles on a logarithmic scale as a function of the (Maxwellian) electron energy.

We fitted the measurements in series A, B, and C in the wavelength regions from 591.75 to 593.0 nm. This region corresponds to the energy range from 1.7 to 4.1 eV, assuming a Maxwellian distribution function. The measurements in series D and E are fitted in the wavelength regions from 590.75 to 593.0 nm. This region corresponds to the energy range from 0.5 to 4.1 eV, assuming a Maxwellian distribution function. Note that in the right-hand side plot of figure 4.8.8 the fitting curves are shown only in the energy range where the data is fitted.

A disadvantage of the fact that we only measure one side of the Thomson scattering spectrum is that the fitting procedure is less sensitive to the electron temperature. This is mainly caused by the fact that the width of the slit has been chosen larger than the image of the laser beam in the slit, as discussed in subsection 3.8.8. This can lead to a slight error in the wavelength axis. This error is in the order of 0.07 nm. However, we compared the measurements with the partly covered camera to measurements with the uncovered camera under the same discharge conditions. This was only possible in the situation where the neon emission was low enough. Measuring the whole spectrum resulted in a difference of 4% in
the electron density and 3% in the electron temperature. These differences are not significant, considering the experimental noise.

Figure 4.8.9 shows the results of the measurements series A, the electron density and the electron temperature as a function of the radial position \( r \). In figure 4.8.9, the markers represent the measurements, and the drawn lines are the fits of the measurements, using the following expression:

\[
    n_e(r) = n_{e0} \left[ 1 - \left( \frac{r}{R} \right)^m \right],
\]

where \( n_{e0} \), and \( m \) are fitting parameters. It was not possible to measure the Thomson scattering profile closer to the tube wall, since the electron density was too low to have a reasonable amount of scattered photons. Figure 4.8.9 also shows the electron temperature as a function of the radial position \( r \). Considering the noise on the measurements, we can conclude that the electron temperature does not vary over the radius.

![Graph](image1.png)

**Fig. 4.8.9:** The electron density (left), and the electron temperature (right) as a function of the radial position (series A).

Figure 4.8.10 shows the results of the measurement series B, C, D, and E, the electron density and the electron temperature on the tube axis as a function of the electrical current. It is clear that the electron density is a linear function of the current. This is in a first order approximation an effect of the increased power density in the discharge.

At the cold-spot temperature of 18°C, the electron temperature does not significantly decrease or increase with the current. At 24°C, the electron temperature first decreases, however, it increases at currents higher than 200 mA. At the other cold-spot temperatures, the electron temperature decreases with the current.

At the cold-spot temperature of 18°C, neon excitation always occurs. For the temperature 24°C, neon excitation is present at currents higher than 200 mA. This is clear in figure 4.8.11, where the emission of the weak neon lines is plotted as a function of the discharge current. Both regions where the emission of the weak neon lines is not zero correspond with a non-decreasing electron temperature in figure 4.8.10. For the higher cold-spot temperatures, neon excitation is absent, and the electron temperature decreases up to the current of 400 mA.
The trends in the electron temperature can be understood by analysis of the mass balance for electrons:

\[
 n_e \cdot n_{He} \cdot k_{He}(T_e) + n_e \cdot n_{Ne} \cdot k_{Ne}(T_e) = -\nabla[D_a(T_e) \cdot \nabla n_e],
\]

(4.8.3)

where \( n_{He} \) is the mercury density, \( n_{Ne} \) is the neon density, \( k_{He} \) is the ionisation reaction coefficient for mercury, \( k_{Ne} \) is the ionisation reaction coefficient for neon, and \( D_a \) is the ambipolar diffusion coefficient. Since the reaction coefficients are exponential functions of the electron temperature and the ambipolar diffusion coefficient is a linear function of this electron temperature, we can neglect the dependency of the right-hand side on the electron temperature. Furthermore, the ambipolar diffusion coefficient is mainly determined by the noble-gas pressure in the tube. As a first order approximation, we can assume the ambipolar diffusion coefficient to be independent of the mercury pressure. Regarding the reaction coefficients we note that at higher electron densities, the electron temperature decreases due to the increase of multistep ionisation contrast to direct ionisation.

In general, the electron temperature is such that the ionisation rate of mercury is much higher than the ionisation rate of neon. So a change in the mercury density will have a large
influence on the electron temperature, as can be concluded from the first term on the left-hand side of expression 4.8.3. When the mercury density is decreased, the electron temperature will increase since the decrease in the chance of an ionisation-collision has to be counterbalanced by increasing the chance of ionisation during a collision, since the loss of charged particles is approximately constant. So the electron temperature decreases with increasing mercury pressure. This effect is clear in figure 4.8.3, where on the average, the electron temperature decreases with increasing mercury pressure.

A change in the current will be followed by a change in the electron density. This change in the electron density influences the electron temperature, by means of enhancing multistep or direct ionisation. The change in the electron density also causes a change in the ambipolar flux to the wall, and subsequently a change in the mercury density, since the mercury density gradient is coupled to the gradient in the mercury ion density. The influence of this change in the mercury density is only significant in the situation with a low mercury pressure in the tube, i.e. at 18 and 24°C. There, the ionisation of neon can be significant, since the mercury ionisation decreases with the mercury density. However, neon requires a higher electron temperature, since the excited states possess a much larger energy than the corresponding states of mercury. The decrease of the electron temperature at low currents can be regarded as the increasing influence of multistep ionisation. The behaviour of the electron temperature at currents higher than 200 mA for the two lowest temperatures can be attributed to the increasing influence of neon ionisation contrast to mercury ionisation. This influence is absent at the two highest temperatures, since the mercury density and the mercury ionisation rate stay high enough.

In the past, several authors report the necessity of taking into account the depletion of the high energetic tail of the electron energy distribution function in mercury-noble-gas discharges, see for instance reference 28. The loss of electron energy in inelastic collisions of electrons with mercury atoms in the ground state is not compensated by the energy gain in the reverse process. This is especially important for the $^3P_{0,1,2}$ triplet, which is mainly populated by collisions between ground state mercury and electrons and is mainly depopulated by resonant emission through the resonant state $^3P_1$. The depletion of the electrons with energies higher than 4.6 eV is more pronounced at low electron densities, since there is less coulomb relaxation. We note that the electron temperature we measured is the temperature determined by the distribution function of electrons with energies lower than 4.1 eV, i.e. in the energy range in which we fit the data. In the investigation of the electron energy distribution function at the onset of neon excitation, the tail of the electron energy distribution function plays an important role. The tail electrons excite and ionise neon atoms. In the neon mercury positive column it is not possible to measure the tail electron distribution function using Thomson scattering due to the high level of plasma emission. However, qualitative information on the amount of high-energetic electrons has already been obtained from the emission measurements. Furthermore, in the next chapter qualitative information on the energetic electrons can be obtained from the neon ion density.

The outer wall temperature of the discharge tube at the position where the Thomson scattering spectra are obtained is plotted in figure 4.8.12, for the series A, B, C, D, and E. Note that series A and B are measured without increasing the ambient temperature, these series are taken at an ambient temperature of 27°C. The other series are taken at an ambient temperature of 50 to 56°C.
Fig. 4.8.12: The outer wall temperature of the tube for series A, B, and C-E.

The accuracy in the measurements is limited by systematic and a statistical errors. We will start with the discussion of the error in the electron density followed by an analysis of the error in the electron temperature. The discussion is an extension of the discussion given in subsection 4.8.1, for the Ar-Hg discharge.

**Electron density.** Apart from the discussion of the errors in section 4.8.1 (the Ar-Hg case), there are a few additional comments to make for the measurements in the Ne-Hg discharge. At first, the influence of covering part of the ICCD camera is also very small, as can be concluded from the comparison between the measurements with and without covering the camera. For the electron density measurements at the two lowest mercury pressures, an additional systematic error is introduced by the correction for the LIF signal. For the 18°C measurements, this additional error is at maximum 30%, for the 24°C this additional error is at maximum 10%. The additional statistical error for these measurements is caused by the fact that the region where the data could be fitted was smaller, resulting in a less accurate fit. This additional statistical error is approximately 10%.

**Electron temperature.** Apart from the discussion of the errors in section 4.8.1, there are a few additional comments to make for the measurements in the Ne-Hg discharge. At first, the influence of covering part of the ICCD camera is also very small, as can be concluded from the comparison between the measurements with and without covering the camera. For the measurements at the two lowest mercury pressures, there is an additional systematic error introduced by the correction for the LIF signal. For the 18°C measurements, this additional error is at maximum 10%, for the 24°C, this additional error is at maximum 5%. For these measurements there is an additional statistical error due to the fact that the wavelength region where the measurements can be fitted is smaller. A smaller region is used in order to limit the influence of the LIF correction. This additional error is approximately 10%.

The Thomson scattering experiments in the Ne-Hg discharge give us valuable information on the density and the temperature of the bulk electrons. The trends we observed are similar to the trends in the Ar-Hg discharge, except for the electron temperature as a function of the discharge current. In the next chapter we will combine the results of the Thomson measurements with the results of the preceding sections to obtain a more complete view of the Ne-Hg discharge and the radial cataphoresis process.
4.8.3 The electron energy distribution function in an Ar-Hg discharge

The measurements described in the preceding two sections show that up to velocities corresponding to the electron energy of 3 eV, the measured Thomson scattering spectra can be described very well using a Maxwellian velocity distribution function. However, since the signal to noise level is not very high, we cannot make any hard statements concerning the electron energy distribution function, especially for higher electron energies. In this section we describe the measurements we performed in order obtain the electron energy distribution function. We especially paid attention to removing the ambiguities in the measured spectra, as described in subsection 3.8.8. The measurements are performed in an Ar/Hg discharge with a mercury pressure of 300 Pa, and a mercury cold-spot temperature of 40°C.

Figure 4.8.13 shows the result of the Thomson scattering measurements for a discharge current of 400 mA. The markers represent the measurements the line represents a Maxwellian fit. The interference filter is blocking radiation with wavelengths below 587 nm and above 596 nm, as can be seen in figure 4.8.13.

Figure 4.8.13 also shows the Thomson spectrum $f_v$ on a logarithmic scale, as a function of the square of the difference between the scattered wavelength and the laser wavelength. It is clear that the Thomson spectrum follows a Maxwellian trend up to at least 2 nm from the laser wavelength. Further away from the laser wavelength, there is a clear deviation from the Maxwellian line.

In the left-hand side plot of figure 4.8.13, there is a strange ‘bump’ near 592 nm. We corrected the spectrum for the transmission of the optical components, for the stray light, for the plasma emission and for the equivalent background illumination (EBI) of the intensifier. Since we use a photon counting method, the influence of the dark-current of the CCD chip and the non-uniformity of the CCD chip is eliminated. There are no spectral lines of argon or mercury in the region around 592 nm. Apart from some experimental artefacts like changing background radiation in the laboratory, the only explanation for the bump is a non-uniformity in the photocathode response. However, we never found this non-uniformity when we measured the spectrum of a Tungsten filament lamp.

Using the expressions 3.8.23 and 3.8.25 we obtain the electron energy distribution function $f_0(E)$. Figure 4.8.14 shows this electron energy distribution function.
In this plot, less data points are shown than in figure 4.8.13. This is because we added adjacent channels in order to decrease the noise on the data. Below 3 eV two adjacent channels are added, above 3 eV four adjacent channels are added. In figure 4.8.14 the noise level is too high to conclude whether or not deviations from a Maxwellian energy distribution function are significant. Increasing the measurement time would decrease this noise level. However, it took us 16 hours to record the Thomson scattering spectrum as shown in figure 4.8.13. Therefore, a better signal to noise ratio can only be achieved by measuring the Thomson scattering spectrum for unreasonable long integration times.

