Plasma Breakdown
of Low-Pressure Gas Discharges

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Chapter 1

General introduction

Abstract. This thesis deals with investigations of breakdown processes in gas discharges. Plasma breakdown can generally be defined as the transition process from an insulating, neutral gas to a conducting, (partially) ionized state. The fundamental processes involved in this phenomenon are the subject of this thesis. This chapter is intended as a general introduction to gas discharges and more specifically, plasma breakdown. Further, it describes the scope of the thesis and the content of the different chapters.
Chapter 1. General introduction

1.1 Gas discharges

People have been fascinated by gas discharges for a very long time. In early times, natural gas discharges such as lightning, aurorae and St. Elmo’s fire were commonly observed and associated with acts of God. In the 18\textsuperscript{th} century, advances in physics, especially the discovery and understanding of electricity, provided a scientific explanation for these impressive phenomena. Nowadays, many of the physical concepts of atmospheric gas discharges are well understood. Nevertheless, the beauty of these natural gas discharges never ceased to impress and inspire humans.

The observations and studies of natural gas discharges encouraged people to try creating their own discharges. A gas discharge can be created by directing electrical energy through a gas. To achieve this it was first of all necessary to create and store considerable amounts of electrical charge. Major breakthroughs in the field of gas discharges therefore came with the invention of charge-storage devices like the Leyden jar by von Kleist and van Musschenbroek, both in 1745 [1], and the invention of the electrochemical battery, by Volta [2] in 1800. These devices provided the energy source for the first man-made gas discharges such as the continuous arc discharges of Petrov in 1803 and Davy in 1809 [3]. Ongoing developments in electrical power sources and vacuum techniques resulted in better control of the discharge properties and in the discovery of more different types of gas discharges. Especially discharges created in glass tubes filled with gas at a low pressure (∼100 Pa) were studied extensively by Faraday in the 1830s [4] and later, during the 1870s by Crookes [5]. Their investigations resulted in a better understanding of the working principles and properties of gas discharges. By the end of the 19\textsuperscript{th} century it was clear that gas discharges were no longer exclusive to nature, humans could also make and control them.

Further studies showed that gas discharges consist of (partially) ionized gas, containing neutral and both positively and negatively charged particles. In 1928, Langmuir [6] introduced the word \textit{plasma} to describe the ionized gas that is created in a gas discharge. Without mentioning any further developments in plasma physics during the last 80 years, we conclude that nowadays gas discharges are known to consist of a collection of different particles, mainly electrons, ions, neutral atoms and molecules. These particles have a variety of interactions with each other, with surrounding wall materials and with electric and magnetic fields present in the discharge. This multitude of particles and interactions makes a gas discharge a complex system that is still not fully understood.

During the last two centuries, there have been numerous investigations on many different types of discharges. These studies were not only aimed at providing further insight in the fundamental processes involved in these discharges, they also investigated possible
1.1. Gas discharges

Applications. Presently, gas discharges are used in an increasing number of applications, ranging from plasma lighting to ozone production, and from etching of computer chips to production of solar cells.

The research presented in this thesis aims at making a contribution to the ongoing process of understanding the fundamental processes involved in gas discharges. This is expected to lead to an improvement of existing discharge applications and possibly to the development of new uses for gas discharges.

1.1.1 Steady-state discharges between parallel electrodes

As a further introduction, we will now look at an illustrative example of a gas discharge. Various modes of a dc discharge between parallel plate electrodes in a low-pressure gas environment will be introduced and described.

The different modes can be identified when looking at the voltage-current characteristic of such a discharge as schematically shown in figure 1.1. This curve can be experimentally reproduced by carefully changing the resistance in the circuit. This subdivision in different discharge modes is applicable for discharges at low pressures (typically $< 10^3$ Pa) with parallel, metal electrodes [7, 8]. The different regions in the $V - I$ characteristic can be
Chapter 1. General introduction

identified as follows:

1. **non-self-sustaining discharge (a-b):**
   When a low voltage is applied to an electrode gap containing neutral gas, an extremely small current ($<10^{-10}$ A) can be observed. This is caused by electrons in the gap created by external sources, for instance cosmic rays or a nearby UV lamp. These few electrons are accelerated towards the anode and cause a very small current. The applied voltage is not high enough to cause ionization of atoms by electron impact, which is observed for higher voltages as will be shown later. Since the discharge needs external sources for the creation of electrons it is **non-self-sustaining**; it will die out when the electron source is removed.

2. **Townsend discharge (b-c):**
   The Townsend discharge is also known as **dark discharge** since there is no appreciable light emission from such a discharge. Starting from the non-self-sustaining discharge, increasing of the applied voltage causes a transition to a **self-sustaining discharge**. The increasing voltage results in a higher electric field inside the discharge gap. The field becomes high enough such that electrons in the gap cause ionization of neutral atoms by electron impact. The result is a multiplication of electrons and ions in the discharge gap. At the cathode surface, new electrons can be emitted into the gas by secondary emission caused by ion impact. This provides a feedback mechanism which sustains a current through the discharge gap. The voltage marking the transition between a non-self-sustaining and a self-sustaining discharge is known as the **breakdown voltage**. In the case of a Townsend discharge, the applied voltage is just above the breakdown voltage and the current is limited to low values by a large external resistance $R$. The number of charged particles in the discharge gap is limited which gives no significant space charge effects and the applied potential in the gap is not disturbed. The $V − I$ characteristic for the Townsend discharge (b-c) is almost flat. This originates from the fact that a small increase in voltage leads to a higher electron multiplication in the gap, producing more electrons and ions, giving more secondary emission at the cathode, leading to even more electrons in the gap and a further multiplication of charges. This means that for a small increase in voltage, the current rises considerably.

3. **subnormal glow discharge (c-d):**
   A further increase of voltage leads to significant space charge effects in the discharge gap. Since there is a big difference between the mobilities of the ions and the electrons, there will be mainly positive space charge. This will concentrate in front of the cathode, creating a **cathode fall** region. The potential drop across the cathode fall is almost equal to the potential difference across the electrodes. In other words, the electric field in this region is higher than when the potential was uniformly distributed over the gap. Since the electron
multiplication increases for increasing electric field, there will be an enhancement of electron multiplication across the cathode fall. The result is that the total voltage needed to sustain the discharge can be reduced. Therefore, the $V - I$ curve shows a drop of voltage for increasing current. This discharge mode is often unstable and goes easily into a glow discharge.

4. **Glow discharge (d-e):**
In this discharge mode, the formation of the cathode fall region has completed; the voltage needed to sustain the discharge is at its minimum. A further increase in current does not lead to a decrease of voltage, but to spreading of the discharge over the electrode surfaces, keeping the current density constant. The glow discharge region ends when the whole surface of the electrodes is covered by the discharge and the current is further increased.

5. **Abnormal glow discharge (e-f):**
The electrodes are fully covered by the discharge and a further current increase leads to an increase of the cathode fall. The voltage across the electrodes rises sharply.

6. **Arc discharge (f-g):**
When the current is further increased a change into an arc discharge is observed. Different processes such as gas heating and thermionic electron emission from the electrodes become important. The result is that the voltage needed to sustain the discharge can be lowered substantially.

Which of these steady-state discharge modes will be formed depends on the external circuit; the voltage that is applied and the current that can be sustained. The voltage across the electrodes for such a system can be described by a load-line, $V = V_{dc} - IR$, where $V_{dc}$ is the dc voltage supplied, $I$ is the current and $R$ the external resistance. The crossing point of the load-line with the $V - I$ characteristic determines at which voltage and current, and in which mode, the discharge will operate. Figure 1.1 shows an example of determining the operating point of a discharge.

The $V - I$ characteristic in figure 1.1 is valid for discharges between parallel plates. However, the basic principles of the different discharge modes, such as cathode fall regions and glow-to-arc transitions, are also observed in low-pressure discharges with different geometries.

The discharge modes presented in this section are all steady-state discharges. This means that they will have the properties described above only after the discharge has stabilized. It is not always clear how this steady-state situation is reached. The evolution from a neutral gas into a steady-state discharge is known as plasma breakdown or discharge ignition and is the subject of this thesis. Especially, features of breakdown into a glow(like)
discharge will be studied. In the next section, plasma breakdown processes will be further introduced.

### 1.2 Plasma breakdown

Plasma breakdown is a fundamental process in gas discharge physics for the simple reason that every plasma has to start at some point in time. Since the variety of gas discharges is enormous, there is also a large range of different types of plasma breakdown. Gas discharges range from the very large scale such as lightning [9] and high altitude sprites [10] to the very small scale microdischarges such as plasma display panels [11] and the plasma needle [12,13]. For many plasmas, for instance discharge lamps and RF etching plasmas, the breakdown phase is only part of the start-up of the system. After breakdown the plasma stabilizes into a steady-state mode and the plasma is used for its intended purpose. For transient discharges, there exists no steady-state situation therefore the breakdown phase is a considerable part of the plasma lifetime. Examples of such systems include dielectric barrier discharges, lightning, and streamer and spark discharges. Also in repetitively pulsed discharges, the breakdown phase is important. The starting conditions for such discharges are determined by the remnants of previous discharges.

Investigations of discharge ignition are almost as old as studies of discharges themselves. Already in 1889, Paschen performed experiments studying the minimum potential difference that was needed to create a spark between two electrodes in a glass tube [14]. He found that this voltage depended on the type of gas, the pressure in the tube, \( p \), and the separation of the electrodes, \( d \). Moreover, the minimum breakdown voltage was a function of the product between pressure and gap distance, \( pd \). Figures showing the breakdown voltage as function of pressure times gap size are nowadays known as Paschen curves; an example is shown in figure 1.2. At that time, the breakdown voltages could be measured experimentally, but the underlying processes causing breakdown, were not properly understood.

Only 20 years later, in 1909, Townsend proposed a theory that could explain the observed breakdown phenomena, including the breakdown voltages measured in Paschen curves [16]. His theory was based on a description of microscopic processes such as ionization of atoms by electron impact, charge multiplication in electron avalanches, and secondary electron emission at the cathode by ion impact. Even nowadays, Townsend theory is commonly used to describe breakdown in low-pressure discharges. A more detailed introduction to Townsend breakdown theory can be found in chapter 2.

The Townsend theory provides an accurate description of breakdown for a large number of low-pressure gas discharges but the set of conditions to which it is applicable is limited.
1.2. Plasma breakdown

Figure 1.2: Paschen curves, showing the breakdown voltage, $V_{br}$, as function of $pd$ for different gases [8,15].

In the 1930s and 1940s, new observations of plasma breakdown under specific conditions showed features that were not in agreement with Townsend theory. Especially, discharges at high pressures and long gaps developed much faster than what could be explained using conventional Townsend breakdown theory. A new type of breakdown theory, known as streamer breakdown was developed by Loeb [17,18], Meek [19] and Raether [20]. In this new theory, space charge effects inside electron avalanches were taken into account, resulting in thin, weakly-ionized channels, known as streamers. Again, in chapter 2, more information on this breakdown theory can be found.

Since the early investigations of Paschen and Townsend and the subsequent development of streamer breakdown theory, many questions have remained unanswered and plasma breakdown is still an active field of research.

For instance, details of breakdown phases in low-pressure dc discharges with uniform fields, such as Townsend discharges [21–24] and glow discharges [25–29] are not fully understood and still being investigated. In pulsed, quasi-dc discharges memory effects, that is influences from previous discharges, play an important role and are studied in detail [30–32].

Further examples of investigations of plasma ignition include RF discharges [33–36], microwave sources [37] and microdischarges [38].

Breakdown phenomena are not only studied at low pressures, but also in high-pressure discharges there are many investigations. Especially streamer discharges are being studied
Chapter 1. General introduction

both experimentally and theoretically, for instance [39–44]. In these type of discharges, the propagation mechanisms and branching effects are not fully understood. Furthermore, special features such as ionization waves with velocities of about $10^7$–$10^8$ m/s, known as fast ionization waves, have been observed for specific conditions, but are only poorly understood [45, 46].

Not only of fundamental interest, but also in many plasma applications, breakdown is an important issue and the subject of research. For instance, (re)ignition is studied in high-intensity discharge lamps [47–50], fluorescent lamps [51–54], plasma display panels [11, 55] and air and water purification systems based on corona discharges [56, 57].

Recently, a special discharge mode in dielectric barrier discharges (DBDs) has been discovered [58, 59]. Under specific discharge conditions, the discharge appears as a diffuse glow covering the entire electrode surface. This in contrast to the normal DBD operating mode in which the discharge consists of many, short-lived discharge filaments. The exact reasons for this special mode are not fully understood, but it is clear that processes in the breakdown phase play a crucial role [60–65].

1.3 Scope of this thesis

The work presented in this thesis was aimed at obtaining a better understanding of the fundamental processes involved in breakdown of low-pressure discharges by experimental investigations.

Although plasma breakdown has been studied for more than a century, and the main concepts have been established, many aspects are still only poorly understood. It remains an active field of research, as shown with some examples in the previous section. Many of the existing studies on plasma breakdown, are modelling investigations. Additionally, most of the experimental investigations were limited to measuring breakdown voltages and discharge currents for various discharge configurations. There are very few reports of properly resolved measurements of plasma properties during breakdown development.

The lack of detailed, experimental studies of plasma properties during breakdown, is mainly due to the highly transient nature of the breakdown process. Measurement equipment capable of following the time evolution of the breakdown process have become available only during the last few years. Additionally, diagnostic techniques for directly measuring plasma properties, such as particle densities and electric fields are experimentally complex. An added complication is the fact that standard methods for measuring charged particle properties are inherently difficult during the breakdown phase because the particle densities are extremely low. In the research described in this thesis we use modern experimental techniques, capable of studying the evolution of the breakdown process with
adequate time-resolution.

Two types of discharges were studied; a pulsed discharge between parabolic, metal electrodes and a parallel-plate, low-pressure dielectric barrier discharge (LPDBD). The breakdown processes in these discharges were studied using different diagnostic techniques. For both discharges, the light emission from the discharge was recorded spatially and temporally resolved. Additionally, voltage and current were monitored during a discharge cycle. This allowed a detailed investigation of the spatial and transient behaviour of the breakdown process.

In parallel to these investigations, a new diagnostic technique was developed to study the time evolution of the electric field during breakdown. Based on measuring Stark effects in Rydberg levels of xenon atoms, it was possible to quantitatively measure electric field strengths during breakdown. The electric field distribution in the parabolic-electrode discharge was studied by this laser-spectroscopic technique.

For the LPDBD additional modelling investigations were performed comparing experimentally observed phenomena with model predictions in order to obtain a better insight in the details of this breakdown process. Similar studies were performed by Brok et al. [66,67] for the parabolic electrode arrangement used in this thesis, but as those investigations form part of a separate thesis dissertation, they are not included here. This thesis focuses on experimental investigations of plasma breakdown phenomena.

1.4 Thesis outline

In general, this thesis can be divided into three parts. In the first part, investigations on a pulsed discharge between parabolic, metal electrodes by light emission imaging experiments are presented. Secondly, the development of an electric field diagnostic based on Stark spectroscopy and the application of this technique to the pulsed discharge with parabolic electrodes are discussed. Finally, a second type of discharge, a low-pressure dielectric barrier discharge, is studied with both experimental and modelling techniques.

In more detail, chapter 2 gives a background on plasma breakdown processes. Basic phenomena are discussed and the Townsend breakdown theory is described in more detail. In the chapters 3 and 4 plasma emission imaging investigations of the pulsed discharge between parabolic electrodes are presented.

A background on the Stark effect is given in chapter 5. This chapter also discusses methods to theoretically calculate Stark effects, including the method of matrix diagonalization that is used in subsequent chapters. A detailed study of Stark effects in xenon atoms in neutral gas is presented in chapter 6. These investigations were intended to test the theoretical calculation method described in chapter 5 and investigate the feasibility of
the technique as an electric field diagnostic for plasma breakdown research.

Chapter 7 contains measurements of electric field strengths in ionization fronts during the breakdown phase of the discharge between parabolic electrodes. The electric field diagnostic was used to investigate breakdown phenomena in the same discharge configuration that was studied by different methods in chapters 3 and 4.

In chapter 8 a second type of discharge, a dielectric barrier discharge in low-pressure argon gas, is introduced. Both experimental and modelling investigations of this discharge are discussed.

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Chapter 2

Background on plasma breakdown

Abstract. The transformation process of a neutral gas into a conducting self-sustaining discharge is known as plasma breakdown or ignition. This chapter gives a theoretical background on plasma breakdown phenomena. It involves fundamental processes such as ionization of atoms by electron impact, charged particles moving in an electric field, charge multiplication in electron avalanches and secondary electron production at the cathode by ion impact. The Townsend breakdown theory describes how an interplay of these microscopical processes can result in the ignition of a discharge between metal, parallel plates in a low-pressure environment. Finally, breakdown in discharges at high pressures and large gaps can not be properly described by the Townsend theory. For these type of discharges streamer and leader breakdown mechanisms are more appropriate. These breakdown theories are briefly introduced.
2.1 Fundamental processes

This section introduces fundamental processes involved in plasma breakdown. In section 2.2 the Townsend breakdown theory is described, while in section 2.3 the main ideas of streamer and leader breakdown are briefly introduced.

2.1.1 Electron avalanches

When a charged particle is placed in an electric field, it will experience an accelerating force. This results in a motion in the direction parallel to the electric field. However, except for very low pressures, in gases (and weakly ionized plasmas) particles collide very frequently with other particles, mainly neutral atoms/molecules. Such collisions change the direction of the moving particle sharply and in a random way, thereby loosing its directed motion. In between such collisions the charged particles are again accelerated by the electric field. Therefore, the motion of charged particles in an electric field is a superposition of a directed motion parallel to the electric field and a random motion. The average, directed motion as a result of an electric field is known as drift. The drift velocity of a particle, \( v_{d,i} \), can be described by:

\[
v_{d,i} = \pm \mu_i F,
\]

where the \( \pm \) sign indicates whether the drifting particle, \( i \), is positively or negatively charged, \( F \) is the electric field and \( \mu_i \) the mobility of particle \( i \). It is clear that particles with opposite charge will drift in opposite directions. The relation between the electric field, \( F \), and the drift velocity is not completely linear since the mobility is a function of electric field strength. It should be noted that since electrons are much lighter than noble gas ions, the mobilities (and drift velocities) of electrons are higher. As an example, figure 2.1 shows the drift velocities of electrons and ions in argon gas.

The fundamental element of the breakdown process is the electron avalanche. Electrons drifting in an electric field towards the anode gain energy from this field and cause ionization of atoms by collisions. This results in a multiplication of electrons and ions. In the 1930s, Raether was the first to visualize an electron avalanche using a cloud chamber experiment \[4\]. Figure 2.2 shows an electron avalanche in a cloud track photograph taken by Raether \[4\].

Electron avalanches evolve not only in time, but also in space, along the direction of the electric field. Often it is most convenient to express the rate of ionization not in terms of ionization frequency, but as an ionization coefficient, \( \alpha \), which is the number of ionization events by electron impact per unit length along the direction of the electric field. The coefficient \( \alpha \) is also known as Townsend’s (first) ionization coefficient.
2.1. Fundamental processes

**Figure 2.1:** Drift velocities of electrons and ions in argon as function of the reduced electric field. The data for the ion drift velocity are taken from [1]. For the electron drift velocities, data from [2] for $E/p < 0.75 \text{ V m}^{-1} \text{ Pa}^{-1}$ and calculations from [3] for $E/p > 0.75 \text{ V m}^{-1} \text{ Pa}^{-1}$ are presented.

**Figure 2.2:** Cloud track photograph visualizing an electron avalanche. The cathode is located at the left, the anode at the right. (by H. Raether [4]).
The ionization coefficient divided by the pressure, $\alpha/p$, is a function of the reduced electric field strength, $E/p$. Additionally, it depends on the type of gas, as shown on figure 2.3. Generally, the ionization coefficient, $\alpha$, can be described by the empirical formula:

$$\frac{\alpha}{p} = A \exp \left(-\frac{Bp}{E}\right),$$  \hspace{1cm} (2.2)

where the constants $A$ and $B$ are gas-specific and are derived from experiments.

For typical discharge conditions studied in this thesis, the applied reduced electric field is around 225 V m$^{-1}$ Pa$^{-1}$ (300 V cm$^{-1}$ Torr$^{-1}$), and the argon pressure is 465 Pa (3.5 Torr). Figure 2.3 shows that this situation corresponds to an ionization coefficient, $\alpha$, of about 2300 m$^{-1}$. This means that when an electron avalanche crosses the discharge gap of 3.3 mm, there will be on average 7.6 ionization events per electron starting from the cathode, resulting in about 200 electrons near the anode.

### 2.1.2 Secondary electron emission

In gas discharges there are several processes that can release electrons from the surfaces of metal electrodes. To extract an electron from a metal surface, a certain amount of energy, known as the *work function*, is needed. Values for the work function depend (strongly)
on parameters such as the type of material and the state of the surface, including surface contamination and roughness.

There are several mechanisms that can provide sufficient energy to release electrons from metal surfaces [6]. The first is thermionic emission in which the electrode material is heated to high temperatures of hundreds or thousands Kelvin. Some of the electrons in the metal gain sufficient energy to escape from the potential well of the metal. A second mechanism for electron release from metals is field emission. A strong electric field near the metal surface changes the potential well into a potential barrier with finite width. This gives the electrons in the metal a chance to tunnel through this barrier and escape the metal. Fields needed to obtain a significant flux of electrons are in the order of $10^8$ V/m. A combination of both mechanisms, high electrode temperatures and strong electric fields, is also possible and is known as thermionic field emission.

Finally, a third mechanism for electron release from metal surfaces is secondary electron emission. In this process a particle impacts on the electrode surface and an electron is released. Secondary electron emission can be caused by various types of particles such as positive ions, excited atoms, electrons and photons. The secondary electron emission is normally characterized by the coefficient $\gamma$, which specifies the number of secondary electrons per incident particle.

For most conditions in low-pressure gas discharges, the ion-impact electron emission is the dominant secondary emission mechanism. In this situation, the electric field of the ion can release electrons from the metal by Auger processes. When an ion approaches the metal surface within distances of atomic dimensions, the electric field of the ion can transform the potential well of the metal into a shallow barrier with finite width. An electron from the metal will tunnel through this barrier and neutralize the ion. When the energy released in this process is more than the work function of the metal, a second electron can be released from the metal.

Not only ions can cause secondary electron emission, but also metastable atoms. The mechanism for this process is very similar to that for ions. The metastable atom is ionized close to the surface and releases a secondary electron by Auger processes.

Secondary electron emission from metals as a result of photons is also possible. When the energy of the photon ($h\nu$) is larger than the work function of the metal, an electron can be released. The $\gamma$ coefficients for this process range from $10^{-3}$ up to $10^{-1}$ for UV/VUV photons. Therefore this process can have a significant effect on plasma breakdown under specific conditions. However, for most breakdown conditions, secondary electron emission by ions is more important than emission by metastables and photons.

Secondary electron emission from dielectric materials is also observed. The processes involved are again similar to those observed in metals. However, the fact that dielectric
surfaces can be charged up complicates the situation. The main characteristics of the 
processes releasing electrons from the dielectric material itself remain the same as for 
metals. However, additional processes that remove charges which are on the surface of the 
dielectric are not fully understood, but can play an important role [7–9].

In most low-pressure gas discharges, and all discharges discussed in this thesis, the 
electrodes are not heated and also no very strong electric fields are present close to the 
electrodes. Therefore, for these cases secondary electron emission by ion impact is the 
dominant process for the release of electrons from metal electrodes.

As noted before, the values for the secondary electron emission depend strongly on the 
surface condition of the electrode. It is well-known that metal surfaces that are cleaned 
by a heat treatment give different secondary electron yields than ‘dirty’ electrodes without 
treatment [10]. Furthermore, it is difficult to identify and separate the different processes 
leading to secondary electron emission [10]. In almost all experiments there are more 
processes occurring simultaneously. Therefore, in practise it is often difficult to identify 
the relevant secondary emission processes and to give a precise value for each contribution 
to the total secondary electron emission coefficient, $\gamma$.

### 2.2 Townsend theory

#### 2.2.1 Breakdown criterion

In the beginning of the twentieth century, Townsend developed a theory that described 
the breakdown process of a discharge at a pressure, $p$, between parallel plate electrodes, 
separated by a distance $d$ [11]. This theory gives an accurate description of the breakdown 
process for a set of conditions in which the product $pd$ is limited to a range of about 
0.1–100 Pa m (0.075–75 Torr cm) and the electrode geometries are relatively simple. The 
microscopical processes, introduced in the previous section, form the fundamental building 
blocks of the Townsend breakdown theory.

In the following discussion of the Townsend theory we assume uniform electric fields 
between parallel, metal electrodes. In this situation, a single electron that starts at the 
cathode will cause an electron avalanche towards the anode. The number of electrons in 
this electron avalanche, $N_e$, is described by:

$$N_e = e^{\alpha x},$$

(2.3)

with $\alpha$ the ionization coefficient, as described in section 2.1.1, and $x$ the distance from the 
cathode. The number of ions, $N_i$, produced in an electron avalanche by ionizing electron
The developing electron avalanche leaves these ions behind in the discharge gap, since the drift velocity of the electrons is much higher than that of the ions. By the time the electrons reach the anode, the ions produced in the avalanche have hardly started their drift process. Nevertheless, the ions will eventually drift towards the cathode and when they do reach the cathode surface there is a certain probability, $\gamma$, that a secondary electron is emitted from the cathode surface. For a discharge gap with a width of $d$, the number of secondary electrons is:

$$N_{e,sec} = \gamma (e^{\alpha d} - 1).$$

The new, secondary electrons will each start a new electron avalanche, producing new electrons and ions in the discharge gap. When other production and loss processes for charged particles, such as photoionization, field emission and electron attachment are neglected, a criterion for self-sustenance of the electron avalanches can be derived. The situation is exactly self-sustaining when the ions resulting from a single electron avalanche produce a single electron by secondary emission at the cathode. This situation corresponds to:

$$\gamma (e^{\alpha d} - 1) = 1,$$

which is known as the breakdown criterion.

With the Townsend theory it is possible to explain Paschen’s law on breakdown voltages [12]. For the case of uniform electric fields in a parallel-plate discharge, the electric field is simply the voltage divided by the gap distance, $E = V/d$. Together with the empirical equation for $\alpha$ (equation 2.2), this can be substituted in the breakdown criterion to find the breakdown voltage, $V_{br}$; that is the voltage needed to achieve breakdown. Firstly, the breakdown criterion (equation 2.6) can be rewritten as:

$$e^{\alpha d} = \frac{1}{\gamma} + 1.$$

Next, equation 2.2 for $\alpha$ and $E = V_{br}/d$ for the electric field are substituted:

$$d = \frac{\exp \left( Bpd/V_{br} \right)}{Ap} \ln \left[ \frac{1}{\gamma} + 1 \right],$$

which can be rearranged to find an expression for the breakdown voltage,

$$V_{br} = \frac{Bpd}{\ln \left( \frac{Apd}{\ln(1+1/\gamma)} \right)}.$$

Evaluating this expression shows that for high $pd$, $V_{br}$ increases quasi-linear with $pd$, while for low $pd$ it also increases sharply. Therefore, the curve of breakdown voltage as function
Chapter 2. Background on plasma breakdown

of $pd$, shows a minimum. This minimum breakdown voltage, $V_{br(min)}$, can be derived by differentiating equation 2.9 and setting it to zero. The result is:

$$V_{br(min)} = 2.718 \frac{B}{A} \ln \left(1 + \frac{1}{\gamma}\right).$$

(2.10)

With these equations for the breakdown voltage, it is possible to calculate Paschen curves and predict the minimum breakdown voltage, provided that the coefficients $A$, $B$ and $\gamma$ are known accurately. The agreement between calculations based on the Townsend theory and experimental data proved to be very good.

2.2.2 Non-self-sustaining discharges

The breakdown criterion in equation 2.6 shows that there are two possibilities for breakdown development when a voltage is applied to an electrode arrangement. The first, when $\gamma(e^{\alpha d} - 1) < 1$, is briefly discussed in this section, the second $\gamma(e^{\alpha d} - 1) \geq 1$ is the topic of the next section.

When the breakdown criterion is not fulfilled, $\gamma(e^{\alpha d} - 1) < 1$, it is clear that electron avalanches can not sustain themselves and therefore will die out. However, if there is a continuous, external source of electrons it is possible to sustain the electron avalanches in the discharge gap and reach a steady-state situation. For instance, an external UV light source can liberate electrons from the cathode surface by photo-electric effects.

In a steady-state situation, we can define $n_0$ as the rate of electrons liberated from the cathode by an external source, $n$ as the rate of electrons arriving at the anode and $n'_0$ as the total rate of electrons emitted from the cathode; that is both by external sources, $n_0$, and by secondary electron emission by ion impact, $\gamma(n - n'_0)$. This gives

$$n'_0 = n_0 + \gamma(n - n'_0) = \frac{n_0 + \gamma n}{1 + \gamma},$$

(2.11)

with

$$n = n'_0 e^{\alpha d},$$

(2.12)

which results in a steady-state situation of

$$\frac{n}{n_0} = \frac{i}{i_0} = \frac{e^{\alpha d}}{1 - \gamma(e^{\alpha d} - 1)},$$

(2.13)

where $n$ and $n_0$ represent rates of electrons and therefore can be replaced by currents $i$ and $i_0$ respectively. It is important to realize that this equation only gives a solution below the breakdown criterion of $\gamma(e^{\alpha d} - 1) < 1$. 

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As soon as the external source of electrons, \( n_0 \), is removed, the discharge current will vanish, therefore the discharge is non-self-sustaining. This type of steady-state discharge was already introduced in figure 1.1 in the previous chapter.

The sensitivity of the discharge current on the incident radiation, as an external source of electrons, is used in a Geiger radiation counter. Also other external electron emission processes can be studied using this type of discharge. Additionally, these types of discharges are often used for determining breakdown coefficients \( \alpha \) and \( \gamma \). By choosing the operating conditions carefully and accurately measuring the current as function of electrode separation, these coefficients can be derived [13–16].

### 2.2.3 Self-sustaining discharges

When \( \gamma(e^{\alpha d} - 1) \geq 1 \), for instance by a small increase of the applied voltage, the number of electrons in the gap keeps increasing over time. Each generation of electron avalanches will contain more electrons than the previous one. The result is an increasing flux of electrons, and therefore an increasing current through the discharge gap. This current keeps rising until it is limited by, for instance, an external resistance. The development of this type of discharge does not depend on external sources of electrons, therefore it is self-sustaining.

**Townsend discharge**

A special situation occurs when the breakdown criterion is exactly met, \( \gamma(e^{\alpha d} - 1) = 1 \). Such a discharge is started with a voltage, just above the breakdown voltage and a very large resistance in series with the discharge. This resistance limits the developing discharge current at very low values between about \( 10^{-10} \) and \( 10^{-5} \) A. The voltage across the electrodes drops a little bit to exactly the breakdown voltage, due to the current flowing through the resistance. In this situation, each electron emitted from the cathode, starts an electron avalanche which produces exactly one secondary electron. This type of discharge was briefly introduced the previous chapter and is known as a Townsend or dark discharge. The charge densities produced in the electron avalanches in such a discharge are very limited. They are not large enough to cause significant space charge fields and affect the electric field inside the gap.

The Townsend theory proved to be very successful in describing the general phenomena observed in these type of discharges. However, some details, for instance current oscillations in certain low-current discharges, are still not fully understood [17–19].
Chapter 2. Background on plasma breakdown

Glow discharge

In situations in which $\gamma(e^{ad} - 1) > 1$, a different type of discharge will develop when a voltage is applied to the electrodes. For most discharges in the range $pd = 0.1–100$ Pa m (0.075–75 Torr cm), the breakdown process transforms the neutral gas into a glow discharge. The glow discharge has been briefly introduced in the previous chapter of this thesis. This type of discharge has been studied extensively in the past and more details can be found in many text books, for instance [6,20]. An interesting property of glow discharges is the characteristic structure of the light emission and the potential distribution in the electrode gap, as schematically shown in figure 2.4. Most of the potential drop across the electrodes is concentrated in a layer close to the cathode, known as the cathode fall. Moving further towards the anode, a bright region with (almost) constant potential, known as the negative glow, and the Faraday dark space are observed. Next, if the discharge gap is long enough, a luminous positive column region is formed in which there is a slowly increasing potential, resulting in a constant, low electric field. Finally, in front of the anode, there is a small potential jump.

Describing breakdown of a neutral gas into a glow discharge, implies that the breakdown process needs to change the potential distribution in the discharge gap from the initial, linear profile from the applied voltage, to the characteristic structure of a glow discharge. This process can be described as follows. At the start of the ignition phase there are electron avalanches resulting in charge multiplication in the discharge gap. Since the voltage is above 24
2.2. Townsend theory

the breakdown voltage, these electron avalanches get stronger each generation. Because the electrons drift much faster than the ions, there will be mainly ions present inside the discharge gap. Therefore, the number of ions in the discharge gap increases with time. At some point, the density of these charges becomes so large that they cause significant electric fields as a result of space charge effects. Because the ion production in electron avalanches is strongest close to the anode, most positive space charge will be found here. The result is a modification of the electric field distribution inside the gap. The original Townsend theory does not take space charge effects into account, therefore it is not able to fully explain the breakdown behaviour. However, the concepts used in Townsend theory remain the same for the situation of breakdown into a glow discharge and these concepts can be used to obtain a qualitative picture of the breakdown process.

The main effect of the space charges is a change of the potential distribution in the gap and therefore a change of the local electric field strengths. Due to the positive space charge in front of the anode, the anode potential will be extended into the gap and there will be field enhancement between the cathode and the space charge region, while there is a decrease of electric field close to the anode. The result is that the electric field in the gap is no longer uniform. The ionization coefficient, $\alpha$, is a function of reduced electric field and therefore no longer constant, but a function of position and time. This complicates the calculation of the number of electrons in an electron avalanche, $N_e$, which is now described by

$$N_e(x, t) = N_0(t) \exp \left( \int_0^d \alpha(x, t) dx \right),$$

(2.14)

where $N_0(t)$ is the number of secondary electrons emitted by the cathode. The electron avalanche development now depends on the local electric field strength in the gap (through the $\alpha$ coefficient). At the same time, the local electric field strength is influenced by the charges produced in previous electron avalanches. The result is a spatially and temporally non-uniform situation, which is difficult to describe quantitatively without using sophisticated modelling methods.

On the other hand, it is possible to obtain a qualitative picture of what will happen during the breakdown process when space charge effects start to play a role. This is schematically indicated in figure 2.5. In the gap, between the cathode and the space charge region, there is an enhanced electric field leading to a higher $\alpha$ coefficient, and therefore stronger electron avalanches. However, these avalanches will only be stronger up to the space charge region, because in between this region and the anode the electric field is decreased and electron multiplication will go down. The space charge region is (approximately) at the same potential as the anode, which means that the anode potential is extended into the discharge gap by the space charge region. The largest part of the
potential difference across the electrodes is now located in the region between the space charge and the cathode. This is the start of the formation of a cathode fall region. During the further development of the breakdown, additional space charges will be added on the cathode-side of the space charge region, extending it further into the gap. Between the space charge region and the anode, there will be many charges, but low electric fields, so no significant charge multiplication. The process of extending the space charge region towards the cathode will continue, further compressing the potential drop in a region close to the cathode. Eventually, the space charge region will reach the cathode surface and the potential across the gap will have taken the characteristic structure of a glow discharge. The potential drop across the cathode layer is approximately equal to the minimum breakdown voltage, $V_{br}(\text{min})$. The thickness of the cathode layer multiplied by the pressure is such that it is close to the $pd$-value for which $V_{br}(\text{min})$ is achieved.

It can be concluded that the Townsend theory provides the basic building blocks for describing breakdown processes, but a quantitative, time- and space-resolved description is non-trivial, even for simple discharge geometries.

Finally, some comments on the time scales involved in this breakdown process. The development of a single electron avalanche is determined by the electron drift velocity, which is typically $10^5$–$10^6$ m/s as shown in figure 2.1. The ion drift velocities are much smaller, typically 50-500 m/s. The full breakdown process needs many generations of avalanches, initiated by secondary electron impact by ions. Therefore, the time needed for ions to drift towards the cathode is the dominant time scale. For breakdown into a glow discharge, the full breakdown process takes typically $10^{-5}$–$10^{-3}$ seconds.
2.3 Streamer and leader breakdown

The breakdown theory described in the previous section is commonly known as the Townsend breakdown mechanism. It is characterized by the continuous development of successive electron avalanches between electrodes by secondary emission processes. During the last century, the Townsend theory has been very successful in explaining breakdown phenomena under various discharge conditions. For instance, explanation of Paschen’s breakdown curves, as discussed in the previous section, and effects of electronegative gases and cathode materials on the breakdown voltage.

However, in the 1930s and 40s, with the ongoing development of experimental equipment for studying the time-development of transient breakdown processes, observations of breakdown phenomena were made that did not fit in the Townsend theory. For instance, Raether’s investigations of developing electron avalanches by cloud chamber experiments [4] showed breakdown features too fast to be explained by Townsend mechanism. These observations were mainly made for discharges with high values of $pd (>10^3$ Pa m); that is high pressures and large gaps. Loeb [21,22], Meek [23] and Raether [24] developed the fundamentals of a new breakdown theory that could explain the observed, high-speed breakdown features. The basis of this new theory, known as streamer breakdown theory, is taking into account space charge effects of a single electron avalanche. In the Townsend theory these space charge effects within electron avalanches are neglected.

In the streamer theory, the breakdown process still starts with the development of an electron avalanche. However, the multiplication of charges in this avalanche is so large that the space charge in the avalanche head starts to modify the applied electric field, before the avalanche can reach the anode. A avalanche-to-streamer transition can be observed in the discharge gap. The space charge field in the avalanche head can be calculated, assuming complete separation between positive and negative charges. The space charge field, $E_s$, produced by a number of charges, $Q$, in a sphere of radius $r$ is [25]:

$$E_s = \frac{Qe}{4\pi \varepsilon r^2},$$

(2.15)

where $e$ is the elementary charge and $\varepsilon$ the vacuum permittivity. The radius, $r$, of the space charge region, that is the avalanche head, can be described by [6,25]

$$r = \sqrt{4Dt},$$

(2.16)

with $D$ the diffusion coefficient and $t$ the time after the start of the avalanche.

By calculating the space charge formation in an electron avalanche and comparing it to the applied electric field, Meek defined a criterion for streamer formation. In simplified
form the avalanche-to-streamer transition occurs when [6]:

$$\exp \left( \int \alpha' \, dx \right) \approx 10^8,$$

(2.17)

where $x$ is the distance travelled by an electron avalanche and $\alpha'$ is the net ionization coefficient, defined as the difference between the ionization coefficient, $\alpha$, and the attachment coefficient, $\eta$. The exact values of the streamer criterion depend on the details of the discharge. This criterion represents the fact that the applied electric field has to be high enough ($\alpha'$) and that the gap has to be long enough.

As soon as Meek’s criterion is fulfilled, a streamer starts to develop. This is a thin weakly-ionized channel that develops between the electrodes at high velocities. Space charge effects in the streamer head modify the electric field distribution in the gap creating an environment for high charge multiplication. Secondary electron avalanches develop in front of the head of the streamer. This makes the streamer develop extremely fast; its speed is characterized by the electron drift velocity, while the ions produced in the streamer have no time to drift to the cathode and cause secondary electron emission. Typical streamer velocities are in the order of $10^6$ m/s, much faster than Townsend breakdown phenomena ($\sim 10^3$ m/s) Many aspects of the streamer breakdown mechanism such as propagation mechanisms and branching are still unknown and it is an active field of research [25–29].

Very large scale discharges, such as lightning can be described by a third type of breakdown mechanism, known as leader breakdown [30–32]. A thin, highly-ionized channel propagates along a path prepared by streamer discharges. When the streamer channel has crossed the discharge gap a return strike occurs, transforming the discharge into a spark.

It should be noted that the type of discharges studied in this thesis all have $pd$ values below 100 Pa m (75 Torr cm) and therefore a Townsend-like breakdown mechanism can be expected.

References


2.3. References


Chapter 3

Plasma emission imaging of a low-pressure argon breakdown

Abstract. Breakdown in a low-pressure argon discharge was investigated experimentally by time-resolved plasma emission imaging with an intensified charge coupled device (ICCD) camera. Basic features of plasma breakdown, such as light emission in front of the anode and crossing of an emission front from anode to cathode were observed. The observations were in agreement with the standard Townsend theory on breakdown into a glow discharge. In addition, a pre-breakdown light flash was observed. This light flash, which occurred at a voltage below the static breakdown voltage, was thought to originate from charges from previous discharges deposited on dielectric surfaces close to the discharge gap. Finally, the time delay as function of the afterglow period of the discharge was measured. The densities of charged particles at the time of ignition influenced the measured time delay. For afterglow periods increasing from 1 to 20 ms, volume recombination of charged particles caused a sharp increase in the time delay from 1 to 9 $\mu$s. For longer afterglow periods (20–200 ms), diffusion became increasingly important and caused a slower increase in time delay (9 to 11 $\mu$s).

3.1 Introduction

Plasma breakdown, the process by which a neutral gas becomes ionized and electrically conducting, is one of the fundamental processes in plasma physics. It occurs in every plasma at the time of ignition and is a highly transient process that involves multiplication of electrons in avalanches and moving ionization fronts. Although plasma breakdown has been studied for more than a hundred years, many aspects are poorly understood and it is still an active area of research.

In 1889, Paschen [1] was the first to predict breakdown voltages based on electrode separation and gas pressure. In the beginning of the twentieth century, Townsend [2] developed a theory describing the breakdown process of low current, low pressure ($\sim 10^2-10^4$ Pa) discharges in uniform electric fields. He introduced the concepts of electron multiplication in avalanches, crossing of ionization fronts and formation of multiple avalanches by secondary electron emission at the cathode. In the last few decades, new breakdown mechanisms, such as streamer [3,4] and leader processes [5,6], were introduced to explain certain high pressure ($\sim 10^4-10^5$ Pa) breakdown phenomena. However, the Townsend theory is still the most suitable theory to describe low-pressure breakdown phenomena. Although many cases of plasma breakdown can be generally described by existing breakdown theories, some of the details of breakdown remain unclear. Furthermore, systems with complex geometries, high pressures or non-uniform electric fields are not fully described by the existing theories.

In many plasma applications breakdown is an important issue that receives considerable attention. Some examples are ignition of fluorescent lamps and high intensity discharge lamps [7,8], gas and water cleaning systems based on corona discharges [9,10] and ozone generation with dielectric barrier discharges [11]. A better understanding of the elementary processes in plasma breakdown is necessary to improve these applications and possibly find new ones.

As mentioned earlier, plasma breakdown into a glow discharge in a uniform electric field is reasonably well understood and can be described with the standard Townsend breakdown theory. More details on the Townsend theory can be found in chapter 2 and reference [12]. However, details of the breakdown process, such as memory effects [13,14] and electrode surface processes [15] remain unresolved. The main limitation in experimental studies has always been the highly transient character of plasma breakdown and the limited time resolution of available experimental techniques. With the ongoing development of measurement equipment such as cameras, oscilloscopes and lasers, more suitable diagnostics have become available and more detailed measurements on plasma breakdown phenomena are now possible.

The aim of the research described in this chapter was to study the fundamental processes
that occur during breakdown at low pressure. We used a simple electrode geometry and observed breakdown in argon when voltage pulses were applied to the electrodes. Current and voltage were measured using standard techniques and plasma emission was measured by imaging the discharge region onto an intensified charge-coupled device (ICCD) camera. Such measurements were made for a variety of discharge conditions. The attention of the experiments was focussed on a single representative discharge condition, of which detailed, time-resolved measurements were made. The results of these detailed experiments were qualitatively compared with a one-dimensional model based on the theory describing low-pressure breakdown into a glow discharge.

This chapter is structured as follows. Section 3.2 contains a description of the discharge apparatus and the ICCD measurement system. In section 3.3, we present the results of the emission imaging experiments and show the different features observed. We also discuss the experimental results and present a simple model of the breakdown process. Section 3.4 contains results and discussion of time delay measurements on our discharge. Section 3.5 is a brief summary.

3.2 Experimental arrangement

The experimental arrangement consisted of a discharge apparatus, which is described in section 3.2.1, and an ICCD measurement system, described in section 3.2.2.

3.2.1 Discharge apparatus

A schematic diagram of the discharge apparatus is shown in figure 3.1. It consisted of two identical metal electrodes in a vacuum vessel. The stainless-steel electrodes were cylindrically symmetric and parabolically shaped. The curvature of the tip was equivalent to a radius of 4 mm. The shape of the electrodes was chosen to be parabolic because this created a discharge that was stable and reproducible every discharge cycle, both in space and time. The surfaces of the electrodes were polished with fine-grained sand paper giving a surface roughness of about 1 \( \mu m \). The electrodes were mounted on an insulating polyester stage, which positioned the electrodes in the centre of the vacuum vessel. The distance between the electrodes was fixed at 3.3 mm for all measurements. The vacuum system consisted of a vacuum vessel, a turbomolecular pump and a rotary vane pump. With this system, a base pressure of \( 10^{-4} \) Pa (\( 10^{-6} \) Torr) could be obtained. A continuous flow of argon gas (purity 99.999\%) was directed through the system. A needle valve controlling the gas flow maintained the pressure in the vessel. Discharges with pressures ranging from 100 to 1000 Pa were studied.
To create a repetitive quasi-dc breakdown we applied a pulsed voltage to the electrodes. A waveform from a function generator was amplified in a power amplifier (Spitzenberger EV600/CuG) to an amplitude of several hundreds of volts. This voltage was applied to the electrode arrangement to create a discharge. The pulse length was kept short to avoid heating of the electrodes and background gas in the glow phase of the discharge. The rise time, length, amplitude and repetition frequency of the pulse could be varied. Typical values were a 10–90% rise time of 25 µs, total length of 100 µs and amplitude of 350 V. Discharges with repetition frequencies between 5 and 5000 Hz and voltage rise times between 2 and 200 µs were studied. The applied voltage amplitude was about 15% above the static breakdown voltage.

The electrode voltage and the discharge current were simultaneously recorded on an oscilloscope. The voltage across the electrode gap was measured using a 100:1 potential divider. The current in the discharge was monitored by measuring the voltage across a 100 Ω resistor connected in series with the electrode arrangement.

The electric field distribution in the discharge area was non-homogeneous due to the electrode geometry. Figure 3.2 shows the results of a calculation of the electric field distribution in the discharge arrangement, when a voltage of 350 V was applied to the top electrode. This image represents a stationary situation, which occurred before the start of the breakdown process. Figure 3.2 shows that the electric field was rather non-homogeneous, mainly on the sides of the electrodes. However, in the discharge gap, between the tips of the electrodes, the electric field was fairly homogeneous. The radial component of the field
3.2. Experimental arrangement

Figure 3.2: Calculated electric field distribution. The top electrode is at a potential of 350 V. The bottom electrode is grounded. The solid and dotted lines indicate equipotential lines, while the arrows represent the electric field vectors. The direction and size of the arrows indicate the direction and magnitude of the electric field at that point. The axes' labels indicate distances in millimetres.

in this region was less than 15% of the axial component.

3.2.2 ICCD measurement system

The ICCD optical system consisted of an imaging lens and an ICCD camera (Andor Technology DH534). Two different lens systems were used to image the discharge region onto the ICCD camera. The first system gave an overview of the entire discharge region, the second an enlarged image of the discharge gap. Both arrangements are shown in figure 3.3. In the first arrangement, a camera macro lens (Sigma 50 mm f2.8) was used to image the entire electrode arrangement onto the ICCD camera. The optical reduction factor of this system was approximately 5:1. In the second arrangement, attention was focused on the discharge gap. A lens with a 200 mm focal length was used to obtain a 1:1 image of the discharge gap.

The ICCD camera had an active area of 13.3×13.3 mm², which consisted of 1024×1024 pixels. The intensifier had a minimum gate duration of 3 ns and a gain of up to 1000 counts per photoelectron. The photocathode was sensitive to all light in the wavelength range from 200 to 850 nm. For this experiment, the transmission of the chamber window meant that
emission in the range 350–850 nm was detected. The camera gating was synchronized with the discharge voltage pulse by means of a delay generator. The delay between the start of the voltage pulse and the gating of the camera could be varied to observe different time intervals in the breakdown cycle.

3.3 Imaging of the breakdown process

In this section the results of our emission imaging experiments are presented and discussed. We investigated discharges with repetition frequencies between 5 and 5000 Hz and rise times from 2 to 200 µs. The observed features of plasma breakdown, which will be further discussed in this section, were qualitatively the same for all of these discharge conditions, the quantitative details did change slightly. A higher repetition frequency lead to an earlier start of the breakdown process, as will be discussed in section 3.4. Changing the voltage rise time also changed the timing of the start of the breakdown process, but it did not significantly affect the properties of the actual process itself. For all rise times investigated, the breakdown process started at about the same applied voltage.

For a qualitative understanding of the breakdown phenomena, we present the results of the experiments studying a single representative discharge condition. The results of the
3.3. Imaging of the breakdown process

Section 3.3.1 contains an overview of an entire breakdown cycle. In section 3.3.2, observations of a pre-breakdown light flash are presented and discussed. Section 3.3.3 focuses on the crossing of a light front in the discharge gap.

3.3.1 Breakdown cycle overview

Breakdown phenomena were studied in a repetitive quasi-dc discharge at a pressure of 465 Pa (3.5 Torr). A positive, pulsed voltage was applied to the top electrode of the discharge apparatus, while the bottom electrode was grounded. Measurements of the discharge voltage and current are shown in figure 3.4. The applied voltage waveform had a 10–90% rise time of 25 µs, total length of 100 µs and amplitude of 350 V. The pulse repetition frequency was 500 Hz.

In a separate experiment, we measured the static breakdown voltage of our discharge to be 300 V, by observing breakdown with minimum voltage using a dc power supply. This implies that the voltage waveform used for our experiments has a maximum which is about 15% above the static breakdown voltage. The actual breakdown voltage of the system, that is the minimum voltage needed to sustain the discharge, was about 10 V higher than the static breakdown voltage. The imaging experiments were performed with this moderate overvoltage in order to minimize statistical jitter and ensure a stable breakdown event for every voltage pulse.

Figure 3.4 shows that in our discharge the plasma current started to rise about 35 µs after the start of the voltage pulse. In the next 35 µs, the current increased to a maximum...
value of approximately 25 mA. At this point the voltage was decreased and the current dropped steeply.

The 5:1 optical system (figure 3.3(a)) was used to measure ICCD images with a long integration time (5 µs). These images are shown in figure 3.5 and give an overview of the plasma emission during a complete breakdown cycle. From this set of images, several features of plasma breakdown could be identified. Firstly, between 10 and 20 µs when the voltage was still rising, a weak pre-breakdown light flash occurred near the anode. Secondly, after 30 µs when the voltage had reached its maximum, there was weak light emission near the anode again. In the next image (35 µs) the light emission had crossed the discharge gap and covered part of the cathode surface. As noted above, the discharge current started to increase at this time. In the next phase, the plasma emission covered the entire cathode surface and stabilized. From 60 to 70 µs the plasma emission pattern was stable. After 70 µs when the voltage was decreased, the current dropped steeply and the plasma died out (80-100 µs).

We were interested in the initial stages of plasma breakdown and hence focused on the first part of the discharge. For this reason, further experiments were performed investigating the pre-breakdown light flash (10 µs) and the crossing of the electrode gap by a light front (30–35 µs) in more detail. These subjects are discussed in the next two sections.

### 3.3.2 Pre-breakdown light flash

The pre-breakdown light flash, observed after approximately 10 µs, was investigated in more detail. The 5:1 optical system (figure 3.3(a)) was used to measure ICCD images with a short exposure time of 100 ns. Figure 3.6 shows the results of these measurements.

A light flash appeared around the tip of the anode after 6 µs. During the next 10 µs, the light front moved away from the anode and died out after approximately 18 µs. During this light flash, the applied voltage was rising from 70 to 210 V and hence was always below the static breakdown voltage.

Further experiments were performed to study the behaviour of the pre-breakdown light flash under different discharge conditions. Voltage rise times were varied from 2.5 to 50 µs and pulse repetition frequencies from 50 to 500 Hz. Further, the polarity of the applied voltage was inverted and finally a small negative voltage (-100 V) was applied between voltage pulses. In all experiments a pre-breakdown light flash was observed, occurring before the main breakdown phase, at a time when the voltage was below the static breakdown voltage.

Emission spectra of the plasma in both the pre-breakdown light flash and the main breakdown phase are presented in figure 3.7. Analysis of the pre-breakdown spectrum shows
Figure 3.5: ICCD images of plasma breakdown. The integration time of the images was 5 $\mu$s. The top electrode is the anode, the bottom one the grounded cathode. Every image is an accumulation of 3000 discharges. White lines indicate the edges of the electrodes. The color scale of the first 3 images (0–20 $\mu$s) is 10 times smaller than the rest of the images. The light spot on the anode in the images was not direct plasma emission, but a reflection of the plasma around the cathode. Light reflecting of the electrode mounting stage can be seen as vertical light and dark lines behind the electrodes in images 40–80 $\mu$s.
mainly atomic argon emission and some (weak) nitrogen lines. Several spectral lines of the 2p to 1s transition (Paschen notation) in atomic argon were detected in the wavelength range between 690 and 860 nm. During the main breakdown phase, both atomic and ionic argon lines were detected. For wavelengths between 690 and 860 nm, the same 2p to 1s lines as during the pre-flash were measured. Additionally, in the wavelength range from 400 to 500 nm, four ionic lines and four atomic lines were measured. These atomic lines were the result of a 3p to 1s transition. The detection of ionic lines indicate the presence of a considerable density of ions during the main breakdown. These spectral investigations indicate that there was only minor ionization during the pre-breakdown phase.

We believe that the pre-breakdown light flash comes from charges left over from previous discharges. Such electrons are accelerated towards the anode when the voltage is applied, causing light emission in small electron avalanches. These avalanches die out because the electrode voltage is not large enough for full breakdown to occur. In one experiment we applied a small negative voltage between voltage pulses, which would act to remove charges from the discharge volume in between breakdown cycles. The fact that the pre-breakdown light flash still occurred in this situation indicates that the leftover charges are probably not in the discharge volume. We tentatively conclude that charges from previous discharges are on the nearby dielectric surfaces of the electrode mounting stage. The exact mechanisms of deposition and removal of charges from these surfaces are not yet clear.
3.3. Imaging of the breakdown process

Figure 3.7: Emission spectra obtained during the pre-breakdown light flash (a) and main breakdown phase (b) of the discharge.

Although we can tentatively conclude that the pre-breakdown light flash is probably caused by surface charges from previous discharges, the origin and exact behaviour of the phenomenon remain unclear at the moment. Further investigations on this topic can be found in chapter 4. For now, we conclude that the effect of the pre-breakdown light flash on the overall breakdown process seems limited. It is possible that charged particles produced in the pre-breakdown light flash provide initial electrons for the electron avalanches in the main breakdown phase. However, the pre-breakdown light flash seems not to change the overall behaviour and development of the breakdown phase dramatically.

3.3.3 Crossing of light front

The main phase of plasma breakdown was studied in more detail, both spatially and temporally. We used the 1:1 ICCD optical system (figure 3.3(b)) to measure ICCD images of the discharge with an exposure time of 100 ns. Figure 3.8 shows the results of the measurements between 29 and 35 µs.