4.8.4 Modulation of the electron density and temperature in a Ne-Hg discharge.

The previously described Thomson scattering measurements are all averaged over the phase of the electric field. This subsection describes the phase-resolved Thomson scattering measurements as performed in a neon-mercury discharge. The neon pressure was 1000 Pa, the discharge current was 400 mA, and the cold-spot temperature was 40°C. Figure 4.8.15 shows the electron density and temperature as a function of time, along with the absolute value of the discharge current, and the emission of the 585.925 nm line of mercury. It is clear that the modulation of the electron temperature is significant, while on the other hand, the electron density can be regarded as constant in time. The emission of the 585.925 nm line of mercury is a superlinear function of the electron temperature. The modulation of the electron temperature and the emission is shifted in phase with respect to the discharge current, this is also found by Drop et al.\textsuperscript{98}

We note that the differences in temperature within one cycle of the electric field are very big, compared to the differences we found in subsection 4.8.2. The modulation of the electron temperature will have a profound impact on the emission spectrum of the lamp. However, the average electron temperature is a good measure for the comparison of electron properties and the discharge emission. Note that the average temperature obtained from figure 4.8.15 is somewhat smaller than the value given in figure 4.8.10. However, this difference is not significant considering the noise of the measurements. The average electron density obtained from figure 4.8.15 is also somewhat smaller than the value given in figure 4.8.10.
The influence of the sinusoidal current waveform on the electron density can be estimated using the characteristic frequency for ambipolar diffusion $v_a$. This frequency can be calculated using the ion mobility $\mu_+$, the characteristic length for ambipolar diffusion $\Lambda_a$ and the electron temperature, using

$$v_a = \frac{\mu_+ T_e}{\Lambda_a} = 2.4 \cdot 10^3 \cdot T_e,$$  \hspace{1cm} (4.8.4)

expressed in Hertz. The electron temperature is of the order of 1.3 eV, so the ambipolar diffusion frequency is of the order of 3 kHz. We used the value for the mobility of mercury ions in neon as measured by Chanin.\textsuperscript{100} We can conclude that the driving frequency is much higher than the frequency for ambipolar loss of charged particles. Therefore, the modulation of the electron density will be negligible. This is also the case for the measurements as plotted in figure 4.8.15.

The influence of the sinusoidal current waveform on the electron temperature can be estimated using the characteristic frequency for energy loss of electrons. This frequency $v_u$ can be written as

$$v_u = \delta \cdot v_m,$$  \hspace{1cm} (4.8.5)

where $v_m$ is the effective collision frequency for momentum transfer, and $\delta$ is the fraction of the electrons energy lost in one collision. For the bulk of the electrons, we can assume that they are only involved in elastic collisions. Therefore, we can use the following expression for $\delta$:

$$\delta = \frac{2m_e}{M},$$  \hspace{1cm} (4.8.6)

where $m_e$ is the electron mass and $M$ is the mass of the dominant heavy particle, i.e. the neon atom. Then the expression for the frequency for energy loss is:

$$v_u = \frac{2e}{M\mu_e}.$$  \hspace{1cm} (4.8.7)

The electron mobility $\mu_e$ is of the order of 50 m\textsuperscript{2}/Vs, so the characteristic frequency for energy loss is of the order of 200 kHz. This frequency is much higher than the driving frequency. Therefore, there will be a significant modulation of the electron energy. The corresponding modulation of the electron temperature is very clear in figure 4.8.15.
4.8.5 Discussion Thomson scattering experiments

We demonstrated that it is possible to measure a Thomson scattering spectrum under severe stray light and plasma emission conditions. Despite this, some improvements of the set-up can be made. Especially the reduction of stray light and plasma emission can be improved.

For the stray light reduction, keeping the optics in the ASE filter free from dust is an important issue. Scattering of the laser beam at the surfaces of the optical components and in the pinholes is the main reason that the stray light level is higher than expected. Furthermore, it is possible to improve the rejection of the ASE filter even further by filtering the laser beam twice. In order to do this, we need an extra, third spatial filter in the set-up, as proposed in subsection 3.8.3. An intermediate solution is to insert one dispersion prism as a predisperser, and to use the first spatial filter as a bandpass filter. In this way we obtain a two-stage ASE filter. The rejection of this filter will be 5 orders of magnitude in the spectral bandpass region of the first filter, and more than five orders of magnitude in the rejection region of this first filter.

The influence of plasma emission can be reduced by improving the synchronisation of the camera to the laser. Especially optical triggering of the ICCD camera would lead to a big improvement. This is not possible at the moment, since the intrinsic delay generated in the gating electronics, the cables and the delay generator is approximately 150 ns. We either need an optical delay line of 150 ns or a jitter-free pre-trigger pulse. The optical gate can then be reduced to approximately 50 ns. This would result in a much better signal to noise ratio for the Thomson spectrum, since at the moment the noise is mainly resulting from the plasma emission. Another improvement would be the use of a better spectrograph. At the moment, the apparatus profile of the spectrograph is rather wide. Therefore, the influence of spectral lines spreads out over the Thomson spectrum. Another improvement could be the use of another laser wavelength. The atomic notch filter we built can also be used for metal vapours other than sodium. Using other metal vapours, the wavelength for Thomson scattering can be changed. This can be interesting when the plasma emission in the 589 nm region is too large for Thomson scattering. Since we used the same materials as Neuman et al., the cell we built is applicable for containing strontium (460.7 nm) and rubidium (780.0 and 794.8 nm). By evaluating the electrochemical activity of the other alkali metal and alkaline earth metals, we believe that the cell is robust enough to contain metal vapours like barium (553.5 nm), potassium (766.5 and 769.9 nm), and calcium (422.7 nm). Due to their extreme aggressiveness, caesium (852.1 and 894.4 nm), and lithium (670.78 and 670.79 nm) will probably react -at least- with the glass to metal seal. Furthermore, we should note that the vapour pressure of Lithium and Calcium is too low to have sufficient absorption at a reasonable temperature. A disadvantage of changing the laser wavelength is the fact that we have to change from the relative stable Rhodamine 6G dye to another -less stable- dye. We had to change the Rhodamine 6G dye at least every hour. The dye replacement frequency could be unrealistically high when using other dyes. An improvement would be the use of a larger dye circulator. When the excimer laser power is at maximum, the dye degrades in approximately 10 minutes. Using a larger circulator for the oscillator stage will increase this degrading-time.

Regarding the detection limit, it is possible to detect Thomson scattering at an electron density of $10^{17}$ m$^{-3}$. We needed half an hour at approximately 1 Watt of laser power to measure this Thomson spectrum. By optimising the set-up, we believe that we can measure down to $10^{16}$ m$^{-3}$. This optimisation includes the dye laser and the ASE filter alignment,
lowering the gate-width of the camera, and lowering the wavelength resolution by, for instance, resolving the Thomson spectrum in 20 channels instead of 400 channels.

In the positive column, the spatial resolution in the direction of the laser beam is not an issue, since the column can be regarded as homogeneous. However, when necessary, the spatial resolution can be increased by changing the ICCD read-out mode from full vertical binning to imaging. Then we can choose the spatial and spectral resolution we want by either integrating vertically or horizontally on the ICCD chip. The use of the photon counting method can further improve the signal to noise ratio. Especially the implementation of an image recognising algorithm will have a positive influence on the measurements. A disadvantage is the long time needed for the readout of the CCD chip compared to the full vertical binning method. This longer readout time increases the measurement time.
5 Synthesis

5.1 Introduction
In the preceding chapter we discussed the results of the experiments we performed. In this chapter these results will be combined and their mutual consistency will be verified. We will briefly discuss the hybrid discharge. The main part of this chapter is devoted to the depleted discharge. For the depleted discharge, the phenomenological description of radial cataphoresis as given in subsection 2.4.2 is verified. Furthermore, a comparison is made between the two discharges. The general aim of this chapter is to obtain a more complete view of the two discharges.

5.2 The hybrid discharge
The most interesting phenomenon in the measurements we performed on the hybrid discharge is the decrease in the intensity of the spectral lines of mercury, while the intensity of the neon lines increases. This effect is shown in figure 4.2.2. An interesting comparison can be made between this figure and figure 4.3.1. The real part of the discharge impedance starts to increase at the same power where the mercury intensity starts decreasing.

The emission pattern suggests that we have a $\gamma$-type ccrf discharge, using Levitsky’s terminology. The ionic current to the dielectric material covering the electrodes results in secondary emission of electrons. As in glow discharges, at the cathode a cathode layer is formed. The coloured layer near the electrodes is similar to the layer at the cathode of a direct current glow discharge. In the sheath region, the energy of the electrons is much higher than in the glow region. This is due to the fact that the electric field is much lower in the glow region than in the sheath region. The low electron energy in the glow explains the fact that we only observed the production of neon radiation in the sheath region.

We will now discuss the effect of increasing the discharge power. An increase in the input power leads to an increase in the discharge voltage, see for instance figure 3.3.6. The voltage across the glow region is negligible compared to the sheath voltages, as can be concluded from the discharge impedance in figure 4.3.1. Therefore, we can assume that the sheath voltage is approximately proportional to the discharge voltage. Because of this, an increase of the discharge power results in an increase of the sheath voltage. The increase of the sheath voltage results in an increase of the energy of the secondary electrons in the sheath, favouring the excitation of neon atoms with respect to mercury atoms. The power deposited in the sheath region also increases with increasing sheath voltage, resulting in the increase of the intensity of the neon lines, and the increase of the real part of the discharge impedance. The power deposited in the glow region also increases with the discharge power. However, due to the increasing glow conductance this increase is smaller than for the sheath power. A possible explanation for the decreasing intensity of the mercury lines with increasing power is depletion of mercury in the sheath region due to the ionic current to the electrodes. The plasma potential increases with the sheath voltage and thus with the discharge power. The ionic current to the electrodes and the resulting mercury depletion will therefore increase with the discharge power. As a result, the intensity of the mercury lines decreases with decreasing mercury density. This effect can dominate the increasing glow intensity.

As a last comment on the hybrid discharge, we will discuss the efficiency for the production of radiation. In almost all ccrf discharges, the bulk of the input power ultimately is absorbed by ions accelerating in the sheath and subsequently deposited at the surface. In ccrf discharges only a small fraction of the energy is used to heat electrons. Only that energy is...
available for the generation of radiation. Therefore, the efficiency is expected to be low, which is clearly demonstrated in the sections 4.2 and 4.3.

5.3 The depleted discharge
This section discusses the results of the experimental study of the depleted discharge. We will start with comparing and cross-linking of the results. Subsequently, we will discuss the results in relation to the phenomenological description of the radial cataphoresis process.

5.3.1 Comparison and cross-linking of the results.
In this subsection we will use simplified analytical models to compare the results of different measurements and different diagnostic techniques.

A. Mercury density versus the electron density
The first comparison we will make is the comparison between the mercury density and the electron density. The mercury density is directly related to the mercury ion density:

\[ n_+ (r) = \frac{k}{e} \frac{\mu_+}{\mu_e} \frac{T_0}{T_e} (n_0 (R) - n_0 (r)), \] (5.3.1)

This expression was derived in section 2.4. For the derivation, we assumed the particle temperatures to be independent of position. This assumption is valid for the electron temperature. However, the heavy particle temperature is not uniform, it is approximately shaped like a parabola. Based on temperature measurements of Kenty et al., it can be estimated that the temperature on the tube axis is 10, 20, and 40% higher than the wall temperature for the currents 100, 200, and 400 mA, respectively. We neglect this temperature difference and assume the gas temperature to be equal to the wall temperature. The mercury ion density profiles resulting from the measurements shown in figure 4.5.1, using expression 5.3.1, are shown in figure 5.3.1. We used the value for the electron temperature obtained from the Thomson scattering measurements. For the mercury density at the wall, we used the wall temperature, and the mercury pressure as determined by the cold-spot temperature.

Fig. 5.3.1: The electron density (left, full markers), the mercury ion density (left, open markers), and the resulting neon ion density (right). The error bars in the plot on the right represent an estimate of the error in the neon ion density.
It is clear that for the two largest currents, the neon ion density is not negligible compared to the mercury ion density. The mercury ion density is approximately equal to the electron density for the current of 100 mA. This conclusion can also be drawn from the emission measurements. These emission measurements show that there is no neon radiation emitted by the lamp in this case. Therefore, we can assume the neon excited state density and the neon ion density to be negligible under these circumstances.

Since we have two species of ions in the discharge, we have to redefine the ambipolar diffusion coefficient. By equating the flux of electrons to the total flux of ions, taking into account both diffusion and drift, we obtain the following expression:

\[
D_a = \left\{ \mu_{Hg^+} - \left( \mu_{Hg^+} - \mu_{Ne^+} \right) \frac{n_{Ne^+}}{n_{Ne^+} + n_{Hg^+}} \right\} \cdot T_e ,
\]

where \( \mu_{Hg^+} \) is the mercury ion mobility, \( \mu_{Ne^+} \) is the neon ion mobility, \( n_{Hg^+} \) is the mercury ion density, \( n_{Ne^+} \) is the neon ion density, and \( T_e \) is the electron temperature expressed in eV. It is clear that the ambipolar diffusion coefficient is dependent on the density of the ions. The mobility of neon ions in 1000 Pa neon is 0.04 m²/Vs,\textsuperscript{102} the corresponding mobility of mercury ions is 0.06 m²/Vs.\textsuperscript{100} The neon ion mobility is lower than the mercury ion mobility. Therefore, an increasing neon ion density will result in slower ambipolar diffusion and thus, better confinement of the electrons.

Since it was not possible to measure the mercury density profile at a higher mercury pressure, we have to look for other ways to obtain information about the mercury ion density. One way is again using the mass balance for mercury, along with the electron density and electron temperature measurements. We know that the mercury ion density is at maximum equal to the electron density. A more elaborate estimate of the mercury ion density can be made using the mass balance of mercury, and the fact that the mercury ground state density is always positive. This leads to a maximum value of the mercury ion density on the axis of the discharge tube. The corresponding minimum value of the neon ion density is presented in figure 5.3.2 as a function of the discharge current.

![Fig. 5.3.2: The minimum neon ion density as a function of the discharge current.](image)

We note that in this subsection, we assumed a constant heavy particle temperature over the tube radius. In practice, the temperature at the tube axis will be significantly higher than the wall temperature. The higher gas temperature will result in a lower mercury ion density on the axis. The correction would amount to a maximum of 10%.
B. The electric field and the electron density and temperature

We measured the electric field in the positive column by using the fact that the column is homogeneous along its length. The electric field can also be calculated from the electron density, the electron temperature, and the corresponding mobility of the electrons. We use the following expression

\[ I = 2\pi \int_0^R j(r)rdr = 2\pi e\mu_e(T_e)E \cdot \int_0^R n_e(r)rdr, \]

which can be rewritten in

\[ E = \frac{I}{2\pi e\mu_e(T_e)\int_0^R n_e(r)rdr}. \]

For the electron mobility, we used the cross-sections for momentum transfer of neon, and mercury, along with Blanc’s law. Figure 5.3.3 shows the electric field as a function of the discharge current.

![Electric Field vs Discharge Current](image)

Fig. 5.3.3: The electric field calculated from the Thomson scattering measurements (square) and the electric field obtained from the positive column voltage measurements (circle).

It is clear that the order of magnitude is the same for both electric field measurements. The trend as a function of the discharge current is also similar. The difference in the absolute values could be due to differences in a few experimental parameters: the discharge geometry, the driving frequency and the wall temperature. We also note that Blanc’s law for the mobility in mixtures is an approximation, see for instance Verweij. Furthermore, we only considered the influence of neon and mercury ground state atoms on the electron mobility.

We can also compare the trends in the electric field and the electron temperature directly, using expression 5.3.3. In this expression, the current and electron density dependency cancel out since the electron density is proportional to the discharge current, as shown in section 4.8. The electron mobility is a decreasing function of the electron temperature. Therefore, the trends in the electric field should be similar to the trends in the electron temperature. When we compare the measured electric field in figure 4.4.5 with the measured electron temperature in figure 4.8.10, we can conclude that they indeed follow the same trend, approximately.

C. The electric field versus the emission spectrum: critical current.

In caesium- and sodium-noble-gas discharges, a large increase of the electric field in the positive column is observed at a certain critical current. At this critical current, the discharge also starts to produce noble-gas radiation, i.e. the emission spectrum contains a significant
contribution of spectral lines of the noble gas. Figure 4.8.10 shows that at a constant discharge current, the electron temperature is higher in a mercury-noble-gas discharge that produces noble-gas radiation (at 18°C) than in a mercury-noble-gas discharge, dominated by mercury (at 40°C). The higher electron temperature results in a lower mobility, and thus higher energy losses. Because of this, the electric field will increase. The increase of the electric field will be present at approximately the conditions where the discharge starts producing noble-gas radiation.

We measured both the emission spectrum and the electric field for the neon-mercury positive column. We can derive a critical current from the emission spectrum and also from the electric field. The critical current inferred from the emission spectrum is already discussed in the preceding chapter, section 4.4. In this subsection, we will compare this critical current for emission with the one inferred from the electric field. We define the lowest current where \( \frac{dE}{dI} \) is zero in the measured \( E(I) \) curve (for instance in figure 4.4.5) as the critical current for the electric field. Figure 5.3.4 shows the values of the two different critical currents. It is clear that the trends in the critical current are the same. The critical currents for the electric field are approximately 25% lower than the ones obtained from the emission spectrum. However, the 1500 Pa measurements deviate significantly more than the rest.

![Graph](image)

**Fig. 5.3.4:** The critical current inferred from the emission measurements (full markers, solid line), and the electric field (open markers, dashed line). In the plot on the left-hand side, the neon pressure is either 300 (square), 500 (circle), 1000 (up-triangle), or 1500 (down-triangle) Pa. In the plot on the right-hand side, the cold-spot temperature is either 18 (square), 20 (circle), 22 (up-triangle), 24 (down-triangle), 27 (diamond), or 30°C (star).

Obviously, the differences in the results of the two ways of deriving a critical current can be attributed to the efficiency of the discharge for producing neon radiation. This efficiency is a function of the electron density, the electron temperature, the mercury density, the amount of self-absorption of the neon lines, the metastable densities, and so on.

At 1500 Pa neon, the critical current for the electric field is much lower than the critical current for the noble-gas emission. Apparently, the discharge conditions do not favour the production of neon radiation. A reasonable assumption is the fact that the electron temperature is low compared to the discharges with a lower neon pressure. It is also reasonable to assume that the self-absorption of the neon lines is more pronounced at the higher neon pressures. The densities of the excited states of neon are higher at higher neon...
pressures. This self-absorption impedes the neon emission in leaving the discharge tube. Both effects cause an increase of the critical current for the noble-gas emission.

Comparing the minimum neon ion density as shown in figure 5.3.3 with the critical current inferred from the electric field as shown in figure 5.3.4, we note that the current where the minimum neon ion density starts to become significant, approximately coincides with the critical current.

**D. The degree of depletion versus the emission spectrum.**

The mercury density is used as a tool to influence the emission spectrum of a mercury-noble-gas discharge. This subsection discusses the relation between the mercury density on the axis of the tube and the emission spectrum as emitted at the tube axis. The mercury density is measured for one specific mercury pressure. Figure 5.3.5 shows the local emission at the discharge axis as a function of the mercury density on the axis.

It is clear that the emission of the mercury lines increases with the increasing mercury density. However, this is not as straightforward as it seems, since the electron density at the conditions of the highest mercury density ($I=100$ mA) is approximately four times lower than at the conditions of the lowest mercury density ($I=400$ mA). As expected, the emission of the neon lines decreases with increasing mercury density.

A similar conclusion can be drawn from the time-resolved emission measurements. These measurements show that the intensity of the mercury lines decreases as a function of the time that the discharge is on, while the intensity of the neon lines decreases. The characteristic time for this increase and decrease is of the same order of magnitude as the characteristic diffusion time for mercury atoms in neon. This suggests that the removal of mercury is the cause for the production of neon radiation. As discussed above, this is also confirmed by the results of the spatially resolved emission measurements that show that the neon radiation is produced in the regions where the mercury density is low.

**E. The density profiles**

Comparing the density profiles can lead to a better understanding of the discharge. Figure 5.3.6 shows the density of the $6^1S_0$ (ground state), the $7^3S_1$, and the $6^3D_2$ states of mercury, the $2p_{10}$ state of neon, the electron density, and the electron temperature in the discharge with a neon pressure of 1000 Pa, and a cold-spot temperature of 18°C.
Fig. 5.3.6: The density profile of the $6^1S_0$ (square), the $7^2S_1$ (circle), and the $6^3D_2$ (up-triangle) states of mercury, along with the density profile of the $2p_{10}$ state of neon (down-triangle), the electron density profile (star), and the electron temperature profile (cross). The current for the plot on the left-hand side is 100 mA, the current for the plot on the right-hand side is 400 mA. In the 100 mA plot, the neon $2p_{10}$ state density is omitted since it is negligibly low. All the quantities are scaled to fit in the plot.

We will first consider the excited states of mercury. Both the excited states of mercury are coupled to the mercury ground state atoms and the electrons. As a first order approximation, we can either assume the $3^3S_1$ state to be populated by electronic excitation via the $3^3P$ states (assumption A), or we can assume the $3^3S_1$ state to be populated directly from the ground state by electronic excitation (assumption B). When we assume the net electronic depopulation of the $3^3S_1$ and the $3^3P$ states to be negligible compared to other depopulation processes like radiative decay, then assumptions A and B lead to the following two expressions for the density of the $3^3S_1$ state $n_2$:

$$n_2 = n_{Hg} \cdot n_e^2 \cdot c_1,$$

(5.3.4)

and

$$n_2 = n_{Hg} \cdot n_e \cdot c_2,$$

(5.3.5)

respectively. In these expressions, $c_1$ and $c_2$ are constants, depending on the electron temperature and other discharge parameters. Figure 5.3.7 shows the density of the $3^3S_1$ state, along with the calculations using expression 5.3.4 and 5.3.5. It appears that assumption B is better for the 100 mA case, while assumption A is better for the 400 mA case. This makes sense, since the $3^3P$ density increases with increasing current. A related argument is the fact that the electron density is much lower in the 100 mA case than in the 400 mA case. Therefore, at the highest current, multistep processes will be more important.

We will now concentrate on the neon excited state density. The electron temperature at 100 mA is 1.9 eV, at 400 mA it is 1.8 eV. Both values are determined using Thomson scattering. We note that the electron temperature we determine is the temperature of the bulk electrons with an energy in the 1.7 to 4.1 eV. For excitation and ionisation of atoms, this energy is not sufficient. Although the bulk temperature is almost the same for the two plots in figure 5.3.6, the neon $2p_{10}$ density is much higher in the 400 mA case. This suggests that the amount of electrons in the tail of the electron energy distribution function is significantly higher in the 400 mA case than in the 100 mA case. This is plausible, since the depletion of the energetic electrons is mainly caused by the improper balance of the electronic excitation and the radiative de-excitation of the $3^3P_1$ state of mercury. The electron-electron collisions, which counterbalance the depletion of the energetic electrons are much more frequent at
higher electron densities, and thus at higher currents. Moreover, the mercury density is much lower at the higher currents, which results in less inelastic collisions of electrons with mercury.

![Graph showing density profiles](image)

Fig. 5.3.7: The measured density of the $^3S_1$ state (solid), along with the calculation using assumption A (dash), and assumption B (dot). The left-hand plot shows the density for the current of 100 mA. The right-hand side shows the density for the current of 400 mA.

The spatial distribution of the energetic electrons could not be measured independent from the emission measurements. However, assuming that the high-energy electrons are lost in inelastic collisions with mercury, their density will be less flat than the bulk electron density. This is because of the fact that the mercury density profile is hollow, and the tail electrons will be lost in the regions where the mercury density is high. The excitation of neon and mercury will be lowered by the depletion of the energetic tail. This could also be a reason for the bell shaped neon excited state density profiles.