The first two images (29.4–30.0 µs) of figure 3.8 show plasma emission in front of the
Chapter 3. Plasma emission imaging of a low-pressure argon breakdown

Figure 3.8: Detailed ICCD images of a moving light front. The integration time of the images was 100 ns. The top electrode was the anode, the bottom one the grounded cathode. Every image is an accumulation of 3000 discharges. The light spot on the anode in the images was not direct plasma emission, but a reflection of the plasma around the cathode.
3.3. Imaging of the breakdown process

In the next images (30.0–31.8 µs) the light front crossed the discharge gap with a velocity of approximately 1800 m/s. Finally, the light front moved down the cathode in a ring shape, covering the cathode surface with plasma.

The regions of plasma emission in the images can be associated with regions with considerable excitation. For argon, the difference in the required energy for excitation and ionization is small and the lifetime of the excited states (i.e. the time delay between excitation and emission) is in the order of tens of nanoseconds. This implies that the images in figure 3.8 can be interpreted as showing a region of ionization building up in front of the anode and then moving across the discharge gap.

The observed features are consistent with standard low-pressure, Townsend breakdown theory, discussed in chapter 2 and reference [12]. The applied electric field accelerates electrons in the discharge volume towards the anode. These electrons will cause electron avalanches, creating more electrons that accelerate towards the anode and ions that drift slowly towards the cathode. At the cathode surface, the incoming ions create new electrons by secondary electron emission, starting new electron avalanches in the discharge gap. The initial electrons that cause the first electron avalanche can originate from previous discharges or from cosmic rays in the natural background radiation.

Initially, because the electron density will be very low, there is no observable light emission. Next, the continuous electron avalanches due to secondary electron emission become stronger with time until they are sufficiently intense to cause observable light emission in front of the anode. Further, the multiple electron avalanches cause a build up of ions in front of the anode. This modifies the potential between the electrodes, when the total charge of the ions in the gap is large enough. The positive ions screen the anode and the anode potential is extended in the electrode gap towards the cathode. The result is a low-field region between the anode and the space charge region and a region with increased field between the space charge and the cathode. This leads to movement of the light front towards the cathode. Qualitatively, this explains all of the features observed in figure 3.8.

In order to understand the process more fully, I constructed a simple, one-dimensional model to investigate the behaviour of electron avalanches and ion density distributions in the discharge gap during breakdown. The purpose of this model is to get a qualitative understanding of the breakdown process. Since the electric field in the discharge gap, as shown in figure 3.2, is fairly homogeneous and we aim at a qualitative comparison with experiments, a simple one-dimensional model is suitable for our purposes. For a detailed, quantitative investigation, a full two-dimensional model would be needed.

The model calculates the number of electrons in electron avalanches and the number of ions left behind in the discharge gap. The reduced electric field in the gap is 225 V m⁻¹ Pa⁻¹ (300 V cm⁻¹ Torr⁻¹), leading to a drift velocity of 7×10⁵ m/s for the electrons and 500 m/s...
for the ions [16,17]. Therefore, the electrons cross the electrode gap almost instantaneously, relative to the time the ions take to drift towards the cathode. During one time step in the model, a complete electron avalanche develops in the discharge gap and disappears into the anode. The number of electrons in the avalanche $N$, grows towards the anode as:

$$N(x) = N(0) \exp(\alpha x),$$

(3.1)

with $N(0)$ the number of electrons at the cathode, $\alpha$ the ionization coefficient and $x$ the distance from the cathode. For our discharge (465 Pa argon, 225 V m$^{-1}$ Pa$^{-1}$) the ionization coefficient $\alpha$ is $2310$ m$^{-1}$ [12]. The electron avalanche leaves behind ions which are assumed stationary within one time step of the model. In the next time step, the drift of the ions is calculated and the ion distribution is moved towards the cathode. The ions that hit the cathode produce new secondary electrons (secondary electron emission coefficient $\gamma$ is 0.01). These electrons at the cathode produce new electron avalanches, adding new ions to the existing distribution. In the following time step, the ion distribution drifts again towards the cathode, producing secondary electrons, and subsequently a new electron avalanche and new ions. At the start of the model, a homogenous electron and ion distribution of $10^6$ cm$^{-3}$ is assumed. Ion density distributions in the electrode gap are calculated for several times during the breakdown phase. The results of the electron avalanche model for times from 0 to 7 $\mu$s are shown in figure 3.9.

The model predicts an increasing ion density in front of the anode as a result of multiple electron avalanches. The electron avalanches become stronger over time due to increasing secondary electron emission at the cathode and this process would also result in observable light emission in the discharge gap. We expect that light emission would be strongest in front of the anode, because here the ionization in electron avalanches is also strongest.

The phase in the breakdown process in which the potential in the gap is modified, is included in the model by introducing an extra low-field region. When the ion density in front of the anode is high enough ($10^9$ cm$^{-3}$), it will shield the anode potential. Hence, at this point the electric field between the space charge region and the anode is assumed to have decreased to an arbitrarily low fraction (5%) of the applied field. New electron avalanches only propagate until they reach the space charge region and undergo much less multiplication in the low-field region. Furthermore, due to the low electric field, the drift velocity of the ions decreases to 50 m/s in the low-field region.

The results of the model calculations including the modified potential are shown in figure 3.10. The space charge which developed in front of the anode extends towards the cathode, crossing the electrode gap. The electron avalanches only propagate until the space charge region, causing light emission in front of the space charge region where the field strength is large enough. In the region between the space charge and the anode, the
3.3. Imaging of the breakdown process

Figure 3.9: Results of the simple electron avalanche model showing the ion density distributions in the discharge gap for different times. A region of positive space charge builds up in front of the anode.

The electric field is very low. Hence, although the density of electrons is high in this region, their energy is too low to cause observable light emission. The result is a narrow front of light emission crossing the electrode gap from anode to cathode.

The time scales involved in the breakdown process are mainly determined by the ion drift velocity, since the breakdown process is controlled by a series of electron avalanches. Several generations of avalanches are needed to build up considerable space charge and extend this space charge region across the discharge gap. Secondary electron emission at the cathode, produced by incoming ions, provide initial electrons for these electron avalanches. Therefore, the rate at which new electron avalanches are produced is determined by the rate of ions reaching the cathode. This rate is controlled by the velocity at which the ions move towards the cathode, the ion drift velocity.

The results of the model, such as the time until observable light emission and the velocity of the moving front agree with experiments within an order of magnitude. However, for our discharge, the initial electron density, ion drift velocity and number of electrons in an avalanche needed to cause observable light emission and potential modification are not exactly known. Hence, a detailed quantitative comparison between the model and the experiments is not possible. On the other hand, the qualitative behaviour of the electron avalanches and ion distributions do not strongly depend on these parameters and are in
agreement with the observations in figure 3.8.

In summary, our observations can be explained as follows. During the start of the main breakdown phase, when the accelerating electric field between the electrodes is strong enough, a continuous electron avalanche becomes stronger until it is large enough to cause observable light emission. By the same process, the number of ions in the discharge gap increases, causing a positive space charge region in front of the anode. The anode potential becomes screened by the ions and the anode potential extends in the discharge gap. This causes the region of maximum ionization in electron avalanches to move from anode to cathode.

3.4 Time delay investigations

In the previous section, we examined the processes that occur during breakdown. In order to get a more complete view of plasma breakdown, it is also necessary to study the way that the conditions existing before the breakdown affect the breakdown process. As we use a series of breakdown pulses, we would expect that the density and distribution of charges that remain from previous pulses would influence each breakdown event. The effect of previous pulses on the breakdown behaviour is sometimes called the ‘memory effect’ [13,14].
3.4. Time delay investigations

Figure 3.11: Measured time delay as function of afterglow period for low-pressure argon breakdown. Both an electrical and an optical measurement technique were used.

In order to study this effect systematically, we varied the time between voltage pulses and measured the time delay between the application of the voltage pulse and the beginning of the breakdown process. Specifically, we altered the time between voltage pulses (i.e. afterglow period) while keeping the voltage pulse itself unchanged. For these studies, we used a voltage pulse with a rise time of 2.5 \(\mu s\) and a duration of 50 \(\mu s\).

Two different measurement methods were used to determine the time delay of the breakdown. The standard, electrical method measured the time between the voltage rise and the start of the current rise during the discharge. The second measurement method used an optical detection system. ICCD images of the crossing light front, similar to those in figure 3.8, were used to determine the time delay, here defined as the time between the start of the voltage rise and the time when the light front reached the middle of the discharge gap.

Figure 3.11 shows the results of these measurements. For afterglow periods shorter than 1 ms, the time delay was constant at 0.4 \(\mu s\), which means that breakdown occurred almost immediately after the start of the voltage rise. For afterglow periods between 2 and 20 ms, the time delay increased with increasing afterglow period. Finally, the time delay increased more slowly for afterglow periods from 20 to 200 ms. Both the electrical and optical measurements showed the same trend for the time delay.

The density distributions of charged particles in the discharge gap at the beginning of the voltage pulse determine to a large extent the time delay of the discharge. Charged
particles, produced during the discharge, decay in the subsequent afterglow period, thereby setting the initial distributions for the next discharge cycle. In order to understand the time delay dependence shown in figure 3.11, we have to understand the way in which the charged particle densities decay during the afterglow period. The main charge loss processes in our decaying plasma are diffusion and recombination. Characteristic times for these processes can be estimated as follows.

The characteristic diffusion decay time $\tau_{\text{diff}}$ can be expressed as $\tau_{\text{diff}} = \Lambda^2 / D$ [12], where $\Lambda$ is a characteristic length scale for diffusion in the discharge and $D$ is the diffusion coefficient. Assuming a diffusion coefficient, $D$, of 25 cm$^2$ s$^{-1}$ [12] and a characteristic length scale, $\Lambda$, of 1 cm, the diffusion time constant is 40 ms. This means that charge loss due to diffusion is only significant for afterglow periods of the order of 40 ms or longer.

Estimation of the characteristic volume recombination time is more complex, and first we have to consider which types of reactions are important. Previous studies on afterglow plasmas in argon [18, 19] identified dissociative recombination of molecular ions as the main recombination process. These studies indicated that the main volume recombination process is a combination of two reactions; conversion of atomic ions to molecular ions and subsequently dissociative recombination. These reactions are

$$\text{Ar}^+ + \text{Ar} + \text{Ar} \rightarrow \text{Ar}_2^+ + \text{Ar},$$

$$\text{Ar}_2^+ + e \rightarrow \text{Ar} + \text{Ar}^*,$$

where $\text{Ar}$ is a ground-state argon atom, $\text{Ar}^+$ an argon ion, $\text{Ar}_2^+$ a molecular argon ion, $\text{Ar}^*$ a metastable argon atom.

In order to estimate the time dependence of charged particle decay due to these processes, we solved this simple set of rate equations. We assumed initial charged particle densities (i.e. densities at the start of the afterglow period) of $10^{12}$ cm$^{-3}$ for both ions and electrons, with a ratio of $n_{\text{Ar}^+} / n_{\text{Ar}_2^+}$ of 100:1. The conversion reaction (3.2) has a rate coefficient $k_1$ of $2 \times 10^{-31}$ cm$^6$ s$^{-1}$ [20]. The dissociative recombination reaction (3.3) has a rate coefficient $k_2$ that depends on electron temperature, and hence we must estimate this. Using data from reference [18] and an estimated electron temperature of 0.5 eV, we derived a value for $k_2$ of $10^{-7}$ cm$^3$ s$^{-1}$.

Figure 3.12 is the result of the calculation, showing the time dependence of charged particle densities in the afterglow period. Two distinct phases of charged particle decay can be identified from figure 3.12. In the first phase, before about 10 ms, $\text{Ar}^+$ is the dominant ion, and charge loss is relatively fast with a characteristic decay time of about 1 ms. During this phase, charge loss will occur as $\text{Ar}^+$ is converted into $\text{Ar}_2^+$, which then recombines with electrons. In the second phase, after about 10 ms, loss of charges is much
slowly having a characteristic decay time of about 10 ms. Here, most of the Ar$^+$ has been converted into Ar$_2^+$, and charge loss occurs via Ar$_2^+$–e recombination.

By comparing these characteristic decay times for volume recombination with the diffusion decay time of 40 ms calculated before, we can identify the dominant charge loss mechanisms. During the first phase of the afterglow, volume recombination, with a characteristic time of 1 ms, is clearly dominant over diffusion losses. In the second decay phase, volume recombination has a characteristic time of 10 ms, which is comparable to the diffusion decay time of 40 ms. During this phase, charge loss occurs through a combination of recombination and diffusion. The transition point between the two decay phases lies around 10 ms.

The time delay measurements in figure 3.11 also show a transition between two regimes. The first regime, characterized by a fast increasing time delay and a second regime with a much slower increase in time delay. Here, the transition between the regimes lies around 20 ms. As mentioned before, the charged particle densities at the beginning of the voltage pulse determine to a large extent the time delay of the discharge. Therefore, the two phases of the measured time delay can be qualitatively explained by the two regimes of charge loss, calculated before.

In summary, the shape of the measured memory curve in figure 3.11 can be qualitatively explained by loss of charges in the afterglow of the previous discharge and the influence of initial charge density on the time delay of the discharge. The sharp increase in time delay
during the first 20 ms of the afterglow period can be explained by a fast loss of charges due to volume recombination. The slow increase in time delay for afterglow periods between 20 and 200 ms, is due to diffusion and dissociative recombination. The loss of charges in this phase is slower than before leading to a slower increase in time delay.

### 3.5 Conclusions

Time-resolved ICCD imaging of breakdown in a low-pressure argon discharge gives a clear view on the basic phenomena of plasma breakdown. These include, the formation of a light emission region in front of the anode and crossing of this light front from anode to cathode. The observed features can be explained by a continuous electron avalanche developing in the discharge gap due to secondary electron emission at the cathode. This avalanche causes light emission due to excitation of the discharge gas and modification of the potential in the electrode gap due to a buildup of space charge. The time scales involved in the breakdown process are mainly determined by the ion drift velocity. We conclude that the breakdown process is controlled by the development of multiple electron avalanches. This is consistent with the standard view of Townsend breakdown into a glow discharge at low pressure.

In addition, we observe a pre-breakdown light flash around the anode at a time when the applied voltage is below the static breakdown voltage, which can not be explained by the standard Townsend breakdown theory. Although the details of this light flash remain unresolved for the moment, it is thought to come from charges from previous discharges on dielectric surfaces close to the electrode gap. Further study is needed to clarify this issue. Chapter 4 contains additional investigations on pre-breakdown phenomena.

Finally, measurements of time delays of the discharge show the effects of the initial conditions on the breakdown behaviour. Longer afterglow periods result in an increase in time delay of the next discharge. Volume recombination and diffusion are identified as the main loss processes for charged particles. These mechanisms can qualitatively explain the behaviour of the time delay of the discharge.

In this chapter breakdown features were studied by measuring plasma light emission. In chapter 7, breakdown is further studied by measuring electric field distributions during discharge ignition.

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References


Chapter 4

Pre-breakdown light emission phenomena in low-pressure argon between parabolic electrodes

Abstract. An experimental study on pre-breakdown light emission in low-pressure argon gas was performed. In a pulsed discharge, pre-breakdown phenomena were observed for repetition rates between 100 and 2000 Hz and pulse duration of 100 $\mu$s. These phenomena were studied with time-resolved emission imaging using an intensified charge coupled device camera. The origin of the pre-breakdown emission was identified as diffusion of volume charges left over from previous discharges. These charges were accelerated towards the anode in small electron avalanches causing excitation of argon atoms. Different spatial distributions of the pre-breakdown light emission for different times between discharges were measured and the effects of the pre-breakdown phenomena on the main breakdown phase were studied using a double voltage pulse. The observed effects were attributed to the distribution of volume charges, left over from previous discharges, in the discharge gap during the pre-breakdown phase.

Chapter 4. Pre-breakdown phenomena in low-pressure argon

4.1 Introduction

Breakdown, also called plasma ignition or gas breakdown, is the process that describes the transition from a gas to a self-sustained discharge. During this ignition phase, an applied electric field changes the electrically insulating gas to a conducting state. Processes such as charge multiplication in electron avalanches and build-up of space charge regions between the electrodes play an important role in breakdown.

In many plasma applications such as high-intensity discharge lamps and fluorescent lamps [1,2], air and water purification systems based on corona discharges [3,4] and plasma display panels [5,6], electrical breakdown is an important topic of research. Some aspects of plasma breakdown under these specific conditions are reasonably well understood. However, details of the breakdown process for a wide range of discharge geometries and conditions and details on the formation of sheath structures in a glow discharge are not fully understood.

In the beginning of the last century, Townsend developed a theory describing the breakdown process between parallel plates in low-pressure gas [7]. This theory is based on a description of the microscopic processes of breakdown such as electron multiplication in avalanches and secondary electron emission at the cathode. The Townsend theory gives an accurate description of breakdown for a large number of gas discharges but the set of conditions when it is applicable is limited. Especially, systems with non-uniform electric fields or complex geometries are not fully described by the existing theory.

In recent years, we have developed an experimental arrangement for the study of plasma breakdown in low-pressure gas. A discharge system consisting of a set of parabolic electrodes driven with a pulsed voltage was built. This discharge was studied by light emission imaging with an intensified charge-coupled device (ICCD) camera. More details on these investigations can be found in chapter 3. In this previous study, features such as light emission in front of the anode and a crossing ionization front were observed. Most of these phenomena were in agreement with Townsend breakdown theory, but not all observed features could be directly explained. Specifically, a weak light flash close to the anode was observed at a time when the voltage was still below the breakdown voltage. At that time it was thought that this phenomenon was caused by charges from previous discharges that had been deposited on nearby dielectric surfaces. However, the exact origin of the light flash and the detailed processes involved, remained unknown.

The aim of this research was to investigate the behaviour and cause of the pre-breakdown emission in more detail and to study the effect of the pre-breakdown phenomena on the main breakdown phase. In order to do this, plasma emission imaging experiments were performed for different pulse shapes and repetition rates of the applied voltage pulse.
4.2. Experimental arrangement

The structure of this chapter is as follows. Section 4.2 contains a brief description of the experimental apparatus. Section 4.3 consists of results and discussion of investigations on the behaviour and origin of the pre-breakdown light emission. Section 4.4 focuses on the effects of the pre-breakdown light flash on the main breakdown process. Finally, section 4.5 contains the conclusions.

4.2 Experimental arrangement

The experimental arrangement is similar to that used in our previous work. Hence, we will only briefly describe its main features here; details about the apparatus can be found in chapter 3.

The discharge apparatus is schematically shown in figure 4.1. It consisted of two cylindrically symmetric stainless-steel electrodes. In the experiments described here, the tips were parabolically shaped with a radius of curvature of 1 mm. This was a minor change, as will be shown in section 4.3.1, from our previous experiments, described in chapter 3, in which the tips had a curvature of 4 mm. The distance between the electrodes was fixed at 3.3 mm for all measurements. The vacuum chamber was evacuated to a base pressure of about $10^{-4}$ Pa after which it was filled with argon gas and kept at a pressure of 465 Pa with a continuous gas flow controlled by a needle valve. The gas entered the vacuum vessel with a velocity of about 2 m/s. A repetitive discharge was created by applying voltage pulses to the electrodes. The output of an arbitrary waveform generator was amplified in a power amplifier.
amplifier (Spitzenberger EV600/CuG) to an amplitude of several hundreds of volts. The rise time, amplitude and repetition rate of the voltage pulses could be varied. For a typical case, the voltage pulse had 100 μs duration and 500 Hz repetition rate with the time in between consecutive discharges being 2 ms. The breakdown voltage of the experimental arrangement was measured to be about 345 V.

An ICCD camera (Andor Technology DH534) was used to record the total light emission from the discharge. The intensifier of the camera had a minimum gate duration of 3 ns and a gain of up to 1000 counts per photoelectron. All light emission in the wavelength range from 350 to 850 nm was detected. A camera macro lens (Sigma 50mm f2.8) imaged the discharge region onto the ICCD camera. A delay generator controlled the gating of the ICCD camera and synchronized it with the voltage pulse. The consecutive discharges were reproducible within 50 ns for each voltage cycle. For the measured images, light from 1000 discharge cycles was accumulated with the ICCD camera.

4.3 Behaviour and origin of pre-breakdown phenomena

This section contains the results and discussion of our investigations on the origin of the pre-breakdown light emission. In section 4.3.1 we briefly describe the entire breakdown cycle and identify the different phases. Section 4.3.2 contains investigations on the effect of pulse repetition frequency on the pre-breakdown emission behaviour. In section 4.3.3 the origin of the pre-breakdown emission phenomena is discussed.

4.3.1 Full breakdown cycle

Plasma breakdown was studied under similar conditions as our previous investigations, presented in chapter 3, and qualitatively similar results were obtained. A discharge was created at a gas pressure of 465 Pa (3.5 torr) by applying positive voltage pulses to the top electrode, while the bottom electrode and the vacuum chamber were grounded. The applied voltage pulse had a rise time of 30 μs, a duration of 100 μs, an amplitude of 380 V and a repetition frequency of 500 Hz. Figure 4.2 presents ICCD images with an integration time of 1 μs, showing an overview of the ignition phase of the discharge. From these images, different features of plasma breakdown can be identified. Firstly, at $t = 6 \mu s$, a weak light flash was observed close to the anode. At this time, the voltage was rising but still below the breakdown voltage and no significant current could be observed ($I < 0.1$ mA). This flash of light was identified as the pre-breakdown light emission observed in the previous investigations in chapter 3. At the time, $t = 26 \mu s$, the main breakdown phase started with weak light emission in front of the anode. This light front crossed the electrode gap.
4.3. Behaviour and origin of pre-breakdown phenomena

Figure 4.2: ICCD images showing an overview of plasma breakdown. The images are accumulations of 1000 discharge cycles; each image had an integration time of 1 µs. The top electrode is the anode, while the bottom electrode is the grounded cathode. The intensity scale of the first five images is five times smaller than the last five images. White lines indicate the edges of the electrodes.

and covered part of the cathode \( t = 30 \, \mu s \). Next, the plasma emission covered the entire cathode and stabilized \( t = 40-50 \, \mu s \). Finally, the discharge extinguished when the voltage was switched off \( t = 70 \, \mu s \).

This breakdown behaviour is the same as described in chapter 3, indicating that changing the curvature of the electrode tips did not change the breakdown phenomena observed. In this chapter we will focus on the characteristics of the pre-breakdown light emission as seen in figure 4.2 around \( t = 6 \, \mu s \).

4.3.2 Frequency dependence of pre-breakdown emission

To investigate the origin of the pre-breakdown light emission we varied the repetition frequency of the pulses while keeping other conditions constant. We measured ICCD images for discharges with repetition frequencies from 100 to 2000 Hz, which corresponds to 10 to 0.5 ms between discharges. In this frequency range the characteristics of the main discharge were similar for all frequencies and the pre-breakdown phenomena were clearly observed. The results for three frequencies are presented in figure 4.3. For repetition frequencies between 100 and 200 Hz, the behaviour was not significantly different from the 200 Hz situation. Additionally, the results from the experiments with a repetition
Figure 4.3: Frequency dependence of pre-breakdown light emission. For the first row of images, the pulse repetition frequency was 200 Hz, in the second row 500 Hz, and in the bottom row 1000 Hz. Each image is an accumulation of 1000 discharges. Note that the intensity scales are different for the different measurement series.

rate of 2000 Hz were very similar to the situation with 1000 Hz. The conditions for the experiments with a repetition frequency of 500 Hz were the same as for the measurements shown in figure 4.2. The detailed measurements in figure 4.3 show that for 500 Hz the pre-breakdown light started at $t = 6 \, \mu s$, had its maximum intensity around $10 \, \mu s$, after which it died out in the next $10 \, \mu s$. It was located around the tip of the anode, extending into the electrode gap.

Decreasing the repetition frequency to 200 Hz resulted in light emission which was spread out over a larger area of the anode. The light emission was weaker, less localized and lasted longer than was the case for 500 Hz. At the maximum emission ($t = 10 \, \mu s$), a stratified structure could be observed. Increasing the repetition frequency to 1000 Hz lead to more localized light emission near the anode tip. Also the duration of the light flash was shorter than for the other measurement series. The intensity of the light was higher than for 200 Hz, but lower than the 500 Hz case.

For all images presented in figure 4.3, light from 1000 discharges was accumulated. The fact that these images show clear features indicates that the statistical effects in the breakdown development were not very large. If there would be considerable statistical
4.3. Behaviour and origin of pre-breakdown phenomena

variations in the breakdown behaviour, the images would be accumulations of different times in the breakdown process and therefore would be smeared out. This was not the case, indicating that statistical effects were limited in our discharge. From additional ICCD imaging experiments, using an integration time of 100 ns, we concluded that the statistical variations were about 50 ns.

To further investigate the effects of diffusion during the off-period of the discharge we performed experiments with negative voltage pulses. These pulses were applied to the bottom electrode, while the top electrode and the vacuum vessel were grounded. In these experiments the grounded vacuum vessel was at the same potential as the anode, while in the previous measurements its potential was equal to the cathode. The differences between the voltage polarities in the pre-breakdown behaviour were minor. The frequency dependence was the same as for positive pulses. The only observed difference between discharges with positive and negative pulses was that the overall intensity of the pre-breakdown emission was weaker for negative voltage pulses.

In additional experiments we used a reversed discharge configuration; we applied normal, positive voltage pulses to the bottom electrode, while the top electrode was grounded. In this situation, the bottom electrode acted as the anode and the top electrode was the cathode. These results showed no significant differences with the original experiments presented in figures 4.2 and 4.3.

4.3.3 Origin of pre-breakdown light emission

The experimental observations in the previous sections raise questions about the origin of the pre-breakdown light emission occurring at voltages below the static breakdown voltage. Our hypothesis is that the pre-breakdown light emission in all our experiments can be explained by volume charges left over from previous discharges. The left-over electrons are accelerated towards the anode as soon as the voltage starts to rise. These electrons gain energy in the electric field and cause small electron avalanches. In these avalanches there is charge multiplication and considerable excitation of argon atoms since the mean free paths for these processes are in the order of 500 $\mu$m, which is much smaller than the characteristic length scales of our discharge. This results in directly observable light emission since the lifetime of excited argon atoms is in the order of nanoseconds.

The dependence of the spatial distribution of the pre-breakdown emission on repetition frequency, as presented in figure 4.3, indicates a volume effect; longer times between discharges lead to more diffusion of volume charges and hence a frequency dependence. If the pre-breakdown phenomena were caused by charges on nearby dielectric surfaces, we would not expect differences in the spatial distributions of the pre-breakdown emission as
function of pulse frequency. Because the spatial characteristics of the main discharge at the end of the pulse were the same for all investigated frequencies, we would expect charges to be deposited at the same locations of the nearby dielectric surfaces. This would result in the same behaviour of the spatial distribution of the pre-breakdown light emission for all repetition frequencies. Since this is not observed, it can be concluded that the pre-breakdown emission is not caused by charges originating from dielectric surfaces as was tentatively suggested in chapter 3.

The spatial variations of the pre-breakdown emission for different frequencies (figure 4.3) can be explained by differences in the charge distribution at the start of the voltage pulse. Since the general characteristics of the main discharge were similar for all experimental conditions under study, the initial distribution of charges is mainly determined by processes in the afterglow of the previous discharge. During the period between voltage pulses, the charge densities decay through both ambipolar diffusion and volume recombination processes.

Additionally, in the experiments with negative pulses, less intense light emission close to the anode was observed. This can be explained by the fact that part of the left-over volume electrons were accelerated to the positive vessel wall instead of the electrode. This supports the hypothesis that left-over volume charges are the origin of the pre-breakdown emission.

Our previous investigations on discharge time delays (chapter 3) showed that for afterglow periods of 1–5 ms, volume recombination was dominant over ambipolar diffusion. At longer afterglow times (20–200 ms) the recombination decay became less dominant and diffusion processes became important. However, these investigations only dealt with the charge densities in the afterglow; they did not describe the spatial charge distributions in the discharge region.

In our discharge geometry, the electric field distribution during the discharge pulse was very inhomogeneous with different fields at different positions. Therefore, even diffusion over small (radial) distances during the afterglow had a large effect on the electric field felt by the electrons at the start of the next voltage pulse. In this way, diffusion had an effect on the path that the accelerated electrons followed and where the electron avalanches reached the anode. A calculation of the static electric field distribution in the discharge volume is shown in figure 4.4.

To estimate the importance of diffusion, we can calculate the mean distance over which the electrons diffuse, \( \bar{r} \), for afterglow periods of 1–5 ms. The mean distance for diffusion of particles in a sphere, can be described by [8]

\[
\bar{r} = \sqrt{\frac{12D\tau_{\text{diff}}}{\pi}},
\]

(4.1)
4.3. Behaviour and origin of pre-breakdown phenomena

Figure 4.4: Calculated static electric field distribution in the discharge region. The top electrode is at a positive potential. The bottom electrode is grounded. The arrows represent the electric field vectors at different points. The background colour indicates electric field strength, with darker colours for higher fields. The axes’ labels show distances in millimetres.

where $\tau_{\text{diff}}$ is the diffusion time and $D$ the diffusion coefficient. Assuming an ambipolar diffusion constant of 25 cm$^2$ s$^{-1}$ [9] and 1 ms afterglow time, the characteristic length is 3 mm, meaning that during the afterglow, charges from the stable plasma around the cathode tip stayed inside the discharge gap, close to the electrodes. At the start of the next voltage pulse, these charges caused avalanches following the electric field lines, ending at the tip of the anode. This made the pre-breakdown light emission to be located close to the anode tip.

For lower repetition rates, i.e. 200 Hz, the afterglow period was longer (5 ms) and the characteristic diffusion length was 7 mm. This means that the charges diffused more in the radial direction and at the next voltage pulse, these charges were accelerated towards the sides of the anode, further away from the tip.

This type of behaviour was confirmed by the experiments shown in figure 4.3. At high repetition frequencies, i.e. 1000 Hz, the pre-breakdown light emission was located near the tip of the anode. It was caused by charges originating from near the centre of the discharge gap. At lower repetition frequencies, the pre-breakdown light flash was more spread out over the anode surface because the initial distribution of charges was more spread out over the discharge volume due to diffusion.
All the observations are consistent with our hypothesis that the origin of the pre-breakdown light emission is volume charges left over from previous discharges. Additionally, our investigations show that the history of the discharge influences the initial conditions and through this the starting behaviour of the discharge. Whether the main breakdown phenomena \((t \approx 25-50 \mu s)\) are influenced by the characteristics of the previous discharge is investigated in the next section.

### 4.4 Effects of starting conditions on breakdown behaviour

In this section, we will describe an experiment that enables us to study the effect of the pre-breakdown phenomena on the main breakdown process in more detail. The initial conditions of the discharge were controlled using a double voltage pulse as will be described in section 4.4.1. Section 4.4.2 contains the results of investigations on the effects of the pre-breakdown emission on the main breakdown behaviour.

#### 4.4.1 Separation of pre-breakdown phenomena from the main breakdown

To control the behaviour of the pre-breakdown light emission we applied a double voltage pulse, shown in figure 4.5. It consisted of the normal voltage pulse preceded by a small pre-pulse. This pre-pulse had a rise time of 10 \(\mu s\), an amplitude of 70% of the main pulse and a total duration of 30 \(\mu s\). The start of the voltage rise of the main pulse was labelled \(t = 0 \mu s\). There was no significant current measured during the pre-pulse; only during the
4.4. Effects of starting conditions on breakdown behaviour

Figure 4.6: ICCD images of plasma breakdown using a double voltage pulse (figure 4.5), separating the pre-breakdown emission from the main breakdown. The repetition frequency of the voltage pulses was 500 Hz. Note that the intensity scale for the middle row of images ($t = 0-26 \, \mu s$) is ten times smaller than for the other images.

Images of plasma emission during breakdown using the double voltage pulse are shown in figure 4.6. These images show that the pre-breakdown emission now occurred during the pre-pulse as intended, and that the main breakdown phase started during the main voltage pulse. The characteristics of the pre-breakdown emission during the pre-pulse ($t = -30-0 \, \mu s$) were very similar to the pre-breakdown behaviour of the previously used voltage pulse (figure 4.3). In this case, however during the voltage rise of the main pulse ($t = 0-25 \, \mu s$) no observable light emission was detected. This confirms that with this double voltage pulse, we could separate the pre-breakdown phenomena from the main breakdown phase and control the initial conditions of the breakdown process.
Chapter 4. Pre-breakdown phenomena in low-pressure argon

Figure 4.7: Images of breakdown from figures 4.2 and 4.6, showing the effect of the voltage pulse waveform on the timing of breakdown events. For the first series a normal voltage pulse was used, while for the second series a voltage pulse with a pre-pulse (figure 4.5) was applied.

4.4.2 Effect on main breakdown behaviour

In the research described here, we investigated the effects of the pre-breakdown phenomena on the ignition phase of the main breakdown. Figure 4.7 shows ICCD images of the main breakdown phase for two different voltage pulses, with and without a pre-pulse.

It is clear from these experiments that removing the pre-breakdown emission from the main pulse with a pre-pulse did not change the qualitative behaviour of the main breakdown process significantly. Characteristic breakdown features such as first light emission in front of the anode, a moving light front crossing the gap and plasma covering the cathode, are seen for both voltage pulses. However, applying a double voltage pulse did quantitatively change the breakdown behaviour. It resulted in a slower discharge ignition process during the main breakdown phase. Crossing of the gap by a light front occurred about 4 µs later than in the normal case. Also, the onset of current development (figure 4.5) started a few µs later. Furthermore, the development of the plasma covering the cathode was significantly slower for the discharge with the double voltage pulse. A slower rise of the measured current confirmed that the discharge development was slower in the case with the double voltage pulse.

The results and discussion in the previous section indicate that the pre-breakdown light emission close to the anode was caused by volume charges left over from the previous discharge which were accelerated towards the anode in small electron avalanches. These avalanches caused a concentration and multiplication of charges in the discharge gap, close
4.4. Effects of starting conditions on breakdown behaviour

to the anode. Additionally, the ions produced in these avalanches and those left over from
the previous discharge, slowly drifted towards the cathode, resulting in secondary electron
emission at the cathode surface. The result of these processes was that the charges left
over from the previous discharge were no longer distributed over the vessel volume as a
result of diffusion, but were concentrated close to the electrode gap.

The pre-breakdown process changed the spatial charge distribution in the discharge
gap. When a normal voltage pulse was applied, the pre-breakdown phase occurred right
before the main breakdown. This caused an increase in charges in the gap during the main
breakdown phase, which influenced the breakdown behaviour. For the double voltage pulse,
the pre-breakdown effects occurred during the pre-pulse, long before the main breakdown
phase.

The electron density in an electron avalanche is proportional to the initial electron
density. An increased initial density, for instance as a result of pre-breakdown processes,
will therefore lead to an increased density in the entire avalanche, resulting in a faster
build-up of space-charge in front of the anode. Subsequently, disturbance of the applied
electric field, indicated by the onset of a moving ionization front, will occur earlier during
the voltage pulse because of the pre-breakdown charge concentration.

Furthermore, the higher charge densities close to the discharge gap also result in a faster
development of the plasma covering the cathode. Again, this feature can be attributed to
a higher charge density close to the electrode tips, resulting in faster charge multiplication
and plasma development. The timing of the different breakdown phenomena changed for
different voltage pulses, but the actual processes involved remained the same.

It can therefore be concluded that the qualitative behaviour of the main breakdown
is not influenced significantly by the pre-breakdown emission. However, the quantitative
details such as ignition delay and plasma development are strongly influenced by the initial
conditions of breakdown set by the previous discharge and the pre-breakdown phenomena.

The effect of previous pulses on the time delay of plasma breakdown is sometimes re-
ferred to as the ‘memory effect’ and has been studied in detail in the past. For instance
in chapter 3 of this thesis and in references [10–12]. From these investigations it was con-
cluded that previous discharges could influence the breakdown time delay, up to hours
after the previous discharge. Marković et al. [13] conclude that for short afterglow times,
less than 10 ms, the start of the breakdown process is dominated by secondary electron
emission caused by left-over ions. For longer afterglow times, secondary emission caused
by metastables becomes important. These results indicate that for our experimental con-
ditions, afterglow periods between 0.5–10 ms, the (pre-)breakdown processes are mainly
caused by charges left over from previous discharges. The effects of metastable atoms from
previous discharges are expected to be minor.
4.5 Conclusions

Using time-resolved ICCD imaging we studied pre-breakdown light emission in a pulsed discharge in low-pressure argon gas. By applying different voltage pulses to create the discharge we studied the origin and effects of these pre-breakdown emission phenomena.

The origin of the pre-breakdown emission was found to be charges left over from previous discharges. These left-over charges are volume charges and not charges on nearby dielectric surfaces, as was tentatively suggested in chapter 3.

The characteristics of the pre-breakdown phenomena are very sensitive to the initial conditions of the discharge. Processes in the afterglow of the previous pulse such as volume recombination and diffusion are therefore important. For afterglow periods in the order of 1 to 5 ms, volume recombination is the main loss process for charge densities. However, the spatial characteristics of the pre-breakdown emission are caused by ambipolar diffusion during the afterglow. Diffusion processes change the spatial distribution of the charges during the afterglow. The very inhomogeneous spatial distribution of the electric field in our discharge makes that different charge distributions, especially in the radial direction, strongly change the position and spatial characteristics of the pre-breakdown light in the next pulse.

From our investigations on the effect of the pre-breakdown phenomena on the main breakdown behaviour, we found that the qualitative behaviour of the main breakdown phase of the discharge is the same with or without pre-breakdown emission. However, the details, especially the timing of the different phases, are influenced by the pre-breakdown processes.

Using a small voltage pulse before the main pulse, we could separate the pre-breakdown light from the main breakdown process. In this case, during the rising edge of the main voltage pulse no light emission could be detected. The voltage pre-pulse was strong enough to attract the left-over electrons towards the anode in small electron avalanches, creating light emission. However, the pre-pulse was not sufficient for full breakdown to occur.

In the normal-pulse discharge, that is with pre-breakdown emission on the rising edge of the voltage, the main breakdown starts earlier and develops faster. This indicates that during the pre-breakdown process, left-over charges from a large volume are concentrated in the discharge gap. The higher initial charge density makes the electron avalanches in the main breakdown stronger and the breakdown development faster. This ability to influence and control the breakdown behaviour of a discharge by adjusting the initial conditions can be useful for industrial applications in which breakdown is an important issue.

For further quantitative details on the various processes involved in the pre-breakdown phase and the plasma afterglow, a modelling study is currently being performed. Our
model is two-dimensional and includes not only the discharge phase of the plasma, but also the afterglow phase, including the relevant processes. Results from these modelling investigations are expected in the near future.

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References


Chapter 5

The Stark effect

Abstract. The Stark effect can be described as the shifting, splitting and mixing of energy levels of an atom or molecule under the influence of an external electric field. This chapter discusses (calculations of) Stark effects in more detail and is intended as an introduction to the experimental investigations, using Stark effects, presented in chapters 6 and 7. Firstly, the discovery of the Stark effect in the beginning of the last century is briefly discussed. Next, calculations of linear and quadratic Stark effects in hydrogen, based on perturbation theory, are presented as an illustrative example. The main part of the chapter deals with the calculation of Stark effects in noble gas Rydberg atoms. Since standard perturbation theory is not suitable for this situation, an alternative method is presented. This method is based on solving the Schrödinger equation including the electric field perturbation by a numerical diagonalization of the total Hamiltonian. In chapter 6, results from this calculation procedure are compared with experimental investigations. Finally, the main ideas of multichannel quantum defect theory are briefly described as a more sophisticated theory to describe Stark effects. This theory can accurately describe Stark effects for atoms with penetrating electron orbits, which is not possible using matrix diagonalization.
5.1 Discovery of the Stark effect

In 1913, Johannes Stark and Antonio Lo Surdo independently discovered the splitting of hydrogen line emission from a discharge under the influence of an electric field [1]. Nowadays, this effect is commonly known as the Stark effect, although it is sometimes referred to as the Stark–Lo Surdo effect.

The discovery of the Stark effect came as a surprise because within the framework of classical physics, the effect of an electric field on the splitting of spectral lines was calculated to be much smaller than what could be observed experimentally at that time. These calculations were based on the fact that an atom can be polarized by an external electric field, $\mathbf{F}$, following [2]

$$ \mathbf{p} = \alpha_{\text{pol}} \mathbf{F}, \quad (5.1) $$

where $\alpha_{\text{pol}}$ is the polarizability of the atom. The corresponding polarization energy is described by

$$ \Delta E = \int \mathbf{F} \cdot d\mathbf{p} = \alpha_{\text{pol}} \int \mathbf{F} \cdot d\mathbf{F} = \frac{\alpha_{\text{pol}}}{2} F^2, \quad (5.2) $$

and can be interpreted as a shift of the energy levels of the atom. This calculation showed that the expected effect had a quadratic dependence on electric field strength.

Stark’s experiments showed not only line shifts that were much larger than expected, also a linear dependence between the shift of the level and the electric field strength was observed. Clearly, Stark’s discoveries could not be explained by the classical atomic theory. At that time Bohr had just developed a new atomic theory which could explain the observed effects. Stark’s investigations therefore made a significant contribution to the acceptance of Bohr’s new quantum theory of the atom.

Over the last century, the Stark effect has received less attention than its magnetic counterpart, the Zeeman effect. This is mainly due to the fact that high electric fields are needed to cause measurable Stark effects, while for only moderate magnetic fields, the Zeeman spectral splitting is obvious. Nevertheless, the Stark effect has played an important role in atomic physics, for instance in the verification of Schrödinger’s quantum mechanics, in the determination of the hydrogen Lamb shift, and in the assignment of mixed states in complex spectra [2].

5.2 Stark effects in hydrogen

The Coulomb electric field inside an atom, $F_{\text{Coulomb}}$, is in the order of

$$ F_{\text{Coulomb}} = \frac{1}{4\pi\varepsilon_0} \frac{e}{a_0^2} = 5 \times 10^9 \text{ V/cm}, \quad (5.3) $$
where $e$ is the elementary charge, $\varepsilon_0$ the vacuum permittivity, and $a_0$ the Bohr radius. Maximum electric fields that are typically available in a laboratory are in the order of $10^6$ V/cm. Therefore externally applied electric fields can be considered as a small perturbation to the internal field of the atom.

In the quantum mechanical description of atoms, the external electric field can therefore be treated as a small perturbation in the Schrödinger equation:

\begin{align*}
\text{unperturbed atom: } & H_0 |\psi_0\rangle = E_0 |\psi_0\rangle \quad (5.4a) \\
\text{perturbed atom: } & (H_0 + H_{\text{Stark}}) |\psi\rangle = E |\psi\rangle. \quad (5.4b)
\end{align*}

In these equations $H_0$ is the unperturbed Hamiltonian, $E_0$ the energies of the states in zero-field, $\psi_0$ the unperturbed wavefunction, $H_{\text{Stark}}$ the perturbation Hamiltonian due to the Stark effect and $\psi$ the perturbed wavefunction. The Stark Hamiltonian, $H_{\text{Stark}}$ can be evaluated using a dipole approximation

$$H_{\text{Stark}} = -P \cdot F,$$

where $P = \sum_i e r_i$ is the atomic electric dipole operator. When we assume the electric field, $F$, to be parallel to the $z$ axis, $H_{\text{Stark}}$ reduces to

$$H_{\text{Stark}} = eFz,$$

(5.6)

To calculate the energies of the atomic levels in an electric field, the perturbed Schrödinger equation (5.4b) needs to be solved. There are different possibilities to achieve this. The first is to use perturbation theory to describe the small corrections to the energies of the atomic levels. For simple systems in low electric fields this method works well. Two examples of more sophisticated methods, matrix diagonalization and multichannel quantum defect theory, are discussed in sections 5.4 and 5.5.

In this section, we will briefly discuss a perturbation approach for calculation of Stark effects in hydrogen. Using quantum mechanics instead of a classical approach, we can explain the linear shifts of hydrogen levels as was observed by Stark in the early 1900s.

Following perturbation theory up to the second order, the energy of a level, perturbed by an electric field can be described as [2]:

\begin{align*}
E &= E_0 + \langle \psi_0 | H_{\text{Stark}} | \psi_0 \rangle + \sum_n \frac{|\psi_0| |H_{\text{Stark}}| \psi_n|^2}{E_0 - E_n} \\
&= E_0 + eF \langle \psi_0 | z | \psi_0 \rangle + e^2 F^2 \sum_n \frac{|\psi_0| z | \psi_n|^2}{E_0 - E_n}. \quad (5.7)
\end{align*}

The first term on the right-hand side is the energy of the level without the electric field. The second term on the right-hand side describes a linear Stark effect, while the third
term adds a quadratic Stark shift. It is important to realize that in general, on ground of parity conservation, the second term on the right-hand side vanishes [3]. The operator \( z \) has the same characteristics as the operator for an electric dipole transition, which is only non-zero for transitions between states of opposite parity, that is \( \Delta l = \pm 1 \).

In second order perturbation theory, a quadratic Stark shift can be expected, because states of different parity are mixed into \(|\psi_0\rangle\). Additionally, selection rules demand \( \Delta M = 0 \) which means that states with the same projection quantum number, \( M \), are mixed. The total Stark effect therefore shows a quadratic dependence on electric field strength.

However, there is an important exception to this situation. In a non-relativistic approach, hydrogen atoms have \( l \)-degenerate levels for given principal quantum numbers \( n \). Therefore, the linear term in equation (5.7) does not vanish and a linear Stark effect can be observed, since first-order terms are much larger than second-order terms.

Following the perturbation theory described before, Schrödinger calculated Stark effects in hydrogen atoms. Using a parabolic coordinate system, he found the following results for the Stark energy shift [2]. The first-order, linear shift is described by

\[
\Delta E_1 = \frac{3}{2} e a_0 n(n_1 - n_2) F, \tag{5.8}
\]

where \( n \) is the principal quantum number and \( n_1 \) and \( n_2 \) are the parabolic quantum numbers [2] which are defined by \( n = n_1 + n_2 + M + 1 \).

The result for the second order, quadratic shift is

\[
\Delta E_2 = -\frac{(4\pi\varepsilon_0)}{16} a_0^3 n^4 [17n^2 - 3(n_1 - n_2)^2 - 9M^2 + 19] F^2. \tag{5.9}
\]

It is clear from equations (5.8) and (5.9) that the Stark shift is a strong function of the principal quantum number \( n \), leading to much larger shifts for highly-excited states.

As an illustrative example we will now look at the \( n = 2 \) levels in hydrogen in more detail. We will follow the discussion as presented in [3]. When fine structure of the hydrogen atom is taken into account, the \( n = 2 \) level is split into 2 levels, \( ^2P_{3/2} \) and \( ^2P_{1/2} \) and a degenerate level, \( ^2S_{1/2} \). Since the latter level is degenerate we expect a linear Stark shift for this level. However, when taking quantum electrodynamical corrections into account, the \( ^2P_{1/2} \) and \( ^2S_{1/2} \) states are not truly degenerate, but separated by the Lamb shift.

When we assume that for \( F = 0 \), the \( ^2S_{1/2} \) state is described by \(|s\rangle\) and \( ^2P_{1/2} \) by \(|p\rangle\) the situation can be described by

\[
H_0|s\rangle = E_s|s\rangle
\]
\[
H_0|p\rangle = E_p|p\rangle, \tag{5.10}
\]

with \( E_s - E_p = \Lambda \approx 1058 \text{ MHz} \), the Lamb shift.
When an electric field $\mathbf{F}$ is present, $|s\rangle$ and $|p\rangle$ are no longer the correct eigenfunctions. There will be mixing of these states and we assume that the new situation can be described by

$$|\psi\rangle = \alpha|s\rangle + \beta|p\rangle. \quad (5.11)$$

Substitution of this equation into the Schrödinger equation (5.4b) results in

$$\alpha(E_s + ezF)|s\rangle + \beta(E_p + ezF)|p\rangle = \alpha E|s\rangle + \beta E|p\rangle. \quad (5.12)$$

Multiplying this equation from the left with $\langle s|$ or $\langle p|$ leads to the following set of equations

$$\alpha(E_s - E) + \beta eF\langle s|z|p\rangle = 0 \quad (5.13a)$$
$$\alpha eF\langle p|z|s\rangle + \beta(E_p - E) = 0. \quad (5.13b)$$

The Stark mixing, described by the elements $\langle p|z|s\rangle$ and $\langle s|z|p\rangle$ can be calculated [3]. For our case the result is

$$\langle s|z|p\rangle = -\langle p|z|s\rangle = \sqrt{3}a_0. \quad (5.14)$$

This expression is inserted into equation (5.13), which is subsequently solved by evaluating the determinant:

$$(E_s - E)(E_p - E) - (\sqrt{3}ea_0F)^2 = 0. \quad (5.15)$$

By setting $E_p = 0$ and $E_s = \Lambda$, the problem has the solution

$$E_{\pm} = \frac{1}{2}\Lambda \pm \sqrt{\frac{1}{4}\Lambda^2 + (\sqrt{3}ea_0F)^2}. \quad (5.16)$$

This describes the Stark effects for the $^2P_{1/2}$ and $^2S_{1/2}$ states in hydrogen.

Figure 5.1 shows a graphical representation of equation (5.16), showing the energy dependence of the levels as function of electric field. It is clear from figure 5.1 that for low electric fields there is a quadratic Stark effect, which gradually changes into a linear Stark effect for high electric fields.

This type of behaviour, the transition from a quadratic to a linear Stark effect in increasing electric field, is not exclusive for these Lamb shifted levels. It is a general characteristic of the Stark effect. For small electric fields the Stark effect follows a quadratic trend, that is for fields producing Stark shifts that are smaller than the energy separation between adjacent levels in zero-field. For large fields, producing Stark shifts large compared to the energy separation of levels in zero-field, the Stark effect is linear to a good approximation. This can be intuitively explained as follows. For small electric fields, the energy levels are non-degenerate resulting in a quadratic Stark effect. For large electric fields, the Stark
shifts are so large that it appears that these levels ‘originate’ from the same, degenerate level at zero-field. The Stark effect is therefore linear.

Calculating Stark effects using perturbation theory, as presented in this section, gives a good description of the observed shifts for levels with the same principal quantum number, \( n \). However, it fails when the shifts become larger than the energy separation between levels with different \( n \), for instance for the Rydberg atoms described in the next section. For that situation a different method to solve the perturbed Schrödinger equation is needed.

## 5.3 Noble gas Rydberg atoms

An atom with a single electron in a highly-excited state is commonly known as a Rydberg atom. The other electrons remain in the lower orbits of the atom, close to the core, shielding off the charge of the nucleus. Since there is one electron in an outer orbit, there is one electron less than the charge of the nucleus close to the core. The outer electron therefore effectively sees a nucleus with a single proton charge, similar to an electron in a hydrogen atom. Therefore, Rydberg atoms behave in many ways similar to hydrogen atoms.

Highly-excited levels in hydrogen atoms can be described by the Rydberg equation

\[
E_n = I - \frac{R}{n^2},
\]

where \( I \) is the ionization energy of the atom, \( E_n \) is the energy above the ground state and \( R \) is the Rydberg constant. This constant is different for different elements; for hydrogen \( R = 1.097 \times 10^5 \text{ cm}^{-1} \).
Rydberg atoms have an energy level structure similar to hydrogen atoms. However, there will be a deviation because the electron orbits are not circular and electrons in a highly-excited orbit can penetrate the shielding cloud of electrons. The result is that the shielding of the core is no longer fully effective, and the Rydberg electron will experience a larger charge of the nucleus. A correction term, named the quantum defect, is needed to account for non-circular electron orbits. The energy level structure of Rydberg atoms can now be described by

\[ E_n = I - \frac{R}{(n - \delta)^2}, \]  

where \( \delta \) is the quantum defect. The magnitude of the quantum defect depends on the angular momentum of the Rydberg electron. States with low angular momentum have more penetrating orbits and therefore a larger quantum defect. Rydberg states with large angular momentum \((l \geq 3)\) generally have very small quantum defects.

For the heavy noble gases, the (fine)structure of atomic Rydberg states can be accurately described by the JK coupling scheme \([4,5]\). This scheme is based on a strong coupling between the orbital momentum of the Rydberg electron, \( l \), and the total angular momentum of the ion core, \( j_c \). This results in an angular momentum, \( K = j_c + l \). Additionally, the angular momentum \( K \) is weakly coupled to the spin of the Rydberg electron, \( s \), which gives the total angular momentum of the atom, \( J = K + s \). The Rydberg energy levels of the atom can now be described by wavefunctions of the form \( \varphi = |nl[K,j]⟩ \), where \( n \) is the principal quantum number of the Rydberg electron.

For Rydberg states in heavy noble gas atoms, one electron is removed from the closed electron shell configuration and moved into an excited state. The remaining ion core is in a \( 5p^5 \) configuration, which gives two possibilities for the total angular momentum of the core:

\[ j_c = l_c + s_c = \frac{3}{2} \text{ or } \frac{1}{2}. \]  

The selection rule \( \Delta j_c = 0 \) forbids transitions between excited states with different core configurations. Therefore, there exist two seemingly separate energy level structures for excited noble gas atoms; one for each core configuration. As an example, a simplified energy level scheme for xenon atoms is shown in figure 5.2. Excited states with a core configuration \( j_c = \frac{1}{2} \) are identified with \( \varphi = |nl'[K,j]⟩ \), adding a ' to the \( l \) quantum number. These states are referred to as primed states, while the states with \( j_c = \frac{3}{2} \) are known as unprimed states.

Figure 5.2 shows that for levels with the same value of \( nl \), the primed states have a higher energy. This is because, following Hund’s rules for atomic energy levels, the ionic core with \( j_c = \frac{1}{2} \) has a higher energy than \( j_c = \frac{3}{2} \). Additionally, the primed and unprimed system have different ionization limits. The result is that for the primed system, the levels
7\(p'\) and higher have an energy above the ionization limit of the unprimed system. These levels are called autoionizing levels, since they can be easily ionized in collisions with other atoms. In the following sections, discussing Stark effects in noble gas atoms, we will use transitions in the unprimed system of xenon. It should be noted that this discussion can also be applied to the primed system of xenon.

To calculate Stark effects in Rydberg atoms it is tempting to base the calculation on a description of the Stark effect in hydrogen, since the energy level structure of Rydberg atoms is similar to the hydrogen atom. However, a description of the Stark effect by perturbation theory, as in the case for hydrogen, is not possible. In Rydberg atoms there are many closely-spaced energy levels and the Stark effects can be strong due to the high principal quantum number. At only moderate electric fields, these systems show many interactions between levels, resulting in numerous anti-crossings of levels. This type of behaviour can not be described by perturbation theory and an alternative method is needed.

### 5.4 Calculation of Stark effects by matrix diagonalization

An interesting method to calculate Stark effects in Rydberg atoms was introduced by Zimmerman et al. [6]. It is based on solving the Schrödinger equation, including the
5.4. Calculation of Stark effects by matrix diagonalization

Electric field perturbation, by a numerical diagonalization of the total Hamiltonian, \( H = H_0 + H_{\text{Stark}} \), for a well-chosen truncated basis.

Firstly, the different steps of the calculation procedure are outlined. More details on each of the steps are given in the remainder of this section. The procedure for calculating Stark effects in Rydberg noble-gas atoms using matrix diagonalization follows the following successive steps.

1. Select a suitable truncated basis set of energy levels. The unperturbed Hamiltonian, \( H_0 \), is diagonal and its elements consist of the zero-field energies of the selected states.