**F. Collisional-radiative model.**

The consistency of the density profiles can also be checked with a collisional-radiative model (CRM). We used the collisional-radiative model for mercury and neon, developed by the PLASIMO-team of the group Equilibrium and Transport in Plasmas at the Eindhoven University of Technology. It is described in detail by Van Dijk *et al.* The model uses the quasi steady state solution. We assumed the neon and the mercury part of the model to be decoupled, except for the influence of both atoms on the electron energy distribution function. The effective lifetime of the resonant states of neon is calculated using the Holstein escape factor. For the other states of neon and all the states of mercury, the escape factor is assumed to be one. This is an estimate, since there will be some re-absorption, especially for resonant radiation of mercury. The escape factor for neon is assumed to be equal to the analytical Holstein-expression at the tube axis and equal to 1/2 at the wall. In between, a third order polynomial function is used. No free parameters are used in the calculation of the density profiles. We used the measured electron density, (bulk) electron temperature, and mercury atom density. For the description of the depletion of the tail of the eedf, we assumed a two-temperature distribution. The tail temperature is calculated from the measured (bulk) electron density, (bulk) electron temperature, and atom densities, using an approach similar to Ligthart *et al.* The ion densities are calculated from the electron density, temperature and mercury atom density as described in part A of this subsection. The heavy particle
temperature is assumed to be parabolic shaped, with a maximum at the discharge tube axis. For the difference between the axis temperature and the wall temperature, we used the estimates as given in part A of this subsection.

Figure 5.3.8 shows the density of the $7^3S_1$, the $6^3D_2$, and the $8^1S_0$ states of mercury as a function of the radial position, along with the density of the $2p_2$ and the $2p_{10}$ states of neon. The agreement is reasonable; the shape of the density profiles is similar. However, the absolute densities differ. For neon the calculated density is approximately one order of magnitude higher than the measured density. This difference can be attributed to experimental errors and approximations in the CRM. Especially the uncertainty in the tail temperature will result in large errors in the density profiles of neon. Only the electrons with an energy higher than 16.5 eV can excite a neon atom. These electrons are situated very far in the tail of the energy distribution function. Therefore, the excitation of neon is very sensitive for errors in the tail temperature.

The results of the CRM calculations can be used to estimate the effect of Penning ionisation. At a discharge current of 400 mA, the rate for the production of the $1s_5$ metastable state is in the order of $10^{22}$ m$^{-3}$/s, as calculated by the CRM. The rate for Penning ionisation of mercury by neon $1s_5$ metastable atoms is in the order of $10^{18}$ m$^{-3}$/s, as calculated using the rate coefficient measured by Wren et al.$^{110}$ At a discharge current of 100 mA, the rate for
Penning ionisation is also negligible. Therefore, the Penning ionisation is not an important process for the quenching of neon metastables at our discharge conditions.

5.3.2 Discussion and synthesis experimental results

This subsection discusses the results of the separate experiments and the cross-linking of the results. In this discussion we will verify whether the phenomenological description given in subsection 2.4.2 is valid. We will start with the discussion of every step in this description, relating it to the experimental results.

Step 1 and 2: An increasing ionisation degree as a result of an increase in the discharge current or a decrease of the mercury pressure. The results of the Thomson scattering experiments in the Ar-Hg and the Ne-Hg discharge show that the electron density is proportional to the discharge current. Therefore, when the mercury density is not increasing, the ionisation degree for mercury increases with the discharge current. Furthermore, the electron density is increasing with decreasing mercury pressure, see figure 4.8.4. Therefore, lowering the mercury pressure also results in an increasing ionisation degree for mercury. This confirms step 2 of the phenomenological description.

Step 3: Significant mercury depletion. This step is confirmed by the ultraviolet absorption measurements. These measurements show higher depletion of mercury atoms at high discharge currents, see figure 4.5.1. The ‘depletion’ of mercury at lower mercury pressures is trivial. Lowering the mercury pressure results in a decrease of the mercury density, if the temperature is not increased.

Step 4: Less inelastic collisions of electrons with mercury. This step can only be confirmed indirectly. We assume that we can use the emission of the spectral lines of mercury as a probe for the amount of inelastic collisions of electrons with mercury atoms. Figure 4.4.4 shows an increase of the spatially integrated intensity of the mercury lines. This falsifies the assumption in step 4. However, we have to consider the amount of inelastic collisions per electron, and not the total amount of the inelastic collisions of all electrons. One way to do this is to scale the total emission to the electron density. Another way to show that the amount of inelastic collisions of an electron with mercury atoms is decreasing is to look at the local emission coefficient for the spectral lines of mercury, instead of the spatially integrated emission. The spatially resolved emission measurements show that on the discharge axis, the emission of the spectral lines of mercury decreases with increasing current and decreasing mercury pressure, see figure 5.3.5. Both effects confirm step 4 indirectly. It is also clear that the emission profiles become hollow when the mercury ground state density profile is hollow. This can be concluded from a comparison of the mercury ground state density in figure 4.5.1 with the emission profiles in figures 4.6.1 and the excited state density profiles in figure 4.6.3. Obviously, these densities are coupled because for the net production of excited mercury atoms, ground state mercury atoms are needed. The similarity in shape of the two density profiles is another confirmation of step 4, the decrease of the inelastic losses in collisions of electrons with mercury atoms.

Step 4 and 5: The increase of the electron temperature and the production of noble-gas radiation. The increase of the electron temperature with decreasing mercury pressure is confirmed by the Thomson scattering measurements in both the Ar-Hg, and the Ne-Hg discharges, see figure 4.8.4, and 4.8.10. However, the electron temperature is not always increasing with increasing discharge current in the mercury-depleted situation. As discussed in section 4.8, this is probably due to the higher probability for multistep processes when the electron density is high. Multistep processes result in a decrease of the electron temperature when the current is increased, see figure 4.8.3 for the Ar-Hg discharge. However, for the Ne-
Hg discharge at the two lowest mercury pressures, the electron temperature is not decreasing with increasing discharge current. The discharge current regions where this occurs correspond to the regions where neon radiation is produced. This can be concluded from the comparison of the electron temperature in figure 4.8.10 with the emission of the neon lines in figure 4.8.11.

In general, whenever noble-gas radiation is produced in either the Ar-Hg or the Ne-Hg discharge, the electron temperature is high. However, a high electron temperature is not enough for the production of noble-gas radiation. The electron temperature in the 18°C case in figure 4.8.10 is approximately constant, while the neon emission differs significantly, as can be seen in the emission profile in figure 4.6.1. The most probable reason for this is the depletion of high energetic electrons, as discussed in the preceding subsection. Moreover, it is also clear in figure 4.6.1 that the production of neon radiation occurs in the regions where the mercury density is low. This is the region where there is not much depletion of energetic electrons.

In conclusion, step 5 is not confirmed in all the experiments, while step 6, the production of noble-gas radiation, is confirmed. For the characterisation of the shape of the electron energy distribution function (eedf) we need more than one temperature. The presence of the high-energetic electrons is reflected in the production of noble-gas radiation. An important conclusion that can be drawn from the emission measurements is the fact that high-energetic electrons are present under the conditions where the discharge current is high and the mercury density is low. The radial cataphoresis process results in the presence of high-energetic electrons under these conditions.

Concerning the simplified model for the mercury atom density and the mercury ion density, we can draw some conclusions. Firstly, the simplified model gives good qualitative results; the trends in the critical current can be explained using this model. Furthermore, this model is useful for the estimation of the degree of depletion. The estimations are validated by the experiments. Finally, the model can be used for the calculation of the mercury ion density from the mercury ground state density. This calculation is validated by the Thomson scattering experiments, as can be concluded from the 100 mA curves in figure 5.3.1 and the related discussion.

In conclusion, the phenomenological description as given in subsection 2.4.2 is validated by the measurements, except for step 5, the increase of the electron temperature. This increase is not always observed. We observed that noble-gas radiation is only produced when the electron temperature is sufficiently high. However, the high electron temperature is not enough, since it is the amount of high energetic electrons that determines the intensity of the spectral lines of the noble gas. The number of high energetic electrons can be independent from the electron temperature.

### 5.4 Comparison performance hybrid and depleted discharge.

As reported in the preceding chapter, control of the emission spectrum of both discharge lamps is possible. The sheath region of the ccrf discharge and the mercury depleted positive column of a neon-mercury discharge are able to produce neon radiation. This is clear in the spectra in figure 4.2.1 and in figure 4.4.1, where neon lines are present. A comparison of both graphs shows an effect similar to Seeliger’s rule. The light produced in the sheath region of the ccrf discharge is orange while the light produced in the positive column of the depleted
discharge is red. This is mainly due to the intensity of the yellow 585.2 nm line, which is much higher in the ccrf discharge than in the depleted discharge.

Figure 4.2.4 shows the main difficulty of the hybrid discharge. The colour temperature shift is very small. When the forward rf power is equal to 24 Watt, the colour shift at the higher If powers is only 500 K. We note that the If discharge current is at maximum in the order of 100 mA, which is very low for a fluorescent lamp. This means that at realistic fluorescent lamp conditions, where the If current is much higher, the colour temperature shift will be even lower. Furthermore, the colour shift will be even lower at higher cold-spot temperatures than the 22°C we used.

The intensity of the neon lines is much higher in the depleted discharge than in the hybrid discharge. This is clear in figure 4.4.2, where even at conditions where a lot of mercury radiation is produced, i.e. at high currents and high mercury pressures, the colour temperature decreases significantly. A disadvantage of the depleted discharge is its sensitivity for changes in the mercury pressure, as can be seen in figure 4.4.2. This necessitates good control of the cold-spot temperature.

The efficiency for the production of noble-gas radiation is much better for the depleted discharge than for the hybrid discharge. Therefore, the depleted discharge offers the best perspective for commercial application. A complication of both the depleted discharge and the hybrid discharge is the necessity of the control of the mercury pressure.
6 General conclusions

The research goal was to obtain a better understanding of the processes involved in the production of noble-gas radiation in mercury-noble-gas discharges. We studied two options to control the noble-gas emission: the hybrid discharge and the depleted discharge lamp. A number of conclusions can be drawn from the experimental study of these discharges. These conclusions are lined up in this chapter, arranged into three categories: (a) the plasma control of the emission spectrum, (b) the application of the discharges in variable colour fluorescent lamps, and (c) the diagnostic study itself. The conclusions in category (a) are related to the first main question: which discharge processes and conditions are important for the production of noble-gas radiation in mercury-noble-gas discharges? The conclusions in category (b) are related to the second main question: how can we produce noble-gas radiation in an efficient way in these discharges? The conclusions in category (c) evolved from the development of the diagnostic techniques.

(a.1) Both the sheath region of a capacitively coupled radio frequency discharge and the radial cataphoresis process can be used to produce noble-gas radiation in a mercury-noble-gas discharge.

(a.2) The phenomenological description of both the radial cataphoresis process and the resulting addition of noble-gas emission to the discharge spectrum, as given in subsection 2.4.2, is validated by the results of the experimental study, with a partial exception for the increase of the electron temperature. The mercury density is the dominant parameter for the production of noble-gas-radiation in the depleted discharge. The effect of mercury on the production of noble-gas radiation is twofold. Firstly, the presence of mercury influences the electron temperature. Secondly, it results in the depletion of high-energetic electrons.

(b.1) The efficiency of the hybrid discharge for the production of noble-gas radiation is too low for the application in a variable colour fluorescent lamp.

(b.2) The depleted discharge can be used in a variable colour fluorescent lamp. The colour temperature of this lamp is controlled by the discharge current. Amplitude modulation of the discharge current enables the independent control of the colour temperature and the luminous flux. The maximum modulation frequency is determined by the characteristic time for the transport of mercury. For the application of the depleted discharge, special care has to be taken in order to control the mercury pressure.

(c.1) It has been demonstrated that Thomson scattering can be performed in high stray-light environments. The combination of a dye laser, an ASE filter, and an atomic notch filter has been used to reduce the influence of stray light in a Thomson scattering set up.

(c.2) The power reflection coefficient of a terminated coaxial line can be used to measure the stray impedances in the terminating radio frequency network.
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Appendices

Appendix A: Zero-dimensional, time-depended model for mercury transport.

In this appendix, we will derive a simplified time-dependent model for the mercury transport in the discharge. We will start with the mass balance for mercury atoms and mercury ions.

\[
\frac{\partial n_{Hg}}{\partial t} = -n_{Hg} n_{e} k_{ion} + D_{0} \nabla^{2} (n_{Hg}),
\]  
(A.1a)

and

\[
\frac{\partial n_{e}}{\partial t} = +n_{Hg} n_{e} k_{ion} + D_{a} \nabla^{2} (n_{e}).
\]  
(A.1b)

The diffusion coefficients are assumed to be independent of position. We assume that the mercury ion density is a constant fraction of the electron density. The spatial dependence of the densities will be neglected and Laplace operator will be approximated by:

\[
\nabla^{2} (n) \approx \frac{\delta(n)}{\Lambda^{2}},
\]  
(A.2)

where \(\delta(n)\) is the difference in \(n\) over the characteristic length \(\Lambda\). This characteristic length is assumed to be constant. Furthermore, we assume the mercury density on the wall, \(n_{Hg}|_{R}\), to be constant. We assume an equilibrium situation for the ion and the neutral density at \(t < 0\). At \(t=0\), the reaction coefficient for ionisation changes from \(k_{ion}|_{0}\) to \(k_{ion}\):  

\[
k_{ion}|_{0} \rightarrow k_{ion}, \text{ at } t = 0.
\]  
(A.3)

For \(t > 0\), \(k_{ion}\) is assumed to be constant. The change in the ion and atom density can be calculated by using the mass balance for ions and atoms, and solving these two differential equations. With these assumptions, the mass balances become:

\[
\frac{\partial n_{Hg}}{\partial t} = -n_{Hg} n_{e} k_{ion} + \frac{D_{0}}{\Lambda^{2}} (n_{Hg}|_{R} - n_{Hg}),
\]  
(A.4a)

and

\[
\frac{\partial n_{e}}{\partial t} = +n_{Hg} n_{e} k_{ion} - \frac{D_{a}}{\Lambda^{2}} n_{e},
\]  
(A.4b)

where the value for \(k_{ion}\) is corrected for the fact that the electron density is not equal to the mercury ion density. The values for the mercury ion density and the mercury atom density are evaluated at the tube axis. The two equations cannot be solved analytically. Therefore, we have to make another approximation. We assume that the densities will reach an equilibrium value when \(t \rightarrow \infty\). We use the following definitions:

\[
n_{Hg}(t) = n_{Hg}(\infty) + \delta_{Hg}(t),
\]  
(A.5a)

and

\[
n_{e}(t) = n_{e}(\infty) + \delta_{e}(t),
\]  
(A.5b)

where \(\delta\) is the deviation from the equilibrium value \(n(\infty)\). We substitute these expressions into expression (A.4), and assume the deviations to be small with respect to the equilibrium values. Obviously, this is an approximation for lower values of \(t\). However, the deviations are very small at higher values of \(t\), since the densities approach their equilibrium value. The resulting equations for the density fluctuations are

\[
\frac{\partial \delta_{Hg}}{\partial t} = \left( \frac{D_{0}}{\Lambda^{2}} + n_{e}(\infty) \cdot k_{ion} \right) \delta_{Hg} - n_{Hg}(\infty) \cdot k_{ion} \cdot \delta_{e},
\]  
(A.6a)

and

\[
\frac{\partial \delta_{e}}{\partial t} = n_{e}(\infty) \cdot k_{ion} \cdot \delta_{Hg},
\]  
(A.6b)
These expressions can be solved analytically, resulting in the following expressions for the densities:
\[
\begin{align*}
n_{Hg}(t) &= n_{Hg}(\infty) + n_{Hg}(0) - n_{Hg}(\infty) \exp(-v \cdot t) \cdot \cos(\omega t + \alpha), \quad (A.7a)
\end{align*}
\]
and
\[
\begin{align*}
n_+ (t) &= n_+ (\infty) + \frac{n_{Hg}(0) - n_{Hg}(\infty)}{n_{Hg}(\infty) k_{ion} \cos(\alpha)} \exp(-v \cdot t) \cdot [-v \cos(\omega t + \alpha) + \omega \sin(\omega t + \alpha)]. \\
&= n_+ (\infty) + \frac{n_{Hg}(0) - n_{Hg}(\infty)}{n_{Hg}(\infty) k_{ion} \cos(\alpha)} \exp(-v \cdot t) \cdot [-v \cos(\omega t + \alpha) + \omega \sin(\omega t + \alpha)], \quad (A.7b)
\end{align*}
\]
where the expressions for \(\alpha\), \(v\), and \(\omega\) are:
\[
\begin{align*}
\alpha &= \arctan \left( \frac{v + 1}{\omega n_{Hg}(\infty) - n_+ (0)} \right), \quad (A.8a)
\end{align*}
\]
\[
\begin{align*}
v &= \frac{1}{2} \frac{D_0}{\lambda^2} n_{Hg}(\infty), \quad (A.8b)
\end{align*}
\]
and
\[
\begin{align*}
\omega &= \frac{1}{2} \sqrt{4n_{Hg}(\infty) k_{ion}^2 \left( \frac{D_0}{\lambda^2} + n_+ (\infty) k_{ion} \right)^2}. \quad (A.8c)
\end{align*}
\]

Since we made a lot of approximations, the expressions for \(\alpha\) and \(\omega\) are not that important. However, the expression for \(v\) is interesting, since it determines the characteristic time for the transport of mercury from the plasma to the wall. This characteristic time is:
\[
\tau = v^{-1} = \frac{\lambda^2}{D_0} \left( 2 \frac{n_{Hg}(\infty)}{n_{Hg}(\infty)} \right) = \tau_D \cdot \left( 2 \frac{n_{Hg}(\infty)}{n_{Hg}(\infty)} \right), \quad (A.9)
\]
where \(\tau_D\) is the characteristic time for the diffusion of mercury atoms. It is clear from this expression that when the resulting mercury atom density at the tube axis is lower than half the density at the wall, then the cataphoresis process is faster than the ‘free’ diffusion of mercury.

**Appendix B: The driving frequency and the influence of averaging over time.**

From the application point of view, it is convenient to use standard equipment and standard discharge conditions for our experimental study. Therefore, for the main part of the experiments we used a standard Philips BRC 411/01 ballast as a power source. The driving frequency is 35 kHz for this power source. The ballast ignites the discharge automatically, preheating the electrodes before the ignition. The main reason for using an alternating current (AC) source is to avoid axial cataphoresis effects: when the time-averaged current is directed towards one of the electrodes, a net transport of mercury atoms will take place towards this electrode. The mercury-noble-gas discharge is very sensitive to axial variations in the mercury density. Therefore, direct current (DC) discharges cannot be used. Another widely used driving frequency for a fluorescent lamp is 50 Hz. However, this frequency leads to serious experimental problems since the discharge extinguishes every half-cycle when the electric field is too low to sustain the discharge. This is due to the fact that the electrons are lost at an ambipolar frequency higher than the driving frequency. For 35 kHz, this is not the case so even when the electric field is zero, enough electrons remain to sustain the discharge. However, there is an important difference between a DC discharge and this 35 kHz discharge. This is due to the fact that there is some modulation of the relevant discharge parameters with
the doubled driving frequency. In the experimental study, we have to be aware of this modulation. This appendix discussed the influence of averaging over time for the ultraviolet absorption measurements, the ‘self-absorption measurements’ and the Thomson scattering measurements.

**B.1 The ultraviolet absorption measurements**

We measured the modulation of the 254 nm output of the source lamp. It is approximately 2 %e. It reasonable to assume that the modulation is this low because of radiation trapping of the resonance radiation in the source lamp. In the source lamp, the mercury pressure is rather high since the cold-spot temperature is 46 ºC. In the probe lamp, the characteristic time for diffusion of neutral mercury atoms is way too low to have significant modulation of the density of these species. The characteristic time can be calculated using the following expression

\[ \tau = \frac{\Lambda^2}{D_{Hg}} \approx \frac{R/2.4}{D_{Hg}} \],

(B.1.1)

where \( \Lambda \) is the characteristic length for diffusion, \( D_{Hg} \) is the diffusion coefficient for mercury atoms measured by Redko et al.\(^9\) For the characteristic length, we used the value as used in the Schottky theory. The resulting characteristic time is approximately 13 milliseconds. This time is much larger than one period of the driving frequency of the discharge. Therefore, we can assume the mercury density profile to be independent of the phase of the driving voltage. On the other hand, the modulation frequency of the 254 nm output of the probe lamp is much higher than the chopper frequency. Therefore, the output of the lock-in amplifier will not be influenced by the modulation of the probe lamp output.

**B.2 The ‘self-absorption measurements’**

We assume the excited state densities to have the same modulation as the emission. The source lamp and the probe lamp are operated at the same conditions. Furthermore, we assume linear absorption, so the transmission can be written as:

\[ t_r = 1 - \alpha \cdot \langle \varepsilon \rangle \],

(B.2.1)

where \( \varepsilon(t) \) is the intensity of the source lamp in the beam which is shone through the probe lamp, and \( \alpha \) is a constant. We assume that \( \varepsilon(t) \) can be written as:

\[ \varepsilon(t) = \varepsilon_0 \cdot \sin^2(\omega t) = \frac{1}{2} \varepsilon_0 \cdot (1 - \cos(2\omega t)) \],

(B.2.2)

where \( \omega \) is the angular frequency of the electric field. For the determination of the influence of self-absorption, the phase of both discharges should be the same. Then the time-averaged value of the transmission is:

\[ \langle t_r \rangle = 1 - \alpha \cdot \langle \varepsilon \rangle = 1 - \alpha \cdot \frac{1}{2} \],

(B.2.3)

On the other hand, the time-averaged intensity of the transmitted radiation divided by the time-averaged intensity of the incident radiation is:

\[ \frac{\langle t_r \cdot \varepsilon(t) \rangle}{\langle \varepsilon(t) \rangle} = 1 - \alpha \cdot \frac{\langle \varepsilon^2(t) \rangle}{\langle \varepsilon(t) \rangle} = 1 - \alpha \cdot \frac{3}{4} \].

(B.2.4)

For uncorrelated phases, the expression for the time-averaged intensity of the transmitted radiation divided by the time-averaged intensity of the incident radiation is:
\[
\frac{\langle t \cdot \varepsilon(t) \rangle}{\langle \varepsilon(t) \rangle} = \frac{\langle (1 - \alpha \cdot \varepsilon(t')) \cdot \varepsilon(t) \rangle}{\langle \varepsilon(t) \rangle} = \frac{(1 - \alpha \langle \varepsilon(t) \rangle) \cdot \langle \varepsilon(t) \rangle}{\langle \varepsilon(t) \rangle} = 1 - \alpha \cdot \langle \varepsilon(t) \rangle = 1 - \alpha \cdot \frac{1}{2}.
\]

This expression is equal to the expression of the time-averaged transmission. However, for the determination of the influence of self-absorption, the time-averaged value of the transmitted radiation (divided by the time-averaged value of the incident radiation) should be used. This quantity is a factor 2/3 smaller for the uncorrelated case than for the in-phase case. The values for the transmission as given in section 3.6.2 are for the uncorrelated case. The correction to the in-phase case doesn’t result in a qualitative difference. Since we only used the transmission in order to estimate the influence of self-absorption, the correction is not carried out.