2. The off-diagonal elements of \( H \) resulting from the Stark Hamiltonian, \( H_{\text{Stark}} = eFz \), are calculated.

3. The total Hamiltonian, \( H = H_0 + H_{\text{Stark}} \), is numerically diagonalized for different values of the electric field, \( F \), and each accessible value of the total magnetic quantum number, \( M \).

4. The energies of the Rydberg levels in an electric field correspond to the eigenvalues of the diagonalized \( H \).

5. Relative line intensities for transitions to Rydberg levels can be calculated using the eigenfunctions of the diagonalized \( H \).

5.4.1 Zero-field energy levels

The first step in the calculation procedure is the construction of the zero-field Hamiltonian, \( H_0 \). This matrix should not only include the level under study, but also all levels with energies close to this level, because of the significant interactions between the different states. In general, the following levels are included in our calculations: for all quantum numbers \( l \), one series of levels with principal quantum numbers \( n \) above, one series below and one series overlapping the level under study are included. Within a \( nl \) series, the number of possible \( K \)-values is \( 2j_c + 1 \) (or \( 2l + 1 \) if \( l < j_c \)), where \( j_c \) is the angular momentum of the ionic core. For every level there are 2 possibilities for the \( J \) quantum number, \( J = K \pm \frac{1}{2} \). This results in a large number of energy levels that need to be included in the zero-field Hamiltonian, \( H_0 \). For instance, for the calculation of Stark effects for the \( 15d[3/2]_1 \) level, there are 288 levels included in the calculation.

Including even more energy levels, further away from the level under study, did not change the calculated results. Therefore, we limited the Hamiltonian \( H_0 \) to the levels described above.
For all energy levels in $H_0$, the zero-field energies are needed. For low $l$-values most of these energies can be found in literature [7]. The energies of the remaining levels were calculated using quantum defects, found in literature, and the Rydberg equation (5.18).

For the levels with $l \geq 3$ the average energy of the multiplets was calculated using quantum defects, while the fine-structure splitting within the multiplets was calculated using the procedure of Kelleher and Saloman [8]. They describe the fine structure of the atom due to the electrostatic interaction between the ion core and the Rydberg electron by

$$\Delta E = -f_2 \langle r_c^2 \rangle \langle r^{-3} \rangle,$$

where $\langle r_c^2 \rangle$ is the mean-square radius of the ionic core, related to the quadrupole moment. For $\langle r^{-3} \rangle$ the value for the hydrogen atom is used [9]:

$$\langle r^{-3} \rangle = \frac{Z^3}{n^3(l+1)(l+\frac{1}{2})l}.$$

The angular part of the coupling, $f_2$, can be evaluated as [4]

$$f_2 = \frac{6h^2 + 3h - 2j_c(j_c + 1)l(l+1)}{4j_c(j_c + 1)(2l-1)(2l+3)},$$

with

$$h = \frac{1}{2} [K(K + 1) - j_c(j_c + 1) - l(l+1)] \equiv j_c \cdot l.$$

With these formulas the energies for the different $K$ components can be calculated. The two values of $J$ for each given $K$ are very close in energy and no splitting of these levels is assumed in zero-field.

### 5.4.2 Off-diagonal elements

The next step in the calculation procedure is the evaluation of the off-diagonal elements of $H$, given by $H_{\text{Stark}}$. For each value of the electric field, $F$, the off-diagonal matrix elements are calculated. This step, together with the subsequent diagonalization, is the heart of the calculation method.

The elements of the Stark Hamiltonian, $H_{\text{Stark}}$, describe the field coupling between two atomic levels. The elements can be calculated by evaluating the expectation value of the dipole operator $z$:

$$\langle H_{\text{Stark}} \rangle = eF \langle (nlj_c)KsJM | z | (n'l'j_c)K'sJ'M' \rangle,$$
which can be calculated using the Wigner-Eckart theorem (in atomic units),
\[
\langle (nlj_c)KsJM | z | (n'l'j_c)K'sJ'M' \rangle \\
= (-1)^{J-M} \begin{pmatrix} J & 1 & J' \\ -M & 0 & M \end{pmatrix} \langle (nlj_c)KsJ || (n'l'j_c)K'sJ' \rangle. \tag{5.25}
\]

Here \(\begin{pmatrix} j_1 & j & j_2 \\ m_1 & m & m_2 \end{pmatrix}\) is a 3\(j\)-symbol. The reduced matrix element can be further simplified using angular momentum algebra:
\[
\langle (nlj_c)KsJ || (n'l'j_c)K'sJ' \rangle \\
= (-1)^{K+s+J'+1}[(2J+1)(2J'+1)]^{1/2} \begin{pmatrix} K & J & s \\ J' & K' & 1 \end{pmatrix} \delta_{SS'} \langle (nlj_c)K || (n'l'j_c)K' \rangle. \tag{5.26}
\]

The \(\delta\) is the Kronecker symbol and the \(\begin{pmatrix} j_1 & j_2 & j_3 \\ l_1 & l_2 & l_3 \end{pmatrix}\) element is a 6\(j\)-symbol. This can be followed by
\[
\langle (nl)j_cK || (n'l')j_cK' \rangle \\
= (-1)^{l+j_c+K'+1}[(2K+1)(2K'+1)]^{1/2} \begin{pmatrix} l & K & j_c \\ K' & l' & 1 \end{pmatrix} \delta_{j_c,j_c} \langle nl || n'l' \rangle. \tag{5.27}
\]

From the Kronecker deltas and the 3\(j\) and 6\(j\) symbols the selection rules for the Stark mixing can be found:
\[
\Delta M = \Delta s = \Delta j_c = 0, \\
\Delta J = \pm 1 \quad \text{for} \quad M = 0, \\
\quad \Delta J = 0, \pm 1 \quad \text{for} \quad M \neq 0, \\
\Delta K = 0, \pm 1, \\
\Delta l = \pm 1. \tag{5.28}
\]

Finally, taking these selection rules into account, the non-zero off-diagonal matrix elements can be described by
\[
\langle (nlj_c)KsJM | z | (n'l'j_c)K'sJ'M' \rangle \\
=(-1)^{J+J'+K+K'+l+j_c+s-M}[(2J+1)(2J'+1)(2K+1)(2K'+1)]^{1/2} \\
\times \begin{pmatrix} J & 1 & J' \\ -M & 0 & M \end{pmatrix} \begin{pmatrix} l & K & j_c \\ K' & l' & 1 \end{pmatrix} \langle nl || n'l' \rangle. \tag{5.29}
\]
Chapter 5. The Stark effect

The problem now has been reduced to evaluating the reduced matrix elements \( \langle nl \| r \| n' l' \rangle \). For a one-electron system

\[
\langle nl \| r \| n' l' \rangle = \begin{cases} 
-\frac{(l + 1)^{1/2}}{} \langle nl \| r \| n' l' \rangle, & l' = l + 1 \\
\frac{1}{2} \langle nl \| r \| n' l' \rangle, & l' = l - 1
\end{cases} \tag{5.30}
\]
is valid. Finally, the radial matrix elements, \( \langle nl \| r \| n' l' \rangle \), can be found in tables calculated by Edmonds et al. [10] using the Coulomb approximation.

5.4.3 Diagonalization of total Hamiltonian

The energies of the different Rydberg states in an electric field correspond to the eigenvalues of the total Hamiltonian, \( H \). These values can be found by numerically diagonalizing matrix \( H \); the diagonal elements are the eigenvalues. Again this procedure is performed separately for each value of the electric field.

In our research, the numerical diagonalization is done with a C program, following the procedure suggested by Vetterling et al. [11]. The real symmetric matrix \( H \) is first reduced to a tridiagonal form using the routine \( \text{tred2} \) [11]. Subsequently, the routine \( \text{tqli} \) [11] finds the eigenvalues and eigenvectors of the tridiagonal matrix. Finally, the eigenvalues corresponding to the energies of the Rydberg states are saved to a file. Additionally, the resulting eigenvectors are used to calculate transition probabilities to Rydberg states, as described in the next section. It should be noted that for each value of \( F \), the numerical diagonalization of matrix \( H \) is performed for each value of the magnetic quantum number \( M \).

5.4.4 Transition probabilities

So far, we have calculated the effects of electric field on the energy level structure of Rydberg levels in atoms. These effects can be experimentally observed by studying transitions to these Rydberg levels. Of course, the probabilities of the different transitions depend on the choice of the lower level. When a certain lower level is chosen, using the eigenvectors from the matrix \( H \) we can calculate the transition probabilities from the lower level to the different Rydberg levels. With the polarization of the laser parallel to the electric field, levels with the same magnetic quantum number, \( M \), are coupled. Therefore, a summation over all transitions with \( \Delta M = 0 \) is needed. The wavefunctions of the energy levels in an electric field can be described by:

\[
\psi^{(n,M)}_{\mu} (F) = \sum_{l,K,J} Q^{(n,M)}_{\mu,l,K,J} (F) |nl[K]_{J}, M\rangle, \tag{5.31}
\]
with $\mu$ ranging from 1 to the dimension of $H$. The coefficients $Q^{(n,M)}(F)$ correspond to the eigenvectors of the matrix $H$.

The intensity of the different transitions from the lower state to Rydberg states can be expressed as

$$I_{\mu}(F) = P_L |\langle \text{lower level, } M \vert z \vert \psi_{\mu}^{(n,M)}(F) \rangle |^2,$$

(5.32)

with $\mu$ ranging from 1 to the dimension of $H$ and with $P_L$ being a coefficient that is proportional to the laser intensity but independent from the electric field $F$. In practice, the coefficient $P_L$ is not used, but the theoretical excitation spectra are normalized to the strongest transition in the wavelength range at zero electric field.

## 5.5 Stark effects calculated by multichannel quantum defect theory

The main limitation of calculating Stark effects by matrix diagonalization, as discussed in the previous section, is the assumption of a hydrogen-like atom with non-penetrating electron orbits. For large atoms and electron orbits with low $l$-values, this assumption is not valid. The atom cannot be accurately described using only hydrogenic wavefunctions and a more sophisticated method is needed.

For the Stark spectroscopy investigations presented in this thesis, the calculation method using matrix diagonalization proved to be sufficiently accurate. Therefore, this theoretical method was used for describing Stark effects. In this section we will briefly outline some of the aspects of using a more sophisticated theory such as multichannel quantum defect theory (MQDT) to describe Stark effects more accurately.

The main idea of the MQDT method is to tailor a suitable wavefunction for the Rydberg electron by dividing its orbit in three regions as shown in figure 5.3. In region 2, the hydrogenic region, the effects from the ionic core are negligible and the potential is purely a Coulomb potential. The wavefunction for this region can therefore be described by hydro-
genic wavefunctions. In region 1, the inner region, the wavefunction cannot be described by simple hydrogenic wavefunctions due to strong interactions between the ionic core and the Rydberg electron. These interactions cause perturbations and mixing between the various Rydberg states. This process can be interpreted as inelastic scattering of the Rydberg electron by the ionic core. The electron approaches the core region in a well-defined quantum state. In the core region, it exchanges energy and angular momentum with the core, after which it leaves in an altered quantum state. The multichannel quantum defect theory, developed by Seaton et al. [12], describes this scattering process in a quantum mechanical way. Finally, in region 3, effects of external electric fields are incorporated. The potential in this region is a superposition of the atomic Coulomb potential and the external electric field.

The complete wavefunction for the atom is constructed by connecting the wavefunctions of the different regions together, using suitable frame transformations. From these tailored wavefunctions, the energies of the Rydberg levels, including Stark effects, can be calculated. More details and examples of calculations of Stark effects by MQDT can be found in the work of Sakimoto [13] and Harmin [14,15].

References


Chapter 6

Investigations of Stark effects in xenon Rydberg states by laser-induced fluorescence-dip spectroscopy

Abstract. Stark effects in Rydberg states of xenon atoms were investigated both experimentally and theoretically. The experimental part consisted of laser-induced fluorescence-dip spectroscopy. Using a 2+1 photon excitation scheme, xenon atoms were excited from the ground state to high-lying Rydberg $ns$ and $nd$ levels. Measurements were made in a controllable electric field environment, produced by applying a pulsed voltage to two parallel, metal electrodes. For energy levels with principal quantum numbers ranging from 12 to 18, Stark shifts of up to 4.8 cm$^{-1}$ (160 pm) were observed for electric fields ranging from 0 to 4000 V/cm. Additionally, mixing of energy levels in high electric fields was measured for $nd$ levels. The experimental results were compared to a theoretical calculation based on solving the Schrödinger equation for a perturbed Hamiltonian. The calculation method proved to be very accurate for predicting Stark effects in Rydberg $nd$ levels, while for $ns$ levels the agreement was only moderate, probably due to deviations from the assumption of a hydrogen-like atom that is used in the calculation. Finally, the feasibility of using measurements of Stark shifts of Rydberg levels as a diagnostic for electric fields in low-pressure discharges was discussed.

6.1 Introduction

The dc Stark effect, as described in more detail in chapter 5, is characterized by shifts and splitting of energy levels of atoms or molecules under the influence of external electric fields. Especially for highly-excited Rydberg states, the Stark effect can be very pronounced, with shifts of energy levels much larger than the energy separation between levels. Over the last few decades, the Stark effect in Rydberg states has been studied extensively, both experimentally and theoretically. The first investigations studied alkali-metal atoms [1–4], while later also noble gases, including xenon, krypton, and argon, were investigated [5–8].

In gas discharge physics, the electric field distribution is an important quantity. In low-pressure discharges, it drives many fundamental processes and is closely connected to other plasma parameters such as charge densities and fluxes. Additionally, the electric field is often used as input for plasma modelling. Therefore, measurements of electric field distributions in plasmas are desirable.

Detection of Stark effects in atoms and molecules by laser spectroscopy can be used as a diagnostic for electric field strengths in plasmas. This technique was first demonstrated in BCl$_3$ [9] and helium discharges [10, 11]. Later, measurements in hydrogen and argon plasmas were performed [12, 13].

In recent years, Czarnetzki et al. [14] introduced a new method for the detection of Stark effects based on laser-induced fluorescence-dip (LIF-dip) spectroscopy. With this double-resonance technique the sensitivity of the measurements in a hydrogen discharge could be improved by an order of magnitude compared to conventional LIF methods. Recently, the use of the LIF-dip technique has been extended to argon discharges [15, 16].

In the research described in this chapter we investigated Stark effects in xenon atoms by LIF-dip spectroscopy. The goal of these investigations was twofold. Firstly, we characterized the shifts of bound Rydberg $ns$ and $nd$ levels in externally applied electric fields for principal quantum numbers ranging from 12 to 18. The experimental Stark spectra were compared with a theoretical calculation based on solving the Schrödinger equation for a perturbed Hamiltonian. Similar investigations, using a different excitation scheme, have been performed for autoionizing $ns$ and $nd$ Rydberg levels in xenon by Ernst et al. [17].

Secondly, we investigated the feasibility of using LIF-dip Stark shift measurements as an electric field diagnostic in low-pressure gas discharges. Our excitation scheme uses the ground state of the xenon atom as the lower level, which makes it possible to measure electric fields not only in plasmas, but also in neutral gas. This is a unique feature for noble gas electric field diagnostics, making investigations of discharge conditions with low degrees of ionization, such as plasma breakdown, possible. Additionally, electric fields in discharges in other gases than xenon can be performed by adding xenon to the discharge
as a trace species.

The structure of this chapter is as follows. Section 6.2 contains experimental investigations, presenting both the experimental arrangement and the results. In section 6.3 the theoretical calculation method is described and the calculation results are compared with the experiments. In section 6.4 we investigate the feasibility of using our experimental technique as an electric field diagnostic in low-pressure plasmas. Section 6.5 contains the conclusions and a short outlook.

### 6.2 Experimental investigations

In this section we present our experimental investigations on Stark effects of xenon $ns$ and $nd$ levels. Firstly, the experimental arrangement is described in section 6.2.1. Next, the results of the measurements are presented in section 6.2.2.

#### 6.2.1 Experimental arrangement

The experimental arrangement consisted of a laser system for the excitation of xenon atoms to Rydberg levels, an electrode arrangement for the application of electric fields and a detection system to monitor fluorescence light.

Stark effects in xenon were measured by probing high-lying Rydberg states by LIF-dip spectroscopy. We used a 2+1 photon excitation scheme, shown in figure 6.1, which was similar to that used by Ebata et al. [18] for investigations in NO and Czarnetzki et al. [14] for atomic hydrogen. Firstly, ground-state xenon atoms were excited to the $6p[1/2]_0$ level in a two-photon transition at 249.629 nm. Subsequently, fluorescence light with a wavelength of 828.2 nm could be observed as a result of decay to the $6s[3/2]_1$ state. A second laser was tuned to probe the transition between the intermediate $6p[1/2]_0$ level and high-lying Rydberg $ns$ and $nd$ states. A resonance for such a transition was detected as a decrease of the fluorescence intensity, because part of the $6p[1/2]_0$ level population was transferred to the Rydberg state. Therefore, when scanning the second laser and monitoring the fluorescence intensity, a transition to a Rydberg state was detected as a dip in the spectrum.

The two lasers necessary for the excitation were both tunable dye lasers, pumped by the same pulsed Nd:YAG laser. The first dye laser was pumped by the third harmonic of the Nd:YAG laser and operated at 499.258 nm. This light was subsequently frequency-doubled by a BBO crystal to create 249.629 nm photons for the two-photon transition. The second dye laser was pumped by the second harmonic output of the same Nd:YAG laser and operated at wavelengths between 580 and 630 nm.
Both beams were sent to a vacuum chamber containing a parallel-plate electrode arrangement. The two beams were parallel to the electrode surfaces and crossed each other in between the electrodes. The energies of both lasers were reduced to a few hundred $\mu$J to avoid initiating a discharge and disturbing the applied electric field.

The fluorescence light was detected perpendicular to the laser beams by an intensified charge coupled device (ICCD) camera. A lens system and an interference filter imaged the fluorescence light onto the ICCD camera, detecting only 828.2 nm light.

We applied a pulsed electric field to parallel plate electrodes, separated by 3 mm, in xenon gas at 50 Pa. The field was pulsed to prevent the formation of a discharge for voltages above the breakdown voltage of the system. Using the electrical scheme shown in figure 6.2, we could apply voltage pulses with a FWHM of about 50 ns and an amplitude of up to 1.5 kV. In this way, the electric field in the gap was controllable and uniform.

Finally, the lasers, the ICCD camera and the voltage pulse were synchronized with a delay generator.

6.2.2 Experimental results

Stark effects for $nd$ levels

We investigated Stark effects for $nd[3/2]_1$ levels with principal quantum numbers $n=12–15$ and $n=18$. Figure 6.3 shows fluorescence-dip spectra for the transition to $15d[3/2]_1$ in
different electric fields. It is clear that the dip at 586.51 nm, corresponding to the $15d[3/2]_1$ level, moves to longer wavelengths for increasing electric fields. Additionally, for an electric field of 2100 V/cm, we observe extra dips at wavelengths 586.40 and 586.28 nm. These dips correspond to transitions to $13f$ levels which are forbidden without an electric field, but become allowed when they are mixed with other levels in high electric fields.

The shifts of $nd[3/2]_1$ levels in electric field for principal quantum numbers $n=12–15$, and $n=18$ are shown in figure 6.4. For applied electric fields between 0 and 4000 V/cm, we measured Stark shifts of up to 4.8 cm$^{-1}$ (160 pm). The measurements show that the observed shift is higher for larger principal quantum numbers, as expected.

Stark effects for $ns$ levels

The shifts of $ns[3/2]_1$ levels in electric fields for principal quantum numbers $n=14–16$ are shown in figure 6.5. Applying electric fields in the range of 0 to 4000 V/cm resulted in Stark shifts of the $ns$ levels with a maximum of 0.6 cm$^{-1}$ (25 pm).

When comparing these results for $ns$ levels with the Stark shifts of the $nd$ levels with similar energies (figure 6.4), it is clear that the shifts of the $ns$ levels were much smaller.

6.3 Theoretical calculations

In this section a theoretical calculation of Stark effects, as previously discussed in chapter 5.4, is presented. Here, the calculation method and procedure are briefly described in section 6.3.1. Some representative examples of calculated results are presented in section 6.3.2. Finally, in section 6.3.3 the calculation results are compared with the experimental results from section 6.2.
6.3.1 Calculation method and procedure

A calculation method for describing Stark structures of Rydberg states in alkali-metal atoms was introduced by Zimmerman et al. [3]. This calculation method was extended by Kelleher and Saloman to describe atoms with anisotropic cores such as barium [4]. In recent years, the method, in slightly modified form, has also been applied to noble gases such as xenon [6], argon [8, 19] and krypton [20]. In these investigations the calculation of the radial matrix elements was simplified by using tables of calculations by Edmonds et al. [21] instead of calculations in the Bates-Damgaard approximation. We will also use this modified Zimmerman method for our calculations. The theoretical calculation method has been extensively described in chapter 5 and the references mentioned above. Therefore, we will only briefly point out the main features.

(i) For Rydberg levels of noble gas atoms, the JK-coupling scheme is most suitable [22]. In this scheme, the orbital angular momentum of the Rydberg electron \( \mathbf{l} \) is strongly coupled to the total angular momentum of the ionic core \( \mathbf{j}_c \). The resultant momentum \( \mathbf{K} \) is coupled to the spin of the Rydberg electron \( \mathbf{s} \), giving the total angular momentum of the atom \( \mathbf{J} \).
Figure 6.4: Shifts of $nd[3/2]_1$ levels as a function of electric field for principal quantum numbers $n=12$ (a), $n=13$ (b), $n=14$ (c), $n=15$ (d) and $n=18$ (e). The squares (■) denote the experimental results, the dots with lines (-•-) represent the results of a theoretical calculation (described in section 6.3). Figure (f) gives an overview of the shifts for all measured $nd[3/2]_1$ levels.
Chapter 6. Stark effects in xenon Rydberg states

Figure 6.5: Shifts of $ns[3/2]_1$ levels as a function of electric field for principal quantum numbers $n=14$ (a), $n=15$ (b) and $n=16$ (c). The squares ($\square$) denote the experimental results, the dots with lines (•••) represent the results of a theoretical calculation (described in section 6.3).

Energy levels can now be described by wavefunctions of the form $\varphi = |nl[K]_J\rangle$, where $n$ is the principal quantum number.

(ii) The energies of Rydberg levels in electric fields can be found by numerically solving the Schrödinger equation for an atom interacting with an electric field $\mathbf{F}$:

$$H\psi = E\psi,$$

(6.1)

with the total Hamiltonian $H$ described by

$$H = H_0 + ezF.$$  

(6.2)

Here, $H_0$ represents the Hamiltonian of the atom in zero field and $ezF$ the dipole interaction of the atom with the applied electric field. The axis $z$ is chosen to be parallel to the electric field vector.
(iii) At the start of the calculation procedure, the diagonal matrix $H_0$, of which the elements are the zero-field energies of the Rydberg levels, is constructed. Not only are the Rydberg levels involved in the laser excitation included, but also neighbouring Rydberg states, because of interactions with these levels. Most of these data can be found in literature [23], but the energies of some levels had to be calculated using the method of Kelleher and Saloman [4].

(iv) The off-diagonal elements resulting from the Stark Hamiltonian, $H_{\text{Stark}} = ezF$, are calculated for each value of the electric field. The radial matrix elements are calculated in the Coulomb approximation, using tables presented by Edmonds et al. [21]. As mentioned in reference [21], these tables are in agreement with the methods used by Zimmerman et al. [3] and Kelleher and Saloman [4].

(v) The total matrix $H = H_0 + H_{\text{Stark}}$ is numerically diagonalized.

(vi) The energies of the Rydberg levels in electric fields correspond to the eigenvalues of the total Hamiltonian $H$. The transition intensities can be calculated using the eigenfunctions of $H$.

The main approximation in this calculation procedure is that the xenon Rydberg atom is a hydrogen-like atom. This assumes non-penetrating electron orbits, which is valid for large values of $l$, but is a less appropriate approximation for small $l$ values.

It should be noted that although this calculation method involves parameters such as quantum defects and zero-field energies, we did not use adjustable parameters to fit the theory to experimental results.

### 6.3.2 Calculation results

As an example of the results of the calculation, a Stark map of energy levels around $15d[3/2]_1$ is presented in figure 6.6, showing the positions of the energy levels as a function of electric field. An excitation spectrum can be calculated from the Stark maps using the eigenfunctions of the Hamiltonian $H$. Without an electric field, the selection rules of JK-coupling only allow certain transitions. Therefore, the calculated excitation spectrum will not show transitions to all levels presented in figure 6.6, but only the transitions allowed by the selection rules. An example of a calculated spectrum for the $15d[3/2]_1$ state in different electric fields is shown in figure 6.7. The zero-field spectrum shows only the allowed transition to the $15d[3/2]_1$ state, the other allowed transitions, $15d[1/2]_1$ and $17s[3/2]_1$, are outside the plotted spectral range. It is clear that the $15d[3/2]_1$ level shifts to longer wavelengths in moderate electric fields. Additionally, in high electric fields, extra transitions which are forbidden in zero field, become allowed.
Figure 6.6: Stark map for Rydberg energy levels in xenon around the $15d[3/2]_1$ state.

Figure 6.7: Theoretical excitation spectrum of the $15d[3/2]_1$ state in different electric fields. An offset in the y scale is used to separate the different spectra. The wavelength scale corresponds to the wavelength of the second laser (figure 6.1), exciting from $6p[1/2]_0$ to Rydberg states.
6.3. Theoretical calculations

6.3.3 Comparison of theoretical and experimental results

From the theoretical spectra, such as shown in figure 6.7, the shifts of the \( nd \) and \( ns \) levels for different electric fields were determined. In figures 6.4 and 6.5, these calculated shifts are compared to the experimental shifts. It can be seen that the calculated results matched the experiments very well for the \( nd \) levels, while for the \( ns \) levels the agreement was only moderate.

These comparisons show that the relatively simple calculation procedure can very accurately describe Stark effects of \( nd \) levels in xenon Rydberg atoms. The assumption of a hydrogen-like atom with a Coulomb potential and non-penetrating orbits appears to be valid for \( nd \) levels. However, for the \( ns \) levels, the calculation gives less accurate quantitative results, probably because the assumption of non-penetrating electron orbits is not valid for these levels.

In both the experiments and the calculations, extra transitions become allowed for high applied electric fields. From our experimental results (figure 6.3), we can determine the energies of the different observed levels and compare them with theoretical Stark maps (figure 6.6). Figure 6.8 shows that the extra transitions observed were also in agreement with the calculated results. They were caused by mixing between the \( 15d[3/2]_1 \) level and levels from the \( 13f \)-manifold.

**Figure 6.8:** Theoretical Stark map for energy levels around \( 15d[3/2]_1 \) with superimposed experimentally observed transitions. The solid lines are calculations, the squares (■) indicate experimental results.
6.4 Feasibility for use as electric field diagnostic

Following the experimental and theoretical results from the previous sections, we now will discuss the possibility of using Stark shift measurements of Rydberg levels in xenon as a diagnostic for electric fields in low-pressure discharges. We present the procedure to measure electric fields and estimate the accuracy and minimum detectable electric field for our proposed diagnostic. Additionally, possible applications for the diagnostic are identified.

The investigations so far have shown that we can measure Stark shifts of Rydberg levels in xenon atoms. Furthermore, we can also accurately predict these Stark shifts for nd levels using a theoretical calculation. Therefore, the nd levels appear to be suitable as an electric field diagnostic by measuring Stark shifts in unknown electric fields. For the ns levels, the observed shifts were much smaller than for nd levels and the theoretical calculation was less accurate, therefore these levels are less suitable as electric field diagnostic.

Quantification of an unknown electric field can be done by matching observed fluorescence-dip spectra with theoretical calculations for different fields. The accuracy of such a diagnostic can be estimated from a comparison between our experiments in known electric fields and the theoretical calculation, shown in figure 6.4. In this estimation we matched theoretically calculated Stark shifts with our measured Stark shifts. The electric fields resulting from this matching were compared to the applied electric fields, which were known. This gives an estimate of the accuracy of the diagnostic for unknown electric fields. Also the minimum detectable electric field, that is the minimum detectable shift, can be estimated from figure 6.4. Obviously, these specifications are different for the different levels of xenon, because they show different Stark shifts. Table 6.1 summarizes the main specifications of the electric field diagnostic using different nd levels.

It should be noted that for n=12–15, the maximum electric field was determined by our experimental arrangement, not by limitations in the measurement method. For n=18, the maximum field was determined by decreasing signal intensities caused by mixing with neighbouring levels.

From table 6.1 it can be concluded that electric fields in the range 250–4000 V/cm can be detected with an accuracy of about 50 to 150 V/cm by measuring Stark shifts of appropriate levels. For field measurements in low electric fields (250–1000 V/cm), using the 18d[3/2]1 level is most suitable. For intermediate fields (1000–2500 V/cm), the 15d[3/2]1 state gives most accurate results. High electric fields (2500–4000 V/cm) were only measured using the 12d[3/2]1 level.

Since we have shown that our theoretical calculation is accurate for calculating Stark shifts for n=12–15 and n=18, it can be expected that it is also accurate for other, similar
6.4. Feasibility for use as electric field diagnostic

Table 6.1: Characteristics of using Stark shift measurements of different levels as an electric field diagnostic

<table>
<thead>
<tr>
<th>Energy level</th>
<th>Minimum detectable field (V/cm)</th>
<th>Measured maximum field (V/cm)</th>
<th>Estimated accuracy (V/cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>12d[3/2]_1</td>
<td>1000</td>
<td>4000</td>
<td>300–150</td>
</tr>
<tr>
<td>13d[3/2]_1</td>
<td>500</td>
<td>2500</td>
<td>150–100</td>
</tr>
<tr>
<td>14d[3/2]_1</td>
<td>300</td>
<td>2500</td>
<td>200–100</td>
</tr>
<tr>
<td>15d[3/2]_1</td>
<td>300</td>
<td>2500</td>
<td>150–75</td>
</tr>
<tr>
<td>18d[3/2]_1</td>
<td>250</td>
<td>1000</td>
<td>50–30</td>
</tr>
</tbody>
</table>

values of $n$. Therefore, in general for the measurement of high electric fields, levels with low $n$ are most suitable and for detection of low electric fields, the levels with high $n$ are best.

In many low-pressure gas discharges, electric fields of up to a few kV/cm can be expected, for instance, sheath regions of glow discharges and moving ionization fronts in plasma breakdown. For these types of discharges, measuring Stark shifts of Rydberg levels is an interesting diagnostic for electric field strengths.

An important advantage of the proposed technique is the use of the ground state of the atom as initial level for excitation. So far, electric field diagnostics for noble gas discharges used metastable atoms as lower level [15,16,19,20]. Using the ground state makes it possible to measure electric fields in neutral gases or plasmas with low degrees of ionization, such as ionization fronts in plasma breakdown.

Additionally, by using a LIF detection scheme, electric field distributions in time-varying plasmas, such as RF discharges, can be monitored. This is not possible using optogalvanic detection methods.

It should be noted that the current technique is limited to discharges containing xenon at relatively low pressure. Firstly, the xenon atoms are needed for detecting Stark shifts. However, xenon may be added to a discharge as a trace gas, extending the possible use of the technique. Additionally, the total gas pressure cannot be too high, because of collisional quenching of the intermediate $6p[1/2]_0$ level, which reduces the intensity of the fluorescence signal. Furthermore, pressure broadening will increase the width of the observed dips, making it more difficult to detect small shifts accurately. The maximum
Chapter 6. Stark effects in xenon Rydberg states

pressure for which this diagnostic can be used was not investigated in detail yet, but was estimated to be around 5000 Pa, limited by pressure-broadening leading to a dip width of about 6 cm$^{-1}$ (200 pm) [24].

6.5 Conclusions

We measured Stark effects in low-pressure xenon gas by LIF-dip spectroscopy. Using a 2+1 photon excitation scheme, starting from the xenon ground state, $nd$ and $ns$ Rydberg levels were investigated in various electric fields. For principal quantum numbers ranging from 12 to 18, Stark shifts of up to 4.8 cm$^{-1}$ (160 pm) were measured for electric fields from 0 to 4000 V/cm.

The Stark effects were also studied using a theoretical calculation. With this relatively simple calculation procedure, we could accurately calculate Stark effects in $nd$ levels. For $ns$ levels, the match with experiments was only moderate. This shows that the main assumption of the calculation, a hydrogen-like atom with non-penetrating electron orbits, is valid for calculating Stark effects of $nd$ Rydberg levels. For $ns$ levels, the deviations were probably caused by deviations from the assumption of a hydrogen-like atom. To accurately calculate Stark effects for these levels, more sophisticated calculation methods are needed.

Measurements of Stark shifts of $nd$ Rydberg levels can be used as an electric field diagnostic in low-pressure gas discharges. With our current technique we can measure electric fields from 250 to 4000 V/cm with an accuracy of about 50 to 150 V/cm. These measurements can be performed with both spatial and temporal resolution. Additionally, the use of the ground state as the lower level in the excitation scheme, makes it possible to measure electric field distributions in neutral gas and weakly ionized plasmas. Furthermore, xenon can be added to a discharge as a trace gas for diagnostic purposes.

A possible application of this diagnostic is the study of electric fields during plasma breakdown. Especially for investigations of the electric field enhancement in moving ionization fronts and the formation process of sheath structures, our proposed technique can be used. Such investigations have been performed and first results are presented in chapter 7.

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References


Chapter 6. Stark effects in xenon Rydberg states


Chapter 7

Measurements of electric field strengths in ionization fronts during plasma breakdown

Abstract. Using laser-induced fluorescence-dip Stark spectroscopy, we performed time-resolved, direct measurements of electric field strengths during the breakdown phase of a low-pressure, pulsed discharge in xenon. With this experimental technique we could for the first time quantitatively measure the time evolution of the driving force of the breakdown process, the electric field. Moving ionization fronts were measured with sub-microsecond resolution. These ionization fronts were sustained by a spatially narrow, rapidly moving region of strong electric field.

Chapter 7. Measurements of electric field strengths in ionization fronts

7.1 Introduction

In this chapter we present time-resolved, direct measurements of electric field strengths in ionization fronts during plasma breakdown. To our knowledge, this is the first time that such measurements have been performed.

Breakdown processes, including ionization fronts, have been studied in detail in many contexts in the past. For instance, the ignition phases of high-intensity discharge lamps [1], fluorescent lamps [2], plasma display panels [3], air purification with corona discharges [4] and dielectric barrier discharges [5]. However, most of these studies were modelling investigations and there are few direct measurements of the plasma properties of ionization fronts in breakdown. Experimental investigations are challenging mainly due to the highly transient nature of plasma breakdown. Additionally, the diagnostics available for direct measurements of plasma parameters such as charge densities and electric field strengths are experimentally complex. Some examples of experimental research on plasma breakdown for specific applications are cross-correlation spectroscopy of dielectric barrier discharges [6], optical and electrostatic investigations of igniting fluorescent tubes [7] and investigations of electric fields in streamers, using the Kerr effect [8]. While these are notable studies, plasma properties were obtained indirectly from measurements of emission [6, 7] or via a model interpretation [8]. There is a lack of direct experimental data on the plasma properties of ionization fronts.

The driving force of ionization fronts is the electric field. Electrons in the discharge gap gain energy in the electric field and cause ionization avalanches. Additionally, due to the electric field ions drift towards the cathode, causing secondary electron emission and new avalanches. Furthermore, the accumulated charges produced in the avalanches cause space charge fields, which modify the applied electric field, influencing the development of subsequent avalanches. Detailed knowledge of the electric field distribution, including its time evolution, is crucial for understanding the rapid, transient processes that occur during breakdown.

The aim of our research was to perform direct, quantitative measurements of electric field strengths in ionization fronts during plasma breakdown. The measurements were performed using laser spectroscopy of neutral gas atoms.

7.2 Experimental arrangement and methods

Recently, we developed an experimental method for measuring electric field distributions in low-pressure xenon gas. Our technique can be used both in neutral gas and plasmas, making it suitable to study plasma breakdown processes. Details on this diagnostic can be found in
chapter 6, here we will only describe its main features. The method is based on measuring Stark effects in Rydberg levels of atoms by laser-induced fluorescence-dip spectroscopy. It was introduced by Czarnetzki et al. [9] for investigations in atomic hydrogen. Later, it has been applied in argon discharges [10,11].

Figure 7.1 shows the excitation scheme used in our investigations. Ground state xenon atoms were excited to the $6p[1/2]_0$ state in a 2-photon transition at 249.629 nm. Using an intensified charge-coupled device (ICCD) camera with an interference filter, we observed fluorescence light at 828.2 nm resulting from the decay of $6p[1/2]_0$ to $6s[3/2]_1$ states. A second laser was tuned to probe transitions from the intermediate $6p[1/2]_0$ state to the high-lying Rydberg $15d[3/2]_1$ level. Excitation on this transition was detected as a decrease (dip) in the fluorescence intensity.

The lasers used in this excitation scheme were both tunable dye lasers pumped by the same pulsed Nd:YAG laser. The energies of the lasers were reduced to a few hundred $\mu$J to avoid saturation effects of the atomic transitions.

We studied plasma breakdown in a discharge between parabolic electrodes in low-pressure xenon gas. In chapters 3 and 4 breakdown phenomena in this discharge were investigated for an argon environment using optical emission imaging with an ICCD camera.

Both laser beams were directed to the electrode arrangement inside the vacuum vessel where they overlapped each other in between the tips of the electrodes as shown in figure 7.2. A cylindrical lens focussed the first laser, operating at 249.629 nm, to a sheet parallel to the discharge axis, with a cross section of 1.0 × 0.15 mm. The second laser, with a wavelength around 586 nm, remained unfocussed with a diameter of about 2 mm. Perpendicular to the laser beams, a lens system including an interference filter imaged the fluorescence light onto an ICCD camera. The signal from a single ICCD bin was monitored during a laser scan. In this ICCD bin, fluorescence light was accumulated from a volume of about 0.15
Chapter 7. Measurements of electric field strengths in ionization fronts

Figure 7.2: Schematic diagram of the electrode arrangement, including both laser beams. Laser beam 1 is focused into a sheet parallel to the discharge axis, while beam 2 remained unfocused. The measurement volume was determined by the focus of laser 1, the lens system imaging the fluorescence light, and the pixel size of the ICCD camera.

Figure 7.3: ICCD emission images of plasma breakdown. The top electrode was the anode, the bottom electrode the grounded cathode. Light from 7500 discharges was accumulated for each image. The white lines indicate the edges of the electrodes. The spot of light on the anode in the images $t = 40.0 \, \mu s$ was not direct plasma emission, but a reflection of the plasma around the cathode.

The discharge system consisted of two cylindrically symmetric electrodes of which the tips had a radius of curvature of 4 mm. They were mounted inside a vacuum chamber creating a 3.3 mm discharge gap. The vacuum vessel was filled with xenon gas at a pressure of 600 Pa. A pulsed discharge was created by applying voltage pulses to the electrodes. These pulses had a rise time of 30 $\mu s$, a total duration of 100 $\mu s$, a repetition rate of 500 Hz and an amplitude of 390 V, which was about 15% above the breakdown voltage. Figure 7.3 shows ICCD images of the breakdown phase of the discharge, using an exposure time of 100 ns. In these measurements all the light from the discharge in the wavelength range 350–850 nm was directly imaged onto the ICCD camera. The images show characteristic features of plasma breakdown. Firstly, a region with light emission

$0.5 \times 0.15 \, \text{mm}$ located on the discharge axis between the electrodes, 0.5 mm in front of the cathode. The measurement volume was determined by the focus of the first laser, the lens system imaging the fluorescence light, and the pixel size of the ICCD camera.
in front of the anode ($t = 26 \mu s$). Subsequently, the light emission became more intense and crossed the electrode gap ($t = 27.5 \mu s$). Finally, the discharge stabilized, covering the cathode surface ($t = 40.0 \mu s$).

The Stark effects were measured from experimental spectra by determining the shifts of the $15d[3/2]_1$ Rydberg level. These observed Stark effects were correlated to electric field strengths by comparison with a theoretical calculation as described in chapters 5 and 6. Electric field strengths for different times during the discharge pulse were measured at a fixed position inside the discharge gap, 0.5 mm in front of the cathode.

The laser scan for a single electric field measurement consisted of 8000 laser shots which took about 13 minutes. A full experiment, measuring the time evolution of electric field strengths during the breakdown phase of the discharge, lasted for about 6 hours. During this period, the timing of the discharge onset slowly drifted to a maximum offset of 1.5 $\mu$s at the end of the measurement series. To compensate for this drift, calibration of the timing of the laser experiments relative to the breakdown events was done by taking reference ICCD images of the plasma light emission before each laser scan. With this procedure an absolute timing accuracy of about 150 ns was achieved for each electric field measurement.

7.3 Results and discussion

Figure 7.4 shows the experimental results; measurements of electric fields during the breakdown phase of the discharge together with the measured electrode voltage, current and plasma light emission. These results show that we were able to measure electric field strengths between 0 and 1600 V/cm at different times during the plasma breakdown process. The resolution of the measurements was about 150 V/cm for fields above 400 V/cm. For lower fields, the Stark shifts are less sensitive to electric field therefore the resolution was lower, about 350 V/cm.

From figure 7.4 can be seen that during the first 25 $\mu$s, while the voltage was rising, there was no measurable current or plasma light emission and the electric field strength increased to about 1000 V/cm. During this phase, the applied electric field rises linearly from 0 to 980 V/cm. Therefore, our measurements indicate that during the first 25 $\mu$s of the pulse there was no discharge and the magnitude of the electric field in the discharge gap was determined by the applied potential.

In the next phase ($t = 26.0–28.0 \mu s$), the current started to develop, rising to a few mA. Figure 7.3 shows that during this phase a light front crossed the electrode gap from anode to cathode. This moving light front was in the investigations in chapter 3 identified as an ionization front.

Now for the first time, we could investigate the local electric field distribution in the
Chapter 7. Measurements of electric field strengths in ionization fronts

Figure 7.4: The top graph shows the measured voltage and current waveforms of the discharge. In the middle graph electric field strengths, measured 0.5 mm in front of the cathode, are presented. The bottom graph shows a magnification of the middle graph for times between 23 and 30 µs and the observed light emission from the discharge, originating from the same volume as the fluorescence light in the laser experiments.
gap during the crossing of this ionization front. From the measurements in figure 7.4 can be seen that the electric field in the gap first increased sharply in about 200 ns, to a value of 1600 V/cm. This was an enhancement of the electric field of about 50% compared to the applied electric field. Subsequently, the measured field decreased rapidly to 800 V/cm at $t = 27.5 \mu s$. These measurements show that the moving ionization front, observed around $t = 27.0 \mu s$, is sustained by a spatially narrow, rapidly moving region of strong electric field.

The observed enhancement of the electric field in the gap is caused by a positive space charge region in front of the anode as a result of ions produced in electron avalanches. This space charge region modifies the potential in the gap, increasing the electric field between the cathode and the space charge region and decreasing the field towards the anode. The continuing electron avalanches in the high field region cause a further extension of the space charge region into the gap. This can be observed as a moving region of enhanced electric field in front of the space charge, followed by a decrease of electric field inside the space charge region. In the high-field region, electron avalanches cause considerable ionization and excitation of atoms. This results in observable light emission since the lifetime of excited xenon is in the order of tens of nanoseconds. In the low-field region, there is less excitation and light emission. Therefore, the ionization front can be observed as a sharp light front crossing the electrode gap.

From the comparison of the timing of the electric field front and the light front, presented in the bottom graph of figure 7.4, it is clear that the electric field front was narrower and moved ahead of the light front by about 300 ns. These differences can be explained by the time needed for electrons to gain energy in the electric field, excite a xenon atom and the subsequent decay of this atom, resulting in light emission. Since these processes have durations in the order of tens to a few hundred nanoseconds, the light emission will be delayed relative to the electric field. Furthermore, due to the statistical nature of these processes, the emitted light will be spatially more spread out than the electric field front, leading to a broader light front.

From the rise time of the electric field enhancement, shown in figure 7.4, we can estimate the (average) speed of the moving ionization front to be in the order of $10^4$ m/s. This is in reasonable agreement with the speed calculated from the plasma light emission data, which gives a velocity in the order of $5 \times 10^3$ m/s. These investigations show that by measuring the time evolution of the electric field distribution in addition to the light emission we can get more detailed information on the duration and timing of the ionization front.

During the period $t = 28–40 \mu s$, after the ionization front crossed the electrode gap, the electric field stabilized at low values around 500 V/cm, while the current reached a maximum of 40 mA, as seen from figure 7.4. The uncertainties in the electric field
measurements in this region were considerable since the resolution of our diagnostic was limited in low electric fields. Nevertheless, a significant electric field was measured during this period, indicating that the discharge in the gap continued to develop. Figure 7.4 also shows that the observed light emission from the measurement volume increases sharply after the ionization front crossed the gap ($t = 28–31$ $\mu$s). This is due to the fact that the measurement volume is on the outer border of the glow region that develops around the cathode during this time, as can be seen in figure 7.3.

Figure 7.5 shows a schematic interpretation of our explanation of the effect of the first ionization front on the potential distribution in the discharge gap. Before the ionization front occurs, the potential in the gap is linear, when small deviations ($<15\%$) due to the curvature of the electrodes are neglected. The build-up of positive space charge in front of the anode leads to (partial) shielding of the anode. The space charge region develops into an ionization front that crosses the discharge gap, extending part of the anode potential further into the gap. However, the ionization front does not shield the full anode potential. After the ionization front crossed the gap, most of the potential drop is located near the cathode, in a (developing) sheath region, but there is still a significant potential difference between this sheath region and the anode. In this region the breakdown process continues and only after $t = 45$ $\mu$s, the modification of the potential in the gap is completed and a stable glow-like discharge is formed.
7.4 Conclusions and outlook

In conclusion, we have shown for the first time that it is possible to directly measure time-dependent electric field strengths in an ionization front during plasma breakdown. We measured electric fields between 0 and 1600 V/cm with a resolution of 150–350 V/cm, depending on the magnitude of the electric field. Previous measurements have shown breakdown to be characterized by an ionization front crossing the discharge gap. These new measurements show that this ionization front is sustained by a spatially narrow, rapidly moving region of strong electric field. It caused an enhancement of the electric field in the gap of about 50%, relative to the applied electric field. Our measurements show that the ionization front did not completely change the potential distribution to that of a self-sustained glow-like discharge; the development towards a steady-state situation continued.

The presented experimental technique provides a useful tool for future investigations of plasma breakdown. Possible investigations include two-dimensional, time-dependent measurements of the electric field in the electrode gap. Furthermore, by using an excitation scheme with a higher Rydberg level, we can increase the sensitivity of our technique in low electric fields as described in chapter 6. This allows a more detailed study of the processes occurring after the (first) ionization front. Finally, the technique can be used to study different discharges; for instance ignition processes in fluorescent tubes. An extension of the technique to higher pressures could open up the possibility to study electric fields in streamer discharges.

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References


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Chapter 8

Experimental and modelling investigations of a dielectric barrier discharge in low-pressure argon

Abstract. The discharge behaviour of a dielectric barrier discharge (DBD) in low-pressure argon gas was investigated by experiments and modelling. The electrical characteristics and light emission dynamics of the discharge were measured and compared with the results of a two-dimensional fluid model. Our investigations showed that the discharge consisted of a single, diffuse discharge per voltage half-cycle. The breakdown phase of the low-pressure DBD (LPDBD) was investigated to be similar to the ignition phase of a low-pressure glow discharge without dielectrics, described by Townsend breakdown theory. The stable discharge phase of the LPDBD also showed a plasma structure with features similar to those of a classical glow discharge. The presence of the dielectric in the discharge gap led to discharge quenching and thus the decay of the plasma. Additionally, the argon metastable density was monitored by measuring light emission from nitrogen impurities. A metastable density of about $5 \times 10^{17} \text{ m}^{-3}$ was present during the entire voltage cycle, with only a small ($\sim 10\%$) increase during the discharge. Finally, a reduction of the applied voltage to the minimum required to sustain the discharge led to a further reduction of the role of the dielectric. The discharge was no longer quenched by the dielectrics only, but also by a reduction of the applied voltage.

8.1 Introduction

A dielectric barrier discharge (DBD), sometimes referred to as a barrier discharge or a silent discharge, is a type of discharge in which at least one of the electrodes is covered with a dielectric material. This dielectric layer acts as a current limiter and prevents the formation of a spark or an arc discharge. The electrical energy coupled into a DBD-plasma is mainly transferred to energetic electrons, while the neutral gas remains close to ambient temperatures. The non-equilibrium plasma that is produced can be operated at elevated pressures ($10^4$–$10^6$ Pa). This combination of plasma properties makes it a unique device with many industrial applications.

DBDs have been extensively studied for over a century. Their principles have been thoroughly investigated and are described in numerous papers, for example [1–4]. Traditional industrial applications range from ozone synthesis in oxygen and air to cleaning of flue gases. Nowadays, DBDs are also used in plasma display panels, high-power CO$_2$ lasers and excimer UV/VUV lamps [3].

For most operating conditions, a DBD consists of a (large) number of discharge filaments, which have a nanosecond duration and are randomly distributed over the dielectric surface. These filaments, also known as microdischarges, are the active regions of a DBD in which active chemical species and UV/VUV radiation can be produced. These microdischarges act as individual discharges which work independently of one another. The discharge dynamics and chemistry of individual microdischarges have been studied in detail, both through modelling and experimental investigations [5–7].

In the 1980s, a different type of discharge mode in DBDs was observed [8, 9]. Under certain operating conditions, the discharge appears as a diffuse glow, covering the entire electrode surface uniformly. Since then numerous investigations have been performed to understand and explain the physical basis of this discharge mode. Several mechanisms have been discussed to explain the generation of diffuse DBDs. These include gas pre-ionization by electrons or metastables from previous discharges [10,11] and interaction between the plasma and the dielectric surfaces [12–14].

Since atmospheric pressure conditions are most suitable for many DBD applications, the research on the properties of the different discharge modes has focused mainly on atmospheric pressure conditions rather than on the low-pressure regime. However, a detailed description of the behaviour of DBDs at low pressure may contribute to a better understanding of the fundamental processes involved in DBDs. Especially the knowledge of the plasma breakdown mechanisms, including the role of the (charged) dielectric surfaces herein, can benefit from an investigation of low-pressure DBDs (LPDBDs).

The aim of the research described in this chapter is to investigate the behaviour of
a LPDBD in 400 Pa argon gas. We characterized the discharge properties by recording voltage and current waveforms and by measuring plasma emission spectrally, spatially and temporally resolved. For a better understanding of the discharge phenomena observed, we compared the experimental results with calculations from a two-dimensional fluid model.

This chapter is structured as follows. Section 8.2 contains a description of the discharge apparatus and the diagnostic system. In section 8.3, we briefly describe the two-dimensional fluid model used for our simulations. The results of the electrical and emission experiments are presented in section 8.4. In this section, these results are also compared with the results of the calculations from the two-dimensional model. Section 8.5 contains a discussion of the results, focusing on the role of the dielectric materials in the discharge behaviour. Further, a simple comparison is made between our current results and similar well-known discharges at high and low pressure. Finally, section 8.6 contains a brief summary of the main conclusions and an outlook.

8.2 Experimental arrangement

The experimental arrangement consisted of a discharge apparatus, which is described in section 8.2.1, and a set of diagnostics, described in section 8.2.2. The experimental arrangement has previously been used for cross-correlation spectroscopy on DBD-microdischarges and diffuse DBDs at atmospheric pressure [6,15].

8.2.1 Discharge apparatus

The discharge apparatus consisted of a vacuum chamber, power supply and electrode arrangement. Figure 8.1 shows a schematic diagram of the discharge apparatus. The actual DBD was created in the discharge cell. Details of this cell are presented in figure 8.2. The discharge cell was made up of two identical square (20 mm × 20 mm) electrodes, covered with dielectric material (Al$_2$O$_3$). A spacer, made of glass, connected the two electrodes together, creating a 5.0 mm discharge gap. Gas entered the discharge region through the gas inlet in the top dielectric plate. The total argon flow was 140 sccm, resulting in a gas flow in the discharge cell with a velocity of about 3 m/s, thus a laminar flow can be assumed.

The discharge cell was mounted in the centre of a vacuum vessel. The vessel was evacuated by a rotary vane pump, creating a residual pressure of about 0.5 Pa. A continuous flow of argon gas (purity 99.999%) was directed through the system. The gas flow was controlled by a flow controller, which maintained the pressure in the vessel at 400 Pa. The argon gas entering the vacuum vessel was introduced directly into the discharge volume.
Figure 8.1: Schematic diagram of the discharge apparatus.

Figure 8.2: Schematic diagram and photograph of the discharge cell. The size of the electrodes was 20 mm × 20 mm and the thickness of the Al₂O₃ dielectric layer was 0.7 mm. The spacer, determining the size of the discharge gap, had a thickness of 5.0 mm. The photograph on the right shows the discharge cell embedded in a block of plastic for insulation purposes.
A sealing ring with a nozzle on the entrance window of the vessel directed the argon flow through a plastic tube to the discharge cell.

To generate the discharge, a sinusoidal alternating voltage was applied to the electrodes. A sinusoidal waveform from a function generator was amplified first with an audio-amplifier and subsequently with an ignition coil. The resulting voltage typically had a peak-to-peak amplitude of several hundreds of volts and a frequency of 5–15 kHz. Two feedthrough windows, made of glass, were used to supply the voltage from the voltage source to the electrodes inside the vacuum chamber. The discharge cell in the chamber was embedded in a block of plastic to prevent discharges between the electrode wires and the wall of the vessel.

8.2.2 Diagnostic system

The behaviour of the discharge was studied using a diagnostic system that could characterize both the electrical properties and the light emission of the plasma.

Electrical properties

The electrical behaviour of the discharge was characterized by measuring the applied voltage and the discharge current. The high voltage applied to the electrodes was measured using a 1000:1 voltage probe. The discharge current was monitored by measuring the voltage across a 100 Ω resistor, connected in series with the discharge cell. Both waveforms were simultaneously recorded on a digital oscilloscope.

During a discharge cycle, the externally applied voltage, \( V_a \), consisted of a voltage difference across the gas gap, \( V_g \), and a voltage across the dielectric barrier plates, \( V_b \). The gap voltage, \( V_g \), and the barrier voltage, \( V_b \), could be calculated from the recorded applied voltage and current using the following relations [11]:

\[
V_g(t) = V_a(t) - V_b(t),
\]

\[
V_b(t) = \frac{2}{C_b} \int_{t_0}^{t} I_d(t')dt' + V_b(t_0),
\]

where \( C_b \) is the capacitance of a single dielectric plate, \( I_d(t') \) the recorded discharge current, \( V_b(t_0) \) the voltage due to charges on the dielectric surfaces that were left over from the previous discharge cycle and \( t_0 \) the starting time of the voltage cycle. The value of \( V_b(t_0) \) is chosen such that there is no auto-polarization. This means that the mean value of the gas voltage, \( V_g \), over a full voltage cycle is equal to 0 V. The value of \( C_b \) was calculated to be 50.6 pF, taking the relative dielectric constant for \( \text{Al}_2\text{O}_3 \), \( \epsilon_r \) equal to 10, a thickness of 0.7 mm for the dielectric plates and a size of 20 mm \( \times \) 20 mm for the electrodes.
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Figure 8.3: Optical system for one-dimensional spatially resolved emission measurements, consisting of two lenses, a slit, a stepper motor and an optical fibre. The stepper motor was used to move the slit vertically, which changed the volume in the discharge that was studied.