**B.3 Thomson scattering**

We assume no correlation between the phase of the electric field and the moment when the Thomson scattering spectrum is measured. This means that we measure the time-averaged Thomson scattering spectrum. The amount of electrons in a certain velocity interval is averaged over time. Since Thomson scattering spectra are a linear function of the electron density, the average Thomson scattering spectrum leads to the average value for the electron density. For the electron temperature the situation is different. Obviously, the averaged Thomson scattering spectrum is different from the Thomson scattering spectrum of the average temperature. We define

\[
\langle f_v \rangle = \frac{1}{T} \int_0^T \frac{1}{\sqrt{\pi \Delta \lambda_{th}(t)}} \exp\left(-\frac{\lambda - \lambda_0}{\Delta \lambda_{th}(t)}\right) dt
\]

as the averaged Thomson scattering spectrum. In this expression, \(T\) is the period of the modulation, and \(\Delta \lambda_{th}(t)\) is the thermal width of the Thomson scattering spectrum:

\[
\Delta \lambda_{th}(t) = 2 \frac{\lambda_0}{c} \sqrt{\frac{kT_e(t)}{m_e}},
\]

where \(\lambda_0\) is the laser wavelength, and \(m_e\) is the electron mass. On the other hand, the expression for the Thomson spectrum of the average temperature is:

\[
f_v(\langle T_e \rangle) = \frac{1}{\sqrt{\pi \Delta \lambda_{th}}} \exp\left(-\frac{\lambda - \lambda_0}{\Delta \lambda_{th}}\right),
\]

where the expression for \(\langle \Delta \lambda_{th} \rangle\) is

\[
\langle \Delta \lambda_{th} \rangle = 2 \frac{\lambda_0}{c} \sqrt{\frac{kT_e}{m_e}}.
\]

Figure B.3.1 shows both the averaged Thomson spectrum, and the Thomson spectrum using the averaged temperature. For the electron temperature we assume

\[
T_e(t) = \langle T_e \rangle + \Delta T \cdot \sin(2\omega t),
\]

where \(\Delta T\) is the amplitude of the modulation, and \(\omega\) is the angular frequency of the driving voltage.
For the calculation of these two profiles we assumed an average temperature of 1 eV and 25% modulation, i.e. \( \Delta T = 0.25 \) eV. It is clear that the deviation between the two spectra is very small. When we use the fitting procedure used for the measurements, the electron temperature inferred from the averaged spectrum is 1.03 eV, so 3% higher than the average temperature.

**Appendix C: The influence of the coaxial water jacket.**

This appendix describes how it can be shown that a water jacket around the tube does not change the emission measurements. We assume the water jacket and the tube to have an infinitely small thickness. Then there is no influence of the walls, we have a tube with a cylinder around it, like in figure C.1.1. In this figure, the definition of the angles \( \alpha_1, \alpha_2, \alpha_3, \) and \( \alpha_4, \) the tube radius \( R_1, \) the cylinder radius \( R_2, \) \( y, y^*, \) the indices of refraction \( n_0 \) and \( n_1, \) and the line \( l \) is shown. We assume the discharge to have cylindrical symmetry.

The two lenses make an image of a certain surface of the focal plane of the first lens. From this surface, we capture the radiation within a certain solid angle. The emission measurements are not affected by the cylinder around the tube when the surface area, the solid angle, and the lateral position of this surface are not changed. We will start with this lateral position.

**C.1 Conservation of lateral position.**

We have to show that the cylinder around the tube does not change the lateral position \( y \) inside the tube. So using the definitions given in figure C.1.1, we have to show that \( y^* = y. \) In order to do this, we have to apply Snell’s law to the boundaries where the index of refraction changes. Furthermore, we have to use the sine rule in order to find a relation between \( \alpha_2, \) and \( \alpha_3. \) The result is:

\[
y = R_2 \cdot \sin(\alpha_1) = R_2 \cdot \frac{n_1}{n_0} \cdot \sin(\alpha_2) = R_2 \cdot \frac{n_1}{n_0} \cdot \frac{R_1}{R_2} \cdot \sin(\alpha_3) = R_1 \cdot \sin(\alpha_4) = y^*. \quad (C.1.1)
\]
So the coaxial boundary rotates the beam, however, since we assumed cylindrical symmetry this has no influence.

![Coaxial Geometry](image1)

**Fig. C.1.1: The coaxial geometry.**

### C.2 Conservation of solid angle.

The solid angle in which the radiation is captured is determined by the focal distance of the first lens $f$, along with the radius of the diaphragm. We have to prove that inside the cylinder on the line $l$, we capture the radiation in the very same solid angle. Note that this is in general not the case for radiation captured by a negative lens like the cylinder. In order to show that in our case the solid angle is not changed by the cylinder, we consider a small volume on the line $l$. We assume this volume to radiate isotropically. The radiation, which is emitted within the solid angle $\delta \Omega$ has to be within the same solid angle after passing the cylinder. In order to show that this is the case, we consider the beam as drawn in figure C.1.1. When we rotate this beam over an angle $\delta \alpha$ around the intersection of the line $l$ and the beam itself, we constructed a second beam which we will call the rotated beam. We now have to show that the angle between the rotated beam and the original beam is $\delta \alpha$, even outside the cylinder. Figure C.2.1 shows the construction we use to prove that this holds.

![Construction for the conservation of the solid angle](image2)

**Fig. C.2.1: Construction for the conservation of the solid angle.**

We have to show that $\delta \alpha$ is equal to $\delta \phi$, then the equivalent of $\alpha_i$ for the rotated beam is equal to $\alpha_i$, and therefore the angles $\alpha_i$ and $\alpha_i$ are the same. We use the first order Taylor expansion (linearisation) for the sine, the cosine, and the tangent of both $\delta \alpha$ and $\delta \phi$, this can be done since the solid angle $\delta \Omega$ is very small. We then obtain the following expression for $\delta \alpha$ and $\delta \phi$. 

\[ \delta \alpha = \delta \phi \]
So the angle between the two beams is always equal to $\delta \alpha$. Therefore, we can conclude that all the radiation and only the radiation emitted in the solid angle $\delta \Omega$ will be captured by the optical system, independent of the lateral position. An important implication is that the line $l$ is positioned in the focal plane of the first lens. This focal plane is different for different lateral positions.

C.3 Conservation of the imaged area.

In order to show that the imaged surface area is also unaffected by the cylinder, we rotate the beam in figure C.1.1 over the angle $\delta \beta$ around the point where it intersects with the first lens. The angle $\delta \beta$ is dependent on the size of the aperture of the optical fibre. This aperture determines the maximum imaged area. For this rotated beam, we can calculate the equivalents of $\alpha_1$, $\alpha_2$, $\alpha_3$, and $\alpha_4$. The construction we use for the proof is shown in figure C.3.1.

![Fig. C.3.1: The construction for the proof of conservation of the imaged area.](image)

The length $l(0)$ is a measure for the imaged area. We will use the linearisation described in the preceding section for $\delta \beta$, and $\delta \varphi$. In the situation without the water jacket, the following expression holds

$$l(0) = f \cdot \delta \beta.$$  \hspace{1cm} (C.3.1)

Using the construction shown in figure C.3.1, we can calculate the equivalent of $\alpha_1$ for the rotated beam, $\alpha_1^*$

$$\alpha_1^* = \alpha_1 - \delta \beta - \delta \varphi.$$  \hspace{1cm} (C.3.2)

For the angle $\delta \varphi$, we use a derivation equivalent to the derivation described in the preceding subsection. The following expression holds:

$$\cos(\alpha_i) \frac{b_1}{b_2} = \frac{(f - R_2 \cdot \cos(\alpha_i)) \cdot \delta \beta}{R_2 \cdot \delta \varphi}.$$  \hspace{1cm} (C.3.3)

This expression can be rewritten in:

$$\delta \varphi = \frac{(f - R_2 \cdot \cos(\alpha_i)) \cdot \delta \beta}{R_2 \cdot \cos(\alpha_i)} = \delta \beta \cdot \frac{f}{R_2 \cdot \cos(\alpha_i)} - \delta \beta.$$  \hspace{1cm} (C.3.4)

We now define for all the angles $\alpha$: \hspace{1cm}
\[ \alpha^* = \alpha + \delta \alpha, \]  
(C.3.5)

and we assume that \( \delta \alpha \ll \alpha \), so we can use the linearisation for all the \( \delta \alpha \)’s. The expression for \( \delta \alpha_i \) is:

\[ \delta \alpha_i = -(\delta \beta + \delta \varphi) = -\delta \beta \cdot \frac{f}{R_2 \cdot \cos(\alpha_i)}. \]  
(C.3.6)

The expressions for \( \delta \alpha_2, \delta \alpha_3, \) and \( \delta \alpha_4 \) can be derived from the expressions relating the adjacent angles \( \alpha \) and the adjacent angles \( \alpha^* \);

\[
\sin(\alpha_i) = \frac{n_1}{n_0} \cdot \sin(\alpha_2), \\
\sin(\alpha_2) = \frac{R_1}{R_2} \cdot \sin(\alpha_3), \\
and \sin(\alpha_3) = \frac{n_1}{n_0} \cdot \sin(\alpha_4). \\
\]  
(C.3.7)

The same expressions hold for the rotated beam, for example:

\[
\sin(\alpha_i + \delta \alpha_i) = \frac{n_1}{n_0} \cdot \sin(\alpha_2 + \delta \alpha_2), \\
\sin(\alpha_2 + \delta \alpha_2) = \frac{n_1}{n_0} \cdot \sin(\alpha_3) + \delta \alpha_2 \cdot \frac{n_1}{n_0} \cdot \cos(\alpha_2), \\
\delta \alpha_2 \cdot \cos(\alpha_2) = \delta \alpha_3 \cdot \frac{n_1}{n_0} \cdot \cos(\alpha_2). \\
\]

Rewriting this expression gives us:

\[ \delta \alpha_2 = \delta \alpha_i \cdot \frac{n_0}{n_i} \cdot \frac{\cos(\alpha_i)}{\cos(\alpha_2)}. \]  
(C.3.8)

This derivation can also be performed for the angles \( \delta \alpha_3 \) and \( \delta \alpha_4 \), resulting in:

\[
\delta \alpha_3 = \delta \alpha_i \cdot \frac{n_0}{n_i} \cdot \frac{R_2}{R_1} \cdot \cos(\alpha_1), \\
\delta \alpha_4 = \delta \alpha_i \cdot \frac{R_2}{R_1} \cdot \cos(\alpha_1). \\
\]  
(C.3.9)

The expression for \( \delta \xi \) is:

\[ \delta \xi = \delta \alpha_4 - \delta \varphi_2. \]  
(C.3.11)

The expression for \( l(\mathbf{x}) \) is:

\[ l(\mathbf{x}) = R_1 \cdot \delta \varphi_2 \cdot \cos(\alpha_2). \]  
(C.3.12)

For \( l(0) \), the following expression holds:

\[ l(0) = l(\mathbf{x}) + x \cdot \delta \xi = R_1 \cdot \delta \varphi_2 \cdot \cos(\alpha_4) + R_1 \cdot \cos(\alpha_4) \cdot \delta \xi, \]

which can be rewritten in:

\[ l(0) = R_1 \cdot \cos(\alpha_4) \cdot (\delta \varphi_2 + \delta \xi), \]

\[ = R_1 \cdot \cos(\alpha_4) \cdot \delta \alpha_4, \]

\[ = R_1 \cdot \cos(\alpha_4) \cdot \delta \alpha_i \cdot \frac{R_2}{R_1} \cdot \cos(\alpha_1), \]

\[ = \delta \alpha_i \cdot R_2 \cdot \cos(\alpha_1), \]
\[
= \delta \beta \cdot \frac{f}{R_2 \cdot \cos(\alpha)} \cdot R_2 \cdot \cos(\alpha), \\
= \delta \beta \cdot f.
\]  
(C.3.14)

So the same expression holds for \( l(0) \), as without the jacket (expression C.3.1). The size of the imaged area is not changed by the water jacket.

For the influence of the jacket on the angle \( \delta \xi \), we need an expression for \( \delta \phi \), this expression is:

\[
\delta \phi = \delta \phi + (\alpha_2' - \alpha_2 - (\alpha_3' - \alpha_3)) = \delta \phi + \delta \alpha_2 - \delta \alpha_3,
\]  
(C.3.15)

The expression for \( \delta \xi \) becomes:

\[
\delta \xi = \delta \alpha_4 - \delta \alpha_2
= \delta \alpha_4 \cdot \frac{R_2 \cdot \cos(\alpha)}{R_1 \cdot \cos(\alpha)} + \delta \beta + \delta \alpha_4 - \delta \alpha_4 \cdot \frac{1}{n} \cdot \frac{\cos(\alpha)}{\cos(\alpha)} + \delta \alpha_4 \cdot \frac{1}{n} \cdot \frac{R_2 \cdot \cos(\alpha)}{R_1 \cdot \cos(\alpha)}
= \delta \alpha_4 \left( \frac{R_2 \cdot \cos(\alpha)}{R_1 \cdot \cos(\alpha)} + 1 - \frac{1}{n} \cdot \frac{\cos(\alpha)}{\cos(\alpha)} + \frac{1}{n} \cdot \frac{R_2 \cdot \cos(\alpha)}{R_1 \cdot \cos(\alpha)} \right) + \delta \beta,
\]

\[
= \delta \beta \left( 1 - \frac{f}{R_1 \cdot \cos(\alpha)} - \frac{1}{R_2 \cdot \cos(\alpha)} + \frac{1}{n} \cdot \frac{f}{R_2 \cdot \cos(\alpha)} - \frac{1}{n} \cdot \frac{1}{R_1 \cdot \cos(\alpha)} \right).
\]  
(C.3.16)

This expression is different from \( \delta \beta \), so the water jacket changes the detection cone, outside the focal plane. This change is symmetrical around the ‘zero beam’.

The cone in which we capture the radiation widens a little. This decreases the spatial resolution of the measurements. However, this does not effect the amount of radiation we capture.

The created image will be changed in the axial direction of the cylinder, due to the refraction at the surfaces of the cylinder. The optical axis is oriented perpendicular to the axis of the cylinder. Besides this, we capture radiation in a very small solid angle. Therefore, we can use the same linearisation as described in the preceding subsections. The focal distance of the first lens in the plane constructed by the axis of the cylinder and the optical axis will change. The change \( \Delta f \) is equal to:

\[
\Delta f = \left\{ \sqrt{R_2^2 - \frac{n_0^2}{n_1^2} y^2} - \sqrt{R_1^2 - \frac{n_0^2}{n_1^2} y^2} \right\} \left( 1 - \frac{n_0}{n_1} \right).
\]  
(C.3.17)

In this expression, the first factor represents the distance that the beam travels through the ‘hollow cylinder’. De shift of the focal ‘plane’ influences the shape of the detection cone. However, since the positive column can be regarded as homogeneous along its axis, this influence has no influence on the measured emission. The solid angle, in which we capture the radiation and the imaged area are not affected by the cylinder. So we can conclude that there is no influence of the cylinder in the plane constructed by the axis of the cylinder and the optical axis.

\section*{C.4 The influence of the shape of the detection cone.}

In the last two subsections, we implicitly assumed that the shape of the detection cone is not influencing the emission measurements. This can be proven in a simple way. We consider a spatially uniform emitting plasma without boundaries that influence the imaging. First we assume the aperture of the fibre to be an infinitely small circle, i.e. a point. Then the area in the focal plane of the first lens, which is imaged on the fibre, is also a point. The two lenses...
capture the radiation emitted in the cone through the imaged point (detection cone) and collect all the radiation on the fibre. However, not all the radiation emitted in the detection cone will be imaged on the fibre. Only the photons that are emitted in the direction of the line that also contains the imaged point in the focal plane of the first lens will be imaged on the fibre. The other photons will not be captured by the first or the second lens or they will be imaged outside the fibre. So in the focal plane, the detection volume is infinitely small, while the solid angle in which the radiation is captured is finite. Outside the focal plane, the detection volume is finite, while the solid angle in which the radiation is captured is infinitely small.

When we now assume the fibre to have finite dimensions, the area in the focal plane, which is imaged on the fibre is finite. Outside the focal plane, we can use the same argument as used above, now stating that only the photons that are emitted in the direction of the line that also contains a point of the imaged area in the focal plane will be guided to the fibre. This means that for a point outside the focal plane, the solid angle in which the radiation is captured is finite. A more general conclusion can be drawn:

_The amount of photons that are captured from a slab perpendicular to the optical axis with a thickness dx, is equal to the amount of photons captured from a slab with a thickness dx situated symmetrically around the focal plane of the first lens._

This conclusion has a very important implication. The shape of the detection cone is not influencing the amount of radiation captured by the optical system. Using Abel inversion, this means that we can use expression 3.6.1, since the amount of photons captured by the optical system is not depending on the position along the optical axis. However, when the emitted radiation is not spatially uniform, the widening of the detection cone will decrease the spatial resolution of the measurements.

**Appendix D: Estimation of the spectral impurity of the ring-dye laser**

We assume that the spectral impurities of the laser are caused by dye jet emission that shines in the direction of the laser beam. The ratio of the solid angle of the laser beam at large distance to the total solid angle is $5 \cdot 10^{-7}$. The maximum dye jet emission is equal to the pump laser power, i.e. 7 W. So the maximum power of the spectral impurities of the laser beam is $4 \cdot 10^{-6}$ W. Approximately 10% of this radiation is emitted in the range 587.6 to 591.6 nm, the region of interest. The dye laser power used for the transmission measurements is higher than 400 mW, so we can conclude that the spectral impurity level in the region of interest is more than 6 orders of magnitude lower than the laser power.

**Appendix E: Calculating the eedf from a Thomson scattering spectrum.**

We assume the velocity distribution function to be isotropic. The drift velocity can be neglected compared to the random velocity of the electrons. The derivation starts with the velocity distribution function in spherical coordinates. Due to the isotropy, this function only depends on the absolute value of the velocity:

$$f_r(v, \theta, \phi) = f_r(|v|).$$  \hspace{1cm} (E.1)

Note that the absolute value signs are used to express the difference between expression E.1 and the Thomson scattering spectrum, i.e. the velocity distribution function in the direction of
the scattering vector for Thomson scattering. The $z$-axis is chosen in the direction of the scattering vector for Thomson scattering. The velocity distribution function in the $z$ direction can be obtained by integrating the distribution as given in expression E.1 over the plane perpendicular to the $z$-axis:

$$f(v_z) = \int_0^\infty dv_x \int_0^\infty dv_y f(v) \cdot v_z,$$  \hspace{1cm} \text{(E.2)}

We now define the velocity $v_r$ perpendicular to $v_z$. Using cylindrical co-ordinates in the plane perpendicular to the $v_z$ axis, the integral can be rewritten as:

$$f_r(v_r) = 2\pi \int_0^\infty dv_z f(v) \cdot v_r.$$  \hspace{1cm} \text{(E.3)}

This expression can be rewritten in

$$f_r(v_r) = \pi \int_{v_{r_{\text{min}}}}^{v_{r_{\text{max}}}} f_r(\sqrt{2}) dv_r = \pi \int_{v_{r_{\text{min}}}}^{v_{r_{\text{max}}}} f_r(\sqrt{2}) dv.$$  \hspace{1cm} \text{(E.4)}

and

$$f_r(v_r) = \pi \int_{v_{r_{\text{min}}}}^{v_{r_{\text{max}}}} f_r(\sqrt{2}) dv = 2\pi \int_{v_{r_{\text{min}}}}^{v_{r_{\text{max}}}} f_r(\sqrt{2}) dv.$$  \hspace{1cm} \text{(E.5)}

Differentiating both sides results in:

$$\frac{df_r(v_r)}{dv_r} = -2\pi \cdot f_r(v_r),$$  \hspace{1cm} \text{(E.6)}

so the following expression holds:

$$f_r(v_r) = \frac{-1}{2\pi v_r} \frac{df_r(v)}{dv}.$$  \hspace{1cm} \text{(E.7)}

Using this expression, the expression for the electron energy distribution function can be obtained:

$$f(E)dE = 4\pi v^2 f(\sqrt{E})dE = -\frac{2}{m} \frac{df_r(v)}{dv} \left|_{v = \sqrt{E/m}} \right. dE,$$  \hspace{1cm} \text{(E.8)}

This expression can be used to calculate the electron energy distribution function from the Thomson scattering spectrum.
Summary

For certain applications, it would be convenient to be able to change the colour temperature and the luminous flux of artificial lighting independently. A promising candidate for this purpose is the fluorescent lamp. This thesis describes the investigation of two variable colour fluorescent lamps. The mercury-noble-gas discharge in these lamps produces both mercury and noble-gas radiation, in contrast to the discharge in normal fluorescent lamps that produces only mercury radiation. The additional noble-gas radiation can be used to change the colour of the lamp. The research goal is to obtain a better understanding of the processes involved in the production of noble-gas radiation in mercury-noble-gas discharges. The two most important questions to be answered are: which discharge processes and conditions are important for the production of noble-gas radiation, and how can we produce this noble-gas radiation efficiently? To answer these questions, we performed an experimental study. The investigation comprises another important goal as well: the improvement and the development of diagnostic techniques.

The first discharge we studied is a fluorescent lamp with an additional capacitively coupled radio frequency (ccrf) discharge. We performed an optical and electrical characterisation of this hybrid lamp. For the electrical characterisation, we developed a new stray impedance characterisation technique. The results of the electrical and optical measurements show that the additional ccrf discharge is inefficient and that the intensity of the noble-gas spectral lines is low. Another important result is the fact that the mercury density significantly influences the emission spectrum.

This result led us to the introduction of a new lamp: the depleted discharge lamp. This is a discharge lamp in which the emission spectrum is controlled by varying the mercury density. A promising way to change the mercury density is using mercury depletion due to radial cataphoresis. The amount of noble-gas radiation produced by this lamp is much higher than the amount produced by the ccrf discharge. Therefore, the research narrowed down to the investigation of the depleted discharge. This discharge has been investigated using the following diagnostic techniques: optical emission spectroscopy, electrical measurements, ultraviolet absorption measurements, spatially- and time-resolved emission spectroscopy and Thomson scattering.

The first emission measurements have been performed to find the conditions where the influence of radial cataphoresis on the emission spectrum is significant. It appears that both the discharge current and the mercury pressure can be used to control the emission of noble-gas radiation. The effect of mercury depletion is observed indirectly in the emission spectrum. In order to proof the mercury depletion directly, we measured the mercury ground state density profile using ultraviolet absorption. The results of these measurements are compared to the results of the spatially resolved emission measurements, which show the regions where noble-gas radiation is produced. The noble-gas radiation is produced only when the mercury density is low. The time-resolved emission measurements show the time-evolution of the spectral lines of mercury and neon after the discharge is switched on. The intensity of the mercury lines decreases with time, while the intensity of the neon lines increases. The characteristic time for this decrease and increase is of the same order of magnitude as the characteristic time for mercury diffusion. The picture of the discharge as created by the measurements mentioned above misses the properties of the bulk of the electron gas.

These properties can be measured using Thomson scattering. There are two problems when Thomson scattering is performed in fluorescent lamps. At first, the electron density is rather low, which results in extremely low signal levels. The second problem is the stray light in the set-up. The first problem is solved by using a powerful laser and a sensitive detector.
The second problem is solved by using a sodium vapour absorption cell in the detection branch of the set-up, which absorbs the stray light. For this option to work, the scattering experiments should be performed at one of the two resonant lines of sodium. This necessitates the use of a dye laser. However, the spectral purity of dye lasers is not sufficient for this experiment. The broadband amplified spontaneous emission (ASE) produced by the laser should be reduced. For this, we designed an ASE filter. This filter consists of two spatial filters and twenty dispersion prisms. The design of the ASE filter is chosen in such a way that the transmission is maximised and the spectral width is minimised. The spectral width of the sodium cell is matched to the width of the ASE filter by adding argon to the cell. The argon atoms broaden the absorption sodium lines. The combination of the sodium absorption cell and the dye laser with ASE filter results in a stray light reduction of more than six orders of magnitude. This reduction is sufficient to perform Thomson scattering in fluorescent lamps. The electron density and temperature have been measured in an argon-mercury and a neon-mercury discharge.

The results of the Thomson measurements combined with the emission and absorption measurements result in the following picture of the radial cataphoresis process and the production of noble-gas radiation. A high current and a low mercury pressure result in a high ionisation degree of mercury. This high ionisation degree results in significant depletion of mercury. As a result, less inelastic collisions of electrons and mercury atoms will occur. The amount of electrons capable of exciting the noble-gas atoms will increase. Therefore, the discharge will start producing noble-gas radiation.