Plasma light emission

The aim of measuring the plasma light emission was to investigate the dynamic behaviour of the discharge. The main part of this diagnostic system was a highly sensitive photomultiplier (Hamamatsu HS5773-04) and a single-photon counting (SPC) module (Becker and Hickl SPC-530). This system was capable of detecting single photons, which made it possible to study the weak plasma emission with adequate spatial, temporal and spectral resolution. A movable lens system provided the spatial resolution, a monochromator spectral resolution and a pattern generator temporal resolution. Each of these subsystems will be discussed in more detail in the remainder of this section.

The optical system, shown in figure 8.3, was used to obtain one-dimensional spatially resolved measurements. It consisted of two lenses, a slit, a stepper motor and an optical fibre. Light emission from the discharge was imaged onto a slit by lens 1. Part of the light emission passed through the slit and was focused onto an optical fibre leading to a photon detector. The vertical position of the slit determined the volume in the plasma that was measured. The slit, together with lens 2 and the fibre, were moved vertically by a stepper motor, allowing the measurement of a one-dimensional emission profile across the discharge gap. The spatial resolution for our experiments was 0.5 mm and was determined by the width of the slit.

Plasma light emission that was focused onto the optical fibre, as described above, was sent through a monochromator. Depending on the width of the monochromator entrance slit, a spectral resolution of 0.2–1.5 nm was achieved.

After the spatial and spectral selection, the remaining signal from the plasma was very weak, consisting only of single photons per discharge cycle. A highly sensitive photomultiplier, with a gain of $10^6$ and operating in a single photon detection mode was used to detect the spatially and spectrally resolved plasma emission. The photomultiplier was cooled down to $+10 \, ^\circ C$ to increase the signal-to-noise ratio.
A temporal resolution of up to 400 ns was achieved using a computer pattern generator (Becker and Hickl, PPG-100). This device created a pattern of 512 consecutive time bins of 400 ns each, corresponding to 512 segments in the computer memory. The start of the pattern was synchronized with the applied voltage waveform, dividing a single voltage cycle into about 357 bins of 400 ns. During each measurement, the detection of a photon by the photomultiplier was stored in the corresponding segment of the computer memory. Typically, signals from up to $10^7$ discharge cycles were accumulated in the memory, which reconstructed the temporal development of the discharge during a single voltage cycle.

The SPC module was originally designed for use in cross-correlation spectroscopy. An example of the application of this technique to DBD microdischarges in atmospheric air can be found in [6]. For our measurements, the experimental setup was modified as described in [15, 16]. The SPC module was triggered by the pattern generator, which made the SPC module act as a ‘simple’ photon counter oscilloscope. Time resolution in our measurements was provided by the pattern generator and proved sufficient to resolve the dynamic behaviour of the discharge.

8.3 Two-dimensional fluid model

For comparison with experiments, we employed a time-dependent, two-dimensional fluid model. In our plasma the mean free paths for electrons and ions are about 100 $\mu$m and 25 $\mu$m respectively. These characteristic lengths are much smaller than the discharge dimensions. Therefore, the LPDBD can be described by a fluid model. Our model was originally developed for use in plasma display technology [17] and was later adapted by Brok et al. to describe breakdown phenomena in long fluorescent tubes [18]. A detailed description of the model can be found in [18] and references therein. Here, only the basic features of this model, in particular the species and reactions that were included as well as the numerical approximation of the discharge geometry, are described.

8.3.1 Model equations

Following conventional fluid models, our model was based on balance equations, derived from the Boltzmann transport equation, and on the Poisson equation. The balance equations were solved for a number of species using the drift-diffusion approximation. The species included in the model, such as electrons, ions and excited atoms, are described in more detail in section 8.3.2. The balance equations for the different species were

$$\frac{\partial n_p}{\partial t} + \nabla \cdot \Gamma_p = S_p,$$  (8.3)
with $\Gamma_p$ the drift-diffusion flux,

$$\Gamma_p = \pm \mu_p E n_p - D_p \nabla n_p,$$

(8.4)

where $n_p$ is the density of species $p$, $S_p$ the source term of species $p$ due to reactions, $\mu_p$ the mobility, $D_p$ the diffusion coefficient of species $p$ and $E$ the electric field.

In order to specify the various reaction rate coefficients and electron transport coefficients as functions of the mean electron energy, an additional balance equation for the electron energy was included:

$$\frac{\partial (n_e \bar{\varepsilon})}{\partial t} + \nabla \cdot \Gamma_{\bar{\varepsilon}} = S_{\bar{\varepsilon}},$$

(8.5)

in which $\bar{\varepsilon}$ is the average electron energy and $n_e$ the electron density. The source term of this equation, $S_{\bar{\varepsilon}}$, represents the energy gained in the electric field and the energy lost in collisions. The electron energy flux, $\Gamma_{\bar{\varepsilon}}$, is described by [19]

$$\Gamma_{\bar{\varepsilon}} = -\frac{5}{3} \mu_e E n_e \bar{\varepsilon} - \frac{5}{3} n_e D_e \nabla \bar{\varepsilon},$$

(8.6)

in which the first term on the right-hand side is the hydrodynamic flux of enthalpy and the second term the heat conduction flux. This approach differed from the commonly used local field approximation, in which these coefficients are specified as a function of the local electric field.

The actual electron transport and reaction rate coefficients were pre-calculated by a Boltzmann solver [20]. The solver calculates the electron energy distribution function at different reduced electric fields. From this, the electron transport and reaction rate coefficients were calculated as a function of the mean electron energy. The resulting data set was used to create a lookup table to be used as input for the model.

The transport coefficients for species other than the electrons and the reaction rate coefficients for heavy particle reactions were found in literature and were used as input for the model as a function of the reduced electric field. Details on the values and origins of these coefficients can be found in section 8.3.2.

Finally, in addition to the balance equations discussed above, the Poisson equation was solved:

$$\nabla \cdot (\epsilon \nabla \varphi) = -\nabla \cdot (\epsilon E) = -\sum_p q_p n_p,$$

(8.7)

in which $\varphi$ is the electric potential as a function of position and time, $\epsilon$ is the permittivity of the medium and $q_p$ the charge of the species $p$. From this equation the electric field and potential distribution in the discharge area were determined.

For each time step, all equations of the model were solved on a rectangular, uniform grid, using a control volume method [21].
### 8.3.2 Species and reactions

The set of species used in the model consisted of electrons, $e$, argon ions, $\text{Ar}^+$, molecular argon ions, $\text{Ar}_2^+$, and three effective excited states of argon atoms. The first effective excited state, called $\text{Ar}^*$, represented the four 4s levels of the argon atom. This species $\text{Ar}^*$ was assumed to be metastable because the electron impact cross section for creating a metastable 4s state is significantly larger than the cross section for the creation of a resonant 4s state. Next, all excited atoms in 4p and higher levels were grouped together in the second effective excited state, $\text{Ar}^{**}$. Finally, a third excited species, $\text{Ar}_r^*$, was used in the model. This species was introduced to take into account collisional quenching from the two metastable 4s states to the two resonant 4s levels. The species $\text{Ar}_r^*$ were lost by deexcitation to the ground level. Details about the choice of the different species used in the model can be found in [18].

The transport coefficients for the different species were taken from literature [20,22–24]. For the charged particles the diffusion coefficients were calculated from the mobilities using the Einstein relation [19]. At the dielectric walls, the incoming electrons were absorbed, charging up the wall. The incoming heavy particles were neutralized or deexcited. The resulting ground state atoms were reflected back into the plasma. Part of the incoming particles caused secondary electron emission at the dielectric wall. The secondary emission coefficients for $\text{Ar}^+$, $\text{Ar}_2^+$, and $\text{Ar}^*$ depend strongly on the properties of the dielectric surface and are not exactly known. We used values of 0.02 for $\text{Ar}^+$ and $\text{Ar}_2^+$ and 0.01 for $\text{Ar}^*$. More details on the boundary conditions of the model can be found in [18].

The set of reactions between the species that were included in the model is listed in table 8.1. Note that the reaction rate coefficients for reactions which include electrons were calculated using a Boltzmann solver.

Finally, electron-electron collisions were not included in the model. This is because inelastic collisions between electrons and the background gas were more common than electron-electron collisions since the ionization degree of the plasma under study was low.

### 8.3.3 Discharge geometry

The experimental discharge geometry is schematically shown in figure 8.4. The dashed box indicates the part of the discharge that was described by the model. Since experiments showed that at low pressures the discharge was homogeneous along the $x$ direction, we decided to only include the centre of the discharge area in the model. On this discharge area, a numerical grid in Cartesian coordinates was defined, consisting of 93 points in the $y$ direction and 15 points in the $x$ direction.

The initial condition for the electrons and the ions was a homogeneous density of
Table 8.1: Reactions included in the model. The third column contains references to the original literature from which the cross sections or rate coefficients were taken.

<table>
<thead>
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<th>No</th>
<th>Reactions</th>
<th>References</th>
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<td>0</td>
<td>Ar + e → Ar + e</td>
<td>[25]</td>
</tr>
<tr>
<td>1</td>
<td>Ar + e → Ar* + e</td>
<td>[26]</td>
</tr>
<tr>
<td>2</td>
<td>Ar + e → Ar** + e</td>
<td>[26]</td>
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<tr>
<td>3</td>
<td>Ar + e → Ar+ + 2e</td>
<td>[25]</td>
</tr>
<tr>
<td>4</td>
<td>Ar* + e → Ar + e</td>
<td>[26]*</td>
</tr>
<tr>
<td>5</td>
<td>Ar* + e → Ar** + e</td>
<td>[27]</td>
</tr>
<tr>
<td>6</td>
<td>Ar* + e → Ar+ + 2e</td>
<td>[28]</td>
</tr>
<tr>
<td>7</td>
<td>Ar** + e → Ar + e</td>
<td>[26]*</td>
</tr>
<tr>
<td>8</td>
<td>Ar** + e → Ar* + e</td>
<td>[27]*</td>
</tr>
<tr>
<td>9</td>
<td>Ar** + e → Ar+ + 2e</td>
<td>[29]</td>
</tr>
<tr>
<td>10</td>
<td>Ar* + Ar* → Ar+ + Ar + e</td>
<td>[30]</td>
</tr>
<tr>
<td>11</td>
<td>Ar* + Ar** → Ar+ + Ar + e</td>
<td>[30]</td>
</tr>
<tr>
<td>12</td>
<td>Ar** + Ar** → Ar+ + Ar + e</td>
<td>[30]</td>
</tr>
<tr>
<td>13</td>
<td>Ar* + e → Ar* + e</td>
<td>[31,32]</td>
</tr>
<tr>
<td>14</td>
<td>Ar* → Ar + hν</td>
<td>[18]</td>
</tr>
<tr>
<td>15</td>
<td>Ar** → Ar* + hν</td>
<td>[18]</td>
</tr>
<tr>
<td>16</td>
<td>Ar+ + 2Ar → Ar2+ + Ar</td>
<td>[33]</td>
</tr>
<tr>
<td>17</td>
<td>Ar2+ + e → Ar* + Ar</td>
<td>[34]</td>
</tr>
</tbody>
</table>

* From the forward reaction using microscopic reversibility.

Figure 8.4: Schematic diagram of the discharge geometry and modelling area.
$10^{13} \text{ m}^{-3}$. The metastable states also had an initial density of $10^{13} \text{ m}^{-3}$. With these initial conditions, the modelled discharge needed several voltage cycles to reach a reproducible discharge for each voltage cycle.

### 8.4 Experimental and modelling results

In this section, we present the results of our experimental and modelling investigations. Measurements of the electrical properties of the discharge and the plasma light emission are compared with calculations from our fluid model. Experiments showed that at a relatively high voltage ($700 \text{ V}_{pp}$), a transient, glow-like discharge developed. By reducing the amplitude of the applied voltage ($440 \text{ V}_{pp}$) we could observe changes in the discharge dynamics. Section 8.4.1 focuses on the properties of the transient, glow-like discharge, whereas section 8.4.2 deals with the effects of reducing the operating voltage.

#### 8.4.1 Transient glow-like discharge

A discharge in 400 Pa argon gas was created by applying a sinusoidal voltage with a frequency of 7 kHz and an amplitude of $700 \text{ V}_{pp}$. The plasma appeared to be diffuse, covering the entire electrode surface.

**Electrical properties**

An electrical characterization of the discharge is presented in figure 8.5. The applied voltage, $V_a$, and discharge current, $I$, were measured and the voltages across the gas gap, $V_g$, and the dielectric barriers, $V_b$, were calculated using equations (8.1) and (8.2). The discharge current waveform shows that the discharge consisted of a single discharge per voltage half-cycle with a duration of about 40 $\mu$s. The similar shape of the positive and negative current pulse indicated that the discharge was similar for both voltage polarities.

The barrier voltage, $V_b$, and the gap voltage, $V_g$, show that the discharge started when the amplitude of the gas voltage rose above the breakdown voltage of the discharge gap ($t = 18 \mu s$). Within a few microseconds the discharge current rose to its maximum ($t = 21 \mu s$). At this point, $V_g$ dropped slightly (about 20 V) and subsequently, over the next 30 $\mu s$, the discharge current decreased to zero, while $V_g$ remained almost constant. During this time, the increasing applied voltage amplitude led to an increase of $V_b$. After the maximum in $V_a$ ($t = 55 \mu s$), the voltage, $V_b$, remained constant, while $V_g$ dropped when the applied voltage amplitude was decreased. Next, $V_g$, changed polarity ($t = 75 \mu s$), now opposing $V_b$. Shortly after the polarity change of the applied voltage, $V_g$ reached
the breakdown voltage again and a new discharge, with opposite polarity, was initiated ($t = 91 \, \mu s$).

The electrical behaviour of the LPDBD is consistent with most of the typical characteristics of DBDs at atmospheric pressure, especially with diffuse DBDs. The discharge ignites when the voltage across the gas gap rises above the breakdown voltage. The gas in the discharge gap breaks down, a plasma is formed and the discharge current rises steeply. During the discharge, charged particles produced in the plasma are deposited on the dielectric surfaces in front of the electrodes, creating an electric field opposing the applied electric field. The resulting total electric field in the gap decreases and the discharge extinguishes. The charges remain on the dielectrics after the discharge ends and cause a residual electric field for the next discharge. After a polarity change of the applied voltage, $V_b$ now enhances the applied voltage and a discharge of opposite polarity can be initiated at relatively low applied fields.

**Spectral characterization**

For the characterization of the light emission of the discharge we recorded a time and space-integrated emission spectrum, which is shown in figure 8.6. The emitted plasma light
8.4. Experimental and modelling results

Figure 8.6: Spectrum of plasma emission. In the wavelength range from 690 to 800 nm, several spectral lines of 4p to 4s transitions in atomic argon could be identified. Further, 5p to 4s transitions in atomic argon were detected for wavelengths between 400 and 500 nm. Additionally, a weak ionic argon line was identified at 385.0 nm and a spectral band of the second positive system of nitrogen (0–0 transition) at 337.1 nm was observed.

consisted mainly of atomic argon lines. Additionally, the spectrum revealed the presence of argon ions and nitrogen molecules in the plasma. The nitrogen emission was probably due to residual gas in the chamber or impurities in the argon gas. From the spectrum in figure 8.6, three emission lines were chosen for further spatio-temporal investigation. Firstly, the strong atomic argon line at 750.4 nm was chosen to characterize the general discharge behaviour. Secondly, the ionic argon line at 385.0 nm was used to investigate ionization processes and the presence of ions during the discharge. Finally, the nitrogen band at 337.1 nm was measured to study the effect of impurities in the discharge.

Spatio-temporal structure of plasma emission

Figure 8.7 presents the results of the spatio-temporally resolved emission spectroscopy at 750.4 nm. The discharge started with some weak light emission in front of the anode ($t = 10 \mu s$). Subsequently, a fast light front crossed the discharge gap from the anode towards the cathode ($t = 18 \mu s$). The moving front had an average velocity of approximately 1700 m/s. The light front did not reach the cathode completely; it stopped at about 2 mm in front of the cathode. Next, a stable light distribution developed in the discharge gap, consisting of an emission region with a width of about 2 mm and a maximum situated at about 2 mm from the cathode ($t = 20 \mu s$). This stable discharge light distribution existed in the gap for 15 $\mu s$ after which it extinguished in the next 30 $\mu s$. In the following period ($t = 60–80 \mu s$) there was no measurable light emission. Subsequently, a new discharge started with the same characteristics as the previous one but with an opposite polarity.
An investigation of the spatio-temporal behaviour of argon ions during the discharge is presented in figure 8.8. Ionic argon line emission with a wavelength of 385.0 nm was measured. This light emission is most likely to come from ions which were excited by direct electron impact excitation. The general discharge behaviour for the ions is very similar to the behaviour of the atomic argon light emission. A light front travelled from anode to cathode with a velocity of about 1700 m/s. Subsequently, a stable discharge developed during the following 5 µs after which the ion light emission died out. One of the differences between the atomic light emission (figure 8.7) and the ionic light emission (figure 8.8) was the starting point of the moving light front. The atomic emission started at the anode surface, while the ionic emission started at about 1 mm in front of the anode. Furthermore, the ionic emission decayed faster than the atomic light and it had a maximum which was located 0.5 mm closer to the cathode than the atomic emission.

Since the lifetimes of the excited states of argon ions and atoms are in the order of nanoseconds, the measured plasma emission can be interpreted as regions of considerable excitation. Furthermore, the difference in the energy required for excitation and ionization is small for argon. Therefore, the measurements in figures 8.7 and 8.8 can be interpreted as an ionization front crossing the discharge gap during the breakdown phase, a stable glow-like discharge in the subsequent phase and a decaying plasma in the final phase. The concept of the development of a moving ionization front can also explain some of
8.4. Experimental and modelling results

Figure 8.8: Spatially and temporally resolved measurements of plasma light with a wavelength of 385.0 nm (ionic argon). The dielectric surfaces were located at positions 0 and 5 mm. The measured applied voltage and current are shown under the graph. The voltage was applied to the bottom electrode.

The differences between the ionic (figure 8.8) and atomic (figure 8.7) argon emission. The electric field strength in a moving ionization front that is crossing the discharge gap will increase with time as was shown in chapters 3 and 7. As a result, the average electron energy in the front will also increase. Because the excitation energy of the argon ions (20.0 eV) is higher than that of the argon atoms (13.3 eV), the ionic argon emission will be observed in a more developed phase of the ionization front, which is closer to the cathode.

The breakdown phase ($t = 10–20$ $\mu$s) of the discharge agrees with the well-known Townsend breakdown mechanism, which describes discharge ignition for low-pressure discharges. More details on the Townsend mechanism can be found in chapter 2 and reference [19]. Townsend theory is based on the development of electron avalanches driven by an electric field present in the discharge gap. Successive generations of electron avalanches are generated by secondary electron emission at the cathode, due to ion bombardment. Since the ion mobility is much smaller than the mobility of the electrons, a positive space-charge region develops in front of the anode. This space-charge modifies the electric field distribution in the gap, leading to the formation of a glow discharge, including features such as the cathode fall region, negative glow and Faraday dark space.

The effect of the dielectric plates was limited during the breakdown phase of the discharge, since the breakdown mechanism was equivalent to the well known low-pressure glow plasmas without dielectrics (chapters 2 and 3 of this thesis and reference [19]). But when the discharge burnt for a few microseconds, charging of the dielectrics caused discharge
extinction. This is not observed in a normal glow-discharge between metal electrodes.

The presence of nitrogen impurities in the discharge was monitored by measuring light emission from the second positive system of N$_2$ at 337.1 nm. The results of these measurements, shown in figure 8.9, reveal a different behaviour compared with the argon line emission. There was a continuous nitrogen emission in the entire discharge gap, with a maximum in the centre. During the discharge the maximum emission slightly increased (about 10%), but it did not decay completely during the decay phase of the plasma. The reason for different structures of the nitrogen light emission is most likely the excitation of the N$_2$(C) level by argon metastables. The energy of the argon metastables (11.6 eV) can be resonantly transferred to nitrogen molecules, exciting the second positive system (excitation energy 11.0 eV). Since the excited state lifetime is in the order of nanoseconds, the nitrogen emission indicates the presence of argon metastables. The metastables were produced during the discharge but remained in the discharge volume during the entire voltage cycle. The spatio-temporally resolved development shown in the figure 8.9 implies that the metastable density at the onset of the breakdown process is about 90% of the maximum density during the discharge of the previous voltage half-cycle.
8.4. Experimental and modelling results

Figure 8.10: Comparison of the modelling results with the experimental data on the discharge voltages. The top figure shows the gas voltage, $V_g$, the bottom figure shows the voltage across the barrier, $V_b$. The solid lines are experimental data, the dotted lines the modelling results.

Modelling results

The fluid model described in section 8.3 was used to simulate the LPDBD. The output of the model consisted of time-dependent, two-dimensional maps of plasma properties such as particle densities, reaction rates, potential and electric field. The full two-dimensional results showed no radial dependencies of the calculated plasma properties. Therefore, a single axial profile was used to represent the whole discharge. In figure 8.10, the model calculations of the discharge voltages are compared with the experimental measurements. The maximum in $V_g$, which corresponds to the breakdown voltage of the discharge, was calculated to be 235 V. This was within 5% of the measured breakdown voltage.

The experimental spatio-temporal investigations of the light emission of the discharge, shown in figure 8.7, were compared with the time-dependent reaction rate of reaction 15 in table 8.1. This reaction represents the decay of all excited argon atoms in levels 4p and higher into the 4s level and can be interpreted as a large part of the total light emission. In the experiments, a single 4p–4s transition was measured at 750.4 nm. The spatio-temporal
Figure 8.11: Spatio-temporal development of the calculated rate of the reaction $\text{Ar}^{*\ast} \rightarrow \text{Ar}^* + h\nu$ (table 8.1). This reaction rate can be interpreted as the main part of the total light emission (cf figure 8.7). The dielectric surfaces were located at positions 0 and 5 mm. The voltage was applied to the lower electrode in the figure, which made it the cathode during the first voltage half-cycle and the anode during the second.

The behaviour of the calculated reaction rate is presented in figure 8.11. The model predicts a single discharge per voltage half-cycle, as was seen in the experiments. Furthermore, the calculated results also show a cathode-directed moving light front and a stable discharge with maximum light emission in front of the cathode. However, in the experiments the moving light front started at the anode surface, while in the calculations it was initiated about 1 mm in front of the anode. Furthermore, the maximum in the light emission during the discharge phase (25–45 $\mu$s) was closer to the cathode in the calculations than in the experiments. These discrepancies in the starting point of the moving front and the position of the maximum of the light emission might be due to surface processes on the dielectrics, such as electron desorption and photoemission, which are not included in the model. The importance of such surface processes in the discharge behaviour of diffuse DBDs at atmospheric pressure has been established [12,14], but the effects in low-pressure DBDs are not known exactly. Electron desorption processes could provide a continuous source of secondary electrons from the surface throughout the discharge cycle. During the ignition phase, these extra electrons can lead to a higher initial charge density in the gap, which could make the developing electron avalanches stronger and the build-up of space charge faster. This can affect the build-up and starting point of the moving light front. During the discharge phase both electron desorption and photoemission processes could provide extra electrons close to the dielectric surfaces. These electrons could affect the charged particle and light distributions in the discharge gap. However, the exact mechanisms and relative importance of the different surface processes are not yet fully understood. Therefore,
8.4. Experimental and modelling results

Figure 8.12: Model calculations of the stable glow phase at \( t = 30 \, \mu s \). The figure shows the distributions in the discharge gap of the potential (a), electric field (b), \( \text{Ar}^{*+} \rightarrow \text{Ar}^* + h\nu \) reaction rate (c) and particle densities (d). The cathode is positioned at \( x = 0 \) and the anode at \( x = 5 \).

Further investigations are needed to identify which processes are important and what their exact influence on the discharge behaviour of the LPDBD will be. Finally, during the time in between two discharges (60–80 \( \mu s \)) there was no light emission at 750.4 nm measured in the experiments, while there was still some emission according to the calculations. Despite these differences the main discharge characteristics were reproduced by the model.

To investigate the plasma structure during the discharge phase, the calculated potential distribution, axial electric field, plasma emission and particle densities at the time of maximum light emission \((t = 30 \, \mu s)\) are shown in figure 8.12. The potential and electric field distributions showed the formation of a cathode fall region with a thickness of about 1.5 mm. In this region, the potential fall was about 215 V and the electric field had a maximum of 2500 V/cm. Additionally, in this region, the argon ion density was several orders of magnitude higher than the electron density, creating a positive space-charge. The
reaction rate shown in figure 8.12(c), which can be interpreted as plasma light emission, indicated that the cathode region was a region without significant excitation of argon atoms. The characteristics of this layer (x = 0–1.5 mm) are similar to the properties of a cathode sheath region in a conventional, low-pressure dc glow discharge.

The region between the positions 1–2.5 mm showed considerable light emission (figure 8.12(c)) but low electric field. These properties are similar to the negative glow region of a glow discharge. Finally, the region x = 2.5–5 mm was again a dark region, which was almost charge neutral and had a low electric field. Because the gas gap was relatively small, there was no formation of a positive column, as is the case in a dc glow in a long tube.

The calculated plasma structure during the discharge phase, presented in figure 8.12, shows several features similar to the well-known, low-pressure glow discharge. Furthermore, the breakdown phase of the LPDBD was also similar to the Townsend breakdown process, known from standard glow discharges. This indicates that the discharge behaviour of the LPDBD can be interpreted as a transient, glow-like discharge.

Figure 8.12(d) shows an almost homogeneous metastable density of about $5 \times 10^{17} \text{ m}^{-3}$ in the discharge gap. This density was present during the entire voltage cycle, with only a small increase ($\sim 10\%$) during a discharge, which is in agreement with the results shown in figure 8.9. At the start of the breakdown phase of the discharge, the metastable atoms can play an important role. Through processes such as metastable-metastable collisions (reactions (10)–(12), table 8.1), the metastables provide extra electrons that create electron avalanches and influence the behaviour of the breakdown process. In between the discharges, at time $t=70 \mu s$, ionization due to metastable-metastable collisions is about an order of magnitude higher than ionization due to electron impact.

8.4.2 Effect of reducing the applied voltage amplitude

Lowering the applied voltage of the discharge to 440 V$_{pp}$ caused changes in the discharge behaviour of the glow-like discharge mode. In the following section, we will characterize the discharge at 440 V$_{pp}$ and compare it with the discharge at 700 V$_{pp}$ as discussed in the previous section.

Electrical properties

Starting with a discharge at 700 V$_{pp}$, decreasing the voltage amplitude resulted in a continuously changing shape of the current waveform. The discharge current amplitude decreased and the discharge occurred at a later time in the voltage cycle. At the minimum voltage required to sustain the discharge (440 V$_{pp}$), the current amplitude was 60 $\mu$A and the peak
8.4. Experimental and modelling results

Figure 8.13: The left part of the figure shows measurements of the applied voltage and discharge current in the LPDBD at an applied voltage of 440 $V_{pp}$. The voltages across the dielectric plates, $V_b$, and the gas gap, $V_g$, were calculated from the measured voltage and current following the procedure outlined in section 8.2.2. The right part of the figure shows the results of the model calculations.

position was close to the maximum of the applied voltage cycle. The measured voltage and current for this minimum applied voltage case are presented in figure 8.13. Although the applied voltage amplitude was 35% lower than before, the maximum gas voltage, $V_g$, was approximately equal to the situation with high applied voltage (figure 8.5). This implies that discharge ignition occurred at the same breakdown voltage as before. However, because the breakdown voltage was reached later in the voltage cycle, there was not enough time to build up a fully developed plasma and a significant barrier voltage, $V_b$, during the discharge. As soon as $V_a$ reached its maximum value and subsequently started to decrease, the discharge could no longer be sustained and extinguished.

Spatio-temporal structure of plasma emission

Figure 8.14 shows measurements of the plasma emission of atomic argon at 750.4 nm. The discharge behaviour was significantly different from the 700 $V_{pp}$ situation presented in section 8.4.1. Again, first plasma emission was seen in front of the anode ($t = 10 \, \mu s$), but no moving, cathode-directed light front was observed. Instead, the maximum of the light emission remained in front of the anode during the entire voltage cycle. When the
voltage increased, the light emission also increased, maintaining the axial shape of the light distribution inside the gap. After the maximum in the applied voltage ($t = 38 \mu s$), the discharge extinguished in the next 20 $\mu s$.

The results of the calculations of the fluid model for the electrical characteristics are shown in figure 8.13 and for the plasma light emission in figure 8.15. The discrepancy between experiments and model in the discharge current at low currents might be due to the effects of parasitic capacitance in the measurement system.

For the spatio-temporal behaviour of the plasma emission, the model predicts maximum light emission close to the anode surface. However, in the experiments the maximum is about 1 mm closer to the anode than in the model. The discrepancy in position of the maximum of the light emission might again be due to surface processes on the dielectrics, which were not taken into account in the model.

The voltage across the dielectric barriers was small ($< 20$ V) during the entire voltage cycle. This indicates that the role of the dielectric was very limited in this situation. Charging of the dielectric was not necessary to quench the discharge since the applied voltage dropped during the discharge, leading to quenching of the discharge. The discharge behaviour is very similar to a low-pressure discharge between metal electrodes, driven by a sinusoidal voltage. The breakdown mechanism is still a Townsend breakdown. However, due to the shape of the applied voltage waveform, the electron avalanches do not create enough space-charges to disturb the potential distribution in the discharge gap significantly.
8.4. Experimental and modelling results

**Figure 8.15:** Spatio-temporal development of the calculated rate of reaction $\text{Ar}^{**} \rightarrow \text{Ar}^* + h\nu$ (table 8.1). This reaction rate can be interpreted as the main part of the total light emission. The dielectric surfaces were located at positions 0 and 5 mm. The voltage of 440 $V_{pp}$ was applied to the lower electrode in the figure, which made it the cathode during the first voltage half-cycle and the anode during the second.

Therefore, the transition to a glow-like discharge structure is prevented.

Figure 8.16 shows several plasma properties calculated by the model for the stable discharge phase ($t = 40 \mu s$). The general behaviour of the potential and electric field distribution in the gap was similar to the high-voltage (700 $V_{pp}$) case. Only in this low-voltage (440 $V_{pp}$) situation, the sheath region was wider and extended to about $x = 2.5$ mm. As a result, the plasma light emission was also positioned further from the cathode surface. However, a significant difference compared with the 700 $V_{pp}$ case in section 8.4.1 was that the entire plasma is now further from charge-neutrality. In the entire gap, the electron density was more than an order of magnitude smaller than the argon ion density.

In conclusion, when the applied voltage was decreased, the breakdown mechanism of the discharge remained the same, but the discharge structure changed. The buildup of space-charge due to electron avalanches was not large enough to significantly change the potential distribution in the gap. This prevented the discharge from developing into a glow-like structure. The discharge structure at 440 $V_{pp}$ appeared to be more similar to a Townsend discharge.

Furthermore, the influence of the dielectric plates on the discharge behaviour was reduced with decreasing voltage. At the minimum applied voltage (440 $V_{pp}$), charge build-up at the dielectric barriers did not quench the discharge; this was done by the decrease in the applied voltage waveform.
Figure 8.16: Model calculations of the discharge with $V_a=440 \ V_{pp}$ at time $t=40 \ \mu s$. Presented are the distributions in the discharge gap of the potential (a), electric field (b), $\text{Ar}^{**} \rightarrow \text{Ar}^{*} + h\nu$ reaction rate (c) and charged particle densities (d). The cathode is positioned at $x=0$ and the anode at $x=5$.

8.5 Discussion

8.5.1 Effects of dielectrics and plasma breakdown

The discharge behaviour of the LPDBD during the breakdown phase can be described by the Townsend breakdown mechanism which was developed to describe the ignition phase of low-pressure discharges between metal, parallel plates [19]. This theory is characterized by multiple electron avalanches driven by the electric field in the gap and secondary electron emission at the cathode due to ion bombardment. The discharge conditions of the LPDBD are considerably different from the standard low-pressure metal electrode discharge. The presence of the insulating, dielectric material covering the electrodes and the sinusoidal shape of the driving voltage change the discharge behaviour. However, during
the breakdown phase the mechanisms driving the ignition process are the same with or without the barriers. Previous investigations on a different discharge system, consisting of a pulsed discharge between parabolic metal electrodes in low-pressure argon gas, presented in chapter 3, also concluded that the ignition phase could be described well by Townsend theory. This indicates that the general concepts of Townsend breakdown are suitable to qualitatively describe breakdown phenomena in a wide range of low-pressure discharges.

The role of the dielectrics during the breakdown phase seems of minor importance, as the breakdown mechanism is qualitatively the same as between metal electrodes. However, the processes on the dielectric surfaces, for instance electron desorption and photoemission, are not completely understood. These additional processes might have an effect on the discharge behaviour at atmospheric pressure [12, 35], but seem to be limited at low pressures.

From more detailed studies, a voltage-controlled transition in the discharge structure could be identified. Our measurements showed that for all applied voltages that were studied, the discharge ignited with the same voltage across the discharge gap. However, the development of the breakdown process proceeded differently for different applied voltages. In the case of a high applied voltage (section 8.4.1), the discharge ignited just after a polarity change in the applied voltage. Here, the charges on the dielectric barrier from the previous discharge caused a residual electric field just below the breakdown voltage of the gap. Only a small additional, external voltage was needed to start the breakdown process. In this case, the gas voltage was sustained long enough for full discharge development. The electron avalanches during the breakdown phase created enough space-charge to change the electric field distribution in the discharge gap. This resulted in the formation of a glow-like discharge structure.

In the low-voltage case (section 8.4.2), the barrier voltage was much smaller and the applied voltage had to rise further to cause breakdown. This happened just before the maximum of the sine wave, which means that the voltage was not above the breakdown voltage long enough for full discharge development. The electron avalanches could not produce enough space-charge to distort the local electric field significantly. Therefore, the formation of a glow-like structure was prevented. The discharge was not quenched by the dielectrics but by a reduction of the applied voltage due to the sinusoidal shape.

These measurements showed that in the case of breakdown in DBDs, the discharge development depends not only on the voltage amplitude but also on the voltage shape. First of all, the voltage has to be above the breakdown voltage to start the ignition process. Secondly, the applied voltage has to be sustained long enough for the plasma to build up in multiple electron avalanches. Additionally, it has to compensate for the opposing electric field generated by particles deposited on the dielectric surfaces.
8.5.2 Comparison with other discharge types

Our experimental and modelling results showed that during the discharge a stable plasma distribution existed in the gap for several microseconds. The properties of this plasma were in good agreement with the well-known structure of a dc glow discharge between metal electrodes [19]. The LPDBD consisted of a region with high electric field similar to a cathode fall, a glow region with plasma emission resembling a negative glow layer and a dark region in front of the anode. This indicates that our discharge can be interpreted, during the stable regime, as a transient glow-like discharge.

In contrast to our low-pressure discharge, in DBDs in argon at medium and atmospheric pressures a filamentary breakdown and the formation of microdischarges are observed [7]. Under these conditions, regions with high space-charge are generated rapidly, leading to thin discharge channels, as described by the streamer breakdown process. At low pressure, a less localized space-charge region is produced in electron avalanches and the formation of a diffuse plasma is observed. The absence of regions with high, localized space-charge during the breakdown phase, as observed in the LPDBD, is also expected to play a role in the formation of diffuse plasmas at atmospheric pressures.

It can be expected that at an intermediate pressure there will be a change from the Townsend breakdown mechanism to the streamer-like breakdown process. It is currently not known how this transition occurs and at which pressures. In order to better understand the discharge, and especially the breakdown phase, measurements of this transition in breakdown mechanism are needed.

8.6 Summary

We investigated DBDs in low pressure argon gas (LPDBD) by experiments and modelling. The experimental investigations consisted of measurements of the electrical properties and the dynamics of the plasma emission of different spectral lines. In our modelling studies, we used a two-dimensional fluid model to simulate the discharge behaviour of the LPDBD.

We conclude that the breakdown phase of the discharge follows the Townsend breakdown mechanism, similar to the breakdown phase in low-pressure dc glow discharges between metal electrodes. This in contrast to the breakdown phase in argon DBDs at medium and atmospheric pressures, which have a filamentary breakdown. During the stable discharge phase, the plasma structure shows features which are also similar to those of a dc glow discharge.

The effects of the dielectric plates are limited during the breakdown phase. Only in the decay phase they cause quenching of the discharge. The influence of surface processes,
such as electron desorption and photoemission, will be investigated in the future.

At voltages close to the minimum value required to sustain the discharge, the role of the dielectric is of even less importance. The discharge is now quenched by a reduction of the applied voltage due to the sinusoidal shape, instead of quenching by charging the dielectrics.

The presence of argon metastables in the discharge was experimentally monitored through light emission from nitrogen impurities. A metastable density of about $5 \times 10^{17} \text{ m}^{-3}$ was present during the entire voltage cycle, with only a small ($\sim 10\%$) increase during the discharge. Reactions between these metastable atoms can provide extra initial electrons which can influence the starting of the breakdown process of the discharge. The exact influence of this large and constant density on the behaviour of the discharge needs to be investigated in future research.

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References


Abstract. In this final chapter, the main goals and achievements of the work described in this thesis are presented. Firstly, the conclusions from the investigations in the different chapters are summarized. Next, there is a general discussion of breakdown processes under different conditions, the effects of initial conditions and the advantages and limitations of the electric field diagnostic. Finally, the main conclusions of the thesis and a short outlook on further research are presented.
9.1 Introduction

The aim of the research described in this thesis was to obtain a better understanding of breakdown processes in low-pressure discharges. In this chapter we first give an overview of the conclusions from the different chapters. Next, there is a general discussion, linking the main results from the different chapters together. Finally, the general conclusions of this thesis and a short outlook on further investigations are presented.

9.2 Overview of the thesis

9.2.1 ICCD imaging

Breakdown in a low-pressure argon discharge between metal, parabolic electrodes was studied by time-resolved ICCD imaging. The results of these investigations, presented in chapters 3 and 4, give a clear view on the basic phenomena of plasma breakdown at low pressure. Characteristic breakdown features that were observed for a variety of discharge conditions include the formation of a light emission region in front of the anode, crossing of the electrode gap by this light front from anode to cathode and finally, a discharge region covering the cathode surface. All of these features were qualitatively in agreement with the Townsend breakdown theory for parallel-plate discharges.

The ICCD imaging investigations also revealed a pre-breakdown light emission feature. During the rise of the applied voltage, but still below the breakdown voltage, a flash of light was observed close to the anode. The origin of this pre-breakdown phenomenon was found to be electron avalanches seeded by volume charges left over from previous discharges. This phenomenon was always observed but its particular characteristics were determined by specific discharge conditions, especially the electrode geometry and the voltage pulse shape.

Finally, it was concluded that the nature of the breakdown process could be influenced by deliberately changing the initial conditions. The qualitative behaviour of the main breakdown phase of the discharge remained the same for a wide variety of discharge conditions. However, the details, especially the timing of the different phases, were influenced by the initial conditions. In chapter 3, time delay investigations show that diffusion and recombination processes in between discharges change the timing of the onset of breakdown. The investigations in chapter 4 conclude that pre-breakdown phenomena further influence the temporal development of the main breakdown process.
9.2.2 Measurements of electric field strengths

An experimental technique to quantitatively measure electric field strengths was developed (Chapter 6). Using laser-induced fluorescence-dip spectroscopy we could detect Stark effects in xenon atoms and measure local electric field strengths. Stark shifts of up to 4.8 cm$^{-1}$ (160 pm) were observed for $ns$ and $nd$ Rydberg states, with principal quantum numbers ranging from 12 to 18, as a result of electric fields between 250 and 4000 V/cm. Additionally, a relatively simple theoretical calculation proved to be very accurate for describing the observed Stark effects in $nd$ Rydberg levels. Finally, the 2+1 photon excitation scheme starting from the ground state of xenon made it possible to measure electric fields both in neutral gas and in (weakly) ionized plasmas.

Electric field distributions during the breakdown phase of a discharge between metal, parabolic electrodes in xenon were measured with this technique (Chapter 7). For the first time, direct measurements of the evolution of electric field during breakdown were obtained. Electric fields between 0 and 1600 V/cm were measured with a resolution of 200–400 V/cm, depending on the magnitude of the electric field. The experiments clearly show that the ionization front, observed by light emission, is sustained by a spatially narrow, rapidly moving region of strong electric field. A 50% enhancement of the applied electric field was measured in the region of this ionization front. Furthermore, it was concluded that the moving ionization front did not fully modify the potential in the electrode gap and the discharge continued developing towards a steady-state operation.

This electric field diagnostic proved to give very useful, new information on breakdown processes. First of all, the driving force of the breakdown process, the electric field, was measured directly, which was not possible before. Furthermore, information on local electric fields was obtained, hence the technique does not suffer from line-of-sight integration effects like the ICCD light emission imaging experiments.

9.2.3 Low-pressure dielectric barrier discharge

Breakdown processes in a second type of discharge, a dielectric barrier discharge (DBD) in low-pressure argon were investigated by different experimental techniques (Chapter 8). Electrode voltage and discharge current were measured and the light emission from the discharge was monitored spatially, temporally and spectrally resolved. Additionally, the discharge was modelled with a two-dimensional fluid model. From these investigations it was concluded that the breakdown process followed a Townsend-like mechanism, in contrast to the filamentary breakdown observed in argon DBDs at medium and atmospheric pressures. During the stable discharge phase, the plasma structure showed features that were very similar to those observed in dc glow discharges. The influence of the dielectric
plates was limited during the breakdown phase, only during the decay phase they caused discharge quenching. Finally, a high metastable density of about $5 \times 10^{17} \text{ m}^{-3}$ was observed during the entire discharge cycle. It was found that processes like electron desorption from dielectric materials and reactions between metastable atoms may provide extra initial electrons which can influence the breakdown process of the discharge. However, the detailed effects of these processes are not yet clear.

### 9.3 Breakdown processes

In this thesis plasma breakdown was studied in two different gases, argon and xenon, and two different electrode configurations, parabolic electrodes and dielectric, parallel-plate electrodes. In this section, the effects of these differences on the breakdown processes will be discussed.

The breakdown phenomena observed in both argon and xenon in discharges between parabolic electrodes were very similar. Both gases showed the characteristic features of breakdown as observed throughout this thesis. Those are, first light emission in front of the anode, crossing of this light front from anode to cathode, the discharge covering the cathode surface, further discharge development inside the electrode gap and eventually a stable discharge around the cathode.

This similarity between breakdown phenomena in argon and xenon can be explained by the similarities in the properties of the two gases. Both are heavy noble gases, which means they have a similar, closed-shell electron structure with similar excitation and ionization energies. This results in ionization coefficients, $\alpha$ (figure 2.3) and breakdown voltages (figure 1.2) that are almost equal for a large range of discharge conditions. Therefore, the qualitative breakdown behaviour will be very similar for both gases.

The small quantitative differences that can be expected include differences in operating voltage (typically 400 V for xenon and 380 V for argon) due to slightly different values of the ionization coefficient, $\alpha$, and secondary electron emission, $\gamma$. Furthermore, the timing of the breakdown phases can be different. Xenon ions have lower drift velocities because of their larger atomic mass. Therefore, the ions will drift slower towards the cathode and the generations of electron avalanches build up slower. The resulting build-up of space charge and subsequent crossing of the ionization front will take longer in the case of xenon. However, these subtle differences have not been investigated further in this thesis.

At first sight the dielectric barrier discharge (Chapter 8) seems quite different compared to the discharge between metal, parabolic electrodes. The discharge configuration is different for the two discharges, especially the electrode shape and the presence of di-
electric material in the gap. But when the breakdown phase is studied in more detail, the similarities are remarkable.

Both discharges show the characteristic features of breakdown into a glow discharge between parallel-plate, metal electrodes. The build-up of a light emission region in front of the anode, crossing of the discharge gap by this light front and the further development into a glow-like discharge structure. The observed breakdown processes can both be interpreted as being driven by multiple electron avalanches resulting from secondary electron emission by ion impact only. We find no evidence for high $\gamma$ coefficients by metastable impact as suggested recently for DBDs in noble gases [1] and nitrogen [2,3].

Similarities between the breakdown processes in different systems have also been observed before [4]. Breakdown in plasma display panel discharge cells and a compact discharge tube also showed the characteristic feature of a moving ionization front. Therefore, it seems that breakdown at low-pressure, or breakdown into a glow-like discharge, is generally characterized by multiple electron avalanches and a moving ionization front crossing the discharge gap.

It can be concluded that the fundamental, microscopic processes of the Townsend theory are sufficient to qualitatively understand breakdown for a large variety of (low-pressure) discharges. However, it remains difficult to accurately predict the details of the breakdown process such as timing of events and voltages needed for breakdown.

Modelling investigations can be very useful to obtain further insight into the quantitative details of the breakdown processes. So far, modelling efforts accurately predicted the general characteristics of the breakdown phenomena, as can be seen in chapter 8 for the low-pressure DBD and in reference [5] for the discharge between parabolic electrodes.

However, these models were unable to predict details of the breakdown process. This is mainly due to the lack of knowledge on the afterglow phase of the discharge and on processes on dielectric and metal electrode surfaces. Further investigations, both experimental and modelling, are needed to clarify these issues in order to more accurately describe breakdown processes.

9.4 Initial conditions

The initial conditions of pulsed discharges are among the most important parameters controlling the details of the breakdown behaviour, especially the timing of events. Not only the densities of particles, but also the distribution of these particles through the discharge volume proved to be important. For breakdown in the repetitively pulsed discharges studied here, the initial conditions are mainly determined by the previous discharge cycle and
the subsequent afterglow phase. Our experiments using double voltage pulses showed that the breakdown behaviour could be influenced by changing the initial conditions. Therefore, in order to control the breakdown phase of a pulsed discharge it is important to be able to control the initial conditions. To achieve this, further studies of these initial conditions and the afterglow phase of the discharge are needed.

However, such investigations are experimentally challenging since the charged and excited particle densities are low during the afterglow. Additionally, the electric fields are small and the discharge does not emit any light. Nevertheless, carefully designed experimental investigations are possible. In numerous studies, Pejović et al. and Petrović et al. [6–12] investigated memory effects of the breakdown time delay in tubes with low-pressure gas. By measuring the breakdown delay as function of the afterglow period they obtained information on the effects of the initial conditions. Additional modelling studies identified different mechanisms to be responsible for discharge ignition for different regimes of the afterglow period. In these investigations only breakdown voltages were measured directly, the processes responsible for the behaviour of the breakdown phase were not measured directly, but derived from modelling.

An interesting example of direct measurements of particle densities in an afterglow is the investigation by Nersisyan et al. [13]. They successfully measured helium metastable densities in the order of $10^{16}$ m$^{-3}$ by laser-collisional-induced fluorescence in a dielectric barrier discharge. They also note an unsuccessful attempt to measure metastable densities by multipass absorption because the densities were below the detection limit of the technique.

Since experimental investigations are challenging, modelling could be very useful for studying afterglow processes. However, to accurately describe the afterglow one must include the relevant processes for this phase. Firstly, diffusion of particles should be considered. In the early afterglow this will (most likely) be ambipolar diffusion, however during longer afterglow periods, this slowly changes to free diffusion [7]. This transition between diffusion processes should be incorporated in an accurate model. Secondly, the relevant volume reactions need to be identified. There is no clear agreement in literature which reactions are dominant under specific conditions. In our simple estimation of charge decay for the time-delay investigations in chapter 3 we followed references [14,15] and only considered atomic to molecular ion conversion and dissociative recombination of molecular ions. However, other studies identify alternative processes that can play a (dominant) role under certain discharge conditions. For instance, three-body recombination, radiative recombination, and neutral-assisted recombination [16,17]. Furthermore, not only recombination reactions should be considered, also reactions including excited species, like metastables, can play an important role. These reactions produce charged particles in collisions be-
tween metastables or collisions between metastable and neutral atoms. A careful study of all possible processes and the corresponding reaction rates is needed to identify the relevant processes for the conditions under study. Finally, surface processes, for instance surface recombination and secondary electron emission by metastables, need to be considered and taken into account accurately.

In conclusion, to obtain a better control of the breakdown behaviour it is necessary to control the initial conditions of the discharge. For this it is essential to understand the afterglow phase in more detail, both by experimental and modelling investigations.

9.5 Electric field diagnostic

The electric field diagnostic that was developed (Chapter 6) is not limited to the study of ionization fronts (Chapter 7); it can also be applied to different discharge situations. Some advantages of our diagnostic compared to other techniques measuring electric fields are the following. First of all, it is a direct measurement; there is no modelling needed for the interpretation of the measurements. The theoretical calculations in chapters 6 and 7 are only used for convenience, the experimental results from chapter 6 can serve as an experimental calibration of the technique.

Further, our technique can be applied in neutral gases and weakly ionized plasmas because of the use of the atomic ground state in our excitation scheme. Additionally, using a small amount of xenon as a trace gas is possible. Other techniques measuring Stark effects for determining electric fields often use metastable states as lower levels, which limits the applicability [18–21]. Furthermore, using a laser-induced fluorescence technique makes spatially resolved measurements possible. Alternative methods, using laser-optogalvanic detection perform line-integrated measurements and have poor temporal resolution [22–25].

Our electric field measurements can be performed with both time- and space-resolution. The time resolution is determined by the pulse length of the lasers and the spatial resolution by the imaging lens system and the binning pattern of the ICCD camera.

Of course there are also limitations to the applicability of the current technique. First of all, the discharge under study needs to be reproducible. During an experiment, the wavelength of the second laser needs to be scanned, therefore many laser shots are necessary for a full experiment. A typical experiment in this thesis took about 8000 laser shots per laser scan at a pulse repetition rate of 10 Hz. The discharge needs to be stable and reproducible during this period. This requirement can be a problem for discharges that experience jitter, like streamer discharges. It should be noted that the measurement time cannot be reduced by increasing the laser power and therefore the signal, because of
saturation effects of the atomic transitions. Using a higher repetition rate laser source or an increased efficiency of the ICCD detector can reduce the measurement time.

Secondly, xenon atoms are needed in the discharge, possibly as trace gas. This is not a fundamental requirement as long as the observed Stark effects in other atoms or molecules are large enough to be detectable and a suitable excitation scheme is available. Alternative techniques using similar excitation schemes in atomic hydrogen [26] and krypton [27] have been reported. The main challenge for extending the technique to lighter noble gas atoms is the two-photon transition exciting from the ground state. For instance, a similar scheme in argon would require a laser operating at 187 nm for the two-photon transition. This is non-trivial to achieve for practical reasons like availability of lasers in this wavelength range and absorption of laser radiation by window materials and air.

A further limitation is the pressure range in which the diagnostic can be used. This is limited to low pressures as discussed in chapter 7. Collisional quenching of the intermediate level reduces fluorescence intensity. Moreover, pressure broadening will increase the width of the observed fluorescence dips, making it more difficult to measure small shifts. The exact values of the pressure broadening depend on the total gas pressure, the type of gas or gas mixture and the atomic transition under study. For a transition to \( n \)d Rydberg states in pure xenon a gas pressure of 5 kPa is calculated to give a broadening of \( 6 \text{ cm}^{-1} \) (200 pm) [28]. However, in a gas mixture containing only a small amount of xenon, the pressure broadening can be less dominant. Pressure broadening coefficients of neon and helium are considerably smaller than for xenon [29]. Calculations by Tamida \textit{et al.} [30] show that at 66.7 kPa, pressure broadening in pure xenon is about an order of magnitude larger than in a mixture of 5% xenon and 95% neon. This indicates that with a proper choice of discharge conditions, it might be possible to use this diagnostic technique at pressures approaching atmospheric pressure. More detailed studies are needed to accurately determine the upper pressure limit for this technique.

Not only pressure broadening limits the applicability of the diagnostic, also other line broadening mechanisms can influence the diagnostic. For instance, Stark broadening which is due to collisions between the radiating atom and charged particles. For the relatively low charge densities that are typically present in low-pressure glow-like discharges (\( \approx 10^{18} \text{ m}^{-3} \)) Stark broadening can be neglected. Truong-Bach \textit{et al.} [31] report Stark broadening of typically 100 pm for transitions in xenon atoms at electron densities in the order of \( 10^{22} \text{ m}^{-3} \), which is in the same order of magnitude as the observed width of the fluorescence dip. For even higher densities (\( 10^{23} - 10^{24} \text{ m}^{-3} \)), Stark broadening can be the dominant broadening mechanism. We can conclude that for discharges with electron densities below about \( 10^{21} \text{ m}^{-3} \), Stark broadening does not limit the applicability of the electric field diagnostic.

A third significant broadening mechanism is Doppler broadening. This mechanism
scales with the square-root of the temperature divided by the mass of the radiating atoms. For a typical temperature of 600 K, the Doppler width of a transition in xenon is in the order of 1 pm, making it negligible compared to other broadening mechanisms. For different types of discharges with higher temperatures and lighter atoms, the Doppler width can be larger, but it is unlikely that it is the limiting factor for the applicability of the diagnostic.

As a final possible limitation for the electric field diagnostic we will discuss the effects of microfields in a discharge. In a quasi-neutral plasma the number of positive and negative charges is equal on a macroscopic scale. However, on a microscopic scale, there will be variations in the local charge densities resulting in positive and negative space charge regions and consequently local electric fields. These electric fields will be superimposed on the global electric field that we intend to measure, resulting in a disturbed measurement. The magnitude of the microfields can be estimated using the theory from Holtsmark [32], which shows that these fields scale with the electron density $n_e^{2/3}$. For typical low-pressure discharge conditions $n_e=10^{18}$ m$^{-3}$, the microfields are in the order of 40 V/cm. This is below the detection limit of our technique, so direct detection of these fields is not possible. However, when measuring external electric fields, these microfields will cause a broadening of the observed fluorescence dip since the microfields have a random distribution which will be superimposed onto the external electric field. This results in a distribution of electric fields that are detected. Therefore, the accuracy of the measurements will decrease.

Finally, some comments on the detection limits of the diagnostic technique. These depend first of all on the type of discharge under study and are determined by the limitations discussed before. However, the detection limits are also determined by the choice of the Rydberg level that is studied. Chapter 5 shows that Rydberg states with higher principal quantum numbers, $n$, experience larger Stark shifts for the same electric field. Since the lower detection limit is determined by the minimum detectable shift of a level, the minimum detectable electric field will be lower for higher Rydberg levels. With the calculation method for Stark effects, discussed in chapter 6, we estimated the minimum detectable field for the 25$d[3/2]_1$ level to be about 35 V/cm.

However, whether this theoretical value for the detection limit is practically achievable is questionable, because the signal-to-noise ratio of the measurement will decrease; the transition probabilities of the transitions decrease for increasing $n$. This can put a practical limit to the highest observable Rydberg state. Additionally, the maximum detectable electric field for these cases is determined by the increasing complexity of the observed spectra due to mixing of other levels with the level under study. Since the levels are closer together and the Stark shifts are larger, the maximum detectable field will decrease for increasing $n$, limiting the range of operation of this diagnostic.
9.6 General conclusions and outlook

In this thesis we investigated plasma breakdown processes in low-pressure gas discharges by experimental methods. From these investigations we conclude that the fundamental processes involved in the breakdown phases of our discharges are the same as described by Townsend’s breakdown theory for parallel-plate discharges. The general concepts of charge multiplication in multiple generations of avalanches, build-up of a space charge region in front of the anode and an ionization front crossing the discharge gap, characterize the breakdown process. However, the details of the process, especially the timing of events, are very sensitive