Regarding the application of the studied discharges, we can conclude that both the sheath regions of the ccrf discharge and the radial cataphoresis process can be used to produce noble-gas radiation in mercury-noble-gas discharges. The efficiency of the ccrf discharge for the production of noble-gas radiation is too low. On the other hand, the efficiency of the depleted discharge lamp offers a much better perspective for commercialisation. For this lamp, amplitude modulation offers the opportunity to control the colour and the luminous flux independently.
Samenvatting

Voor sommige toepassingen is onafhankelijke regelbaarheid van de kleurtemperatuur en de lichtstroom van kunstmatige verlichting gewenst. Dit proefschrift beschrijft het onderzoek aan de meest veelbelovende kandidaat hiervoor: een kleurinstelbare fluorescentielamp. De kwik-edelgasontlading in deze lamp zendt zowel kwikstraling als edelgasstraling uit, dit in tegenstelling tot de ontlading in normale fluorescentielampen die alleen kwikstraling produceert. Het toevoegen van edelgasstraling kan gebruikt worden om de kleur van de lamp te veranderen. Het doel van het onderzoek is het zo goed mogelijk begrijpen van de processen die belangrijk zijn voor de productie van edelgasstraling in kwik-edelgasontladingen. De belangrijkste vragen die beantwoord moeten worden zijn: wat zijn de belangrijke processen en omstandigheden voor de productie van edelgasstraling en hoe is edelgasstraling het meest efficiënt te produceren? Een experimentele studie moet deze vragen beantwoorden. Een nevendoel van deze experimentele studie is het ontwikkelen en verbeteren van diagnostische technieken.

Als eerste lamp is een fluorescentielamp met een extra capacitief gekoppelde radiofrequente ontlading bestudeerd. Het elektrisch gedrag en het emissiespectrum van de ontlading in deze hybride lamp zijn onderzocht. Voor de bepaling van de ontladingsimpedantie is een nieuwe methode ontwikkeld om de parasitaire impedanties te bepalen. De resultaten van de optische en elektrische metingen geven aan dat de extra ontlading weinig licht produceert en inefficiënt is. Ook wijzen de metingen uit dat de kwikdichtheid de intensiteit van de edelgasspectraallijnen significant beïnvloedt. Deze ontdekking heeft geleid tot een nieuwe lamp, de zogenaamde depletielamp. Dit betreft een fluorescentielamp waarin de hoeveelheid edelgasstraling geregeld wordt door de kwikdichtheid te variëren. Een manier om de kwikdichtheid te regelen is het gebruik maken van kwikdepletie door het radiale kataforeseproces. De hoeveelheid edelgasstraling die geproduceerd wordt door de depletielamp is veel hoger dan door de hybride lamp. Daarom is het onderzoek toegepast op het bestuderen van de ontlading in de depletielamp. Het onderzoek aan deze ontlading is uitgevoerd met behulp van de volgende diagnostieken: optische emissiespectroscopie, elektrische metingen, absorptiespectroscopie in het ultraviolet, plaatsopgeloste en tijdsopgeloste emissiespectroscopie en Thomsonverstrooiingsmetingen.

De eerste emissiemetingen zijn uitgevoerd om de condities waar de invloed van de kwikdichtheid op het emissiespectrum duidelijk zichtbaar is in kaart te brengen. Er is gebleken dat de kwikdruk en de ontladingsstroom beide gebruikt kunnen worden om de hoeveelheid edelgasstraling te regelen. De absorptiemetingen in het ultraviole zijn uitgevoerd om de kwikdichtheid in de ontlading te bepalen. Het resultaat van deze metingen geeft een direct bewijs voor de depletie van kwik. De plaatsopgeloste emissiemetingen geven aan waar de edelgasstraling geproduceerd wordt in de ontlading. Het blijkt dat de edelgasstraling geproduceerd wordt in de gebieden waar de kwikdichtheid laag is. De tijdsopgeloste metingen laten zien wanneer edelgasstraling geproduceerd wordt na het aanzetten van de ontlading. De intensiteit van de spectraallijnen van kwik neemt af met de tijd, terwijl de intensiteit van de neon lijnen juist toeneemt. De karakteristieke tijd voor deze toename en afname is van dezelfde orde als de karakteristieke tijd voor kwikdiffusie. Het beeld dat door bovenstaande metingen gecreeëerd wordt mist alleen de eigenschappen van het elektronengas.

Het elektronengas is bestudeerd met behulp van Thomsonverstrooiing. Er zijn twee problemen met de toepassing van Thomsonverstrooiing in fluorescentielampen. Ten eerste is de elektronendichtheid erg laag. Dit leidt tot een extrem laag Thomsonsignaal. Ten tweede is het parasitaire strooilichtniveau zeer hoog in vergelijking met het Thomsonsignaal. Het eerste probleem is overwonnen door een krachtige laser te gebruiken, in combinatie met een
gevoelige detector. Het tweede probleem is opgelost door gebruik te maken van een natrium-absorptiecel die het strooilicht absorbeert. De verstrooiingsmetingen dienen dan uitgevoerd te worden bij de golflengte van één van de resonante lijnen van natrium. Dit maakt het gebruik van een kleurstoflaser noodzakelijk. Het nadeel van deze laser is het feit dat de spectrale zuiverheid van de laserstraal niet hoog genoeg is. De laser produceert namelijk ook een breedbandig ASE (Amplified Spontaneous Emission) signaal. De spectrale zuiverheid is verhoogd met behulp van een ASE filter. Dit filter bestaat uit twee ruimtelijke filters en twintig dispersieprisma’s. Het ontwerp is zo uitgevoerd dat de transmissie maximaal is en de spectrale breedte minimaal. De spectrale breedte van de natriumcel is aangepast aan de breedte van het ASE filter door argon aan het natrium toe te voegen. De argon atomen verbreden de natrium absorptielijnen. De combinatie van de kleurstoflaser met het ASE filter en de natriumcel resulteert in de onderdrukking van het parasitaire strooilicht met zes ordes. Dit is voldoende om Thomsonverstooiing uit te voeren in een fluorescentielamp. De elektronendichtheid en de elektronentemperatuur zijn gemeten in een argon-kwik- en een neon-kwikontlading.

De resultaten van de Thomsonmetingen, gecombineerd met de emissie- en absorptiemetingen resulteren in het volgende beeld van het kataforeseproces en de productie van edelgasstraling: Een hoge stroom of een lage kwikdruk leiden tot een hoge ionisatiegraad. De hoge ionisatiegraad resulteert in significante kwikdepletie. Hierdoor vinden er minder inelastische botsingen van elektronen met kwikatomen plaats. De dichtheid van de elektronen, die in staat zijn om een edelgasatoom aan te slaan, zal stijgen. Dit resulteert in de productie van edelgasstraling.

Met betrekking tot de toepassing van de bestudeerde ontladingen kan geconcludeerd worden dat zowel de elektrodegebieden van de capacitief gekoppelde radiofrequente ontlading als radiale kataforese gebruikt kunnen worden voor de productie van edelgasstraling in een kwik-edelgasontlading. De efficiëntie van de radiofrequente ontlading is te laag. Daarentegen biedt de veel betere efficiëntie van de depletielamp wel voldoende perspectieven voor een commerciële toepassing. Amplitude modulatie van de ontladingsstroom biedt dan de mogelijkheid om de kleur en de lichtstroom van deze lamp onafhankelijk van elkaar in te stellen.
Dankwoord

Als eerste wil ik mijn eerste promotor Gerrit Kroesen bedanken voor het vertrouwen, de vrijheid en de rust die hij me de afgelopen jaren gegeven heeft. Ook bedank ik Frits de Hoog die mede-initiatiefnemer van dit project was. Uiteraard gaat mijn dank ook uit naar de stagiaires Peter van den Oever, Danny van den Akker, Guido Schiffelers, Arnold van der Starre, Cécile Mercusot en Bart de Loos die elk hun steentje aan dit proefschrift bijgedragen hebben. Verder is Hans Freriks onmisbaar geweest voor de technische ondersteuning en voor het ontwerpen van de opstellingen. Ook Loek Baede en Lambert Bisschops bedank ik voor de technische bijstand. Het modelleerteam van ETP wil ik bedanken voor de prettige samenwerking, in het bijzonder Jan van Dijk, Bart Hartgers en de stagiaires Bart Vorselaars en Astrid Stijfs. Joost van der Mullen bedank ik voor de vele verlichtende discussies en het onder de aandacht brengen van het belang van de kwispelende elektronenstaart. De samenwerking met de medewerkers van de faculteitswerkplaats en de gemeenschappelijke technische dienst is zeer vruchtbaar gebleken, iedereen bedankt. Marco Haverlag, Henk Vogels, Rolf de Man en John Gielen en met name Frans Ligthart van Philips Lighting Eindhoven dank ik voor hun belangstelling en expertise. Tweede promotor Bill Graham en de leden van de leescommissie Niek Lopes Cardozo, Daan Schram en Marnix van der Wiel dank ik voor hun kritische kijk op mijn proefschrift. Verder ben ik mijn (oud) collega’s van de groep EPG dankbaar voor de gezelligheid: Rina Boom, Jean-Charles Cigal, Marianne van den Elshout, Marc van de Grift, Gerjan Hagelaar, Daiyu Hayashi, Tarik Hbid, Marcel Hemerik, Wilfred Hoeven, Carole Maurice, Gabriela Paeva, Wijnand Rutgers, Eva en Winfred Stoffels, Geert Swinkels en Eddie van Veldhuizen.

Naast bovengenoemden dank ik mijn ouders. Zij stonden aan de basis van mijn opleiding, hun advies en aanmoedigingen zijn zeer belangrijk geweest. Verder dank ik alle vrienden en familie die de afgelopen jaren interesse getoond hebben in mijn werk. Als laatste dank ik Esme die altijd geïnteresseerd was en me altijd gesteund heeft, maar die vooral het leven buiten mijn promotiewerk kleur gaf.
Curriculum Vitae

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Stellingen

behorende bij het proefschrift

Plasma Control of the Emission Spectrum of Mercury-Noble-Gas Discharges.

door

Leon Bakker

Eindhoven, 7 december 2000
I
Radiale kataforese kan gebruikt worden om het emissiespectrum van een kwik-
edelgasontlading te variëren.
dit proefschrift

II
De invloed van parasitaire verstrooiing tijdens Thomson- en Ramanverstrooiingsexperimenten wordt drastisch gereduceerd door het gecombineerde gebruik van een kleurstoflaser, een amplified spontaneous emission filter en een natrium-absorptiecel.
dit proefschrift

III
In de plasmafysica wordt het belang van eenvoudige, benaderende diagnostieken vaak ten onrechte onderschat ten opzichte van dat van zuivere maar complexe diagnostieken.

IV
Bij de beoordeling van wetenschappelijke prestaties wordt vaak ten onrechte het vinden belangrijker gevonden dan het zoeken.

V
Kennis van andere vakgebieden en wetenschappen leidt tot een beter begrip en een betere beoefening van het eigen vakgebied.

VI
Handhaving van de verkeersregels voor bromfietsers is een betere maatregel ter vermindering van ongevallen dan het gedogen van overtredingen en het zogenaamde zichtbaar maken van de bromfietser.

VII
Recente opiniepeilingen† wijzen uit dat de term fiduciair geld voor de Euro ongepast is.
† NIPO, 26 september 2000.
VIII
In het huidige, gesponsorde topsportklimaat geldt voor veel sporters het motto: *geld is tijd*.

IX
In tegenstelling tot wat het vaak vertoonde kuddegedrag van haar leden doet vermoeden, biedt een studentenvereniging uitstekende mogelijkheden tot persoonlijke ontwikkeling en ontplooiing.

X
Beleefde automobilisten hinderen de snelle doorstroming op de snelweg tijdens het zogenaamde ritsen.

XI
Zolang de aanwezigheid van camera's het gedrag van de gefilmede beïnvloedt kan er geen sprake zijn van real-life soap.

XII
Alhoewel de mobiele telefoon veelal gebruikt wordt voor sociale doeleinden, resulteert het gebruik ervan vaak in asociaal gedrag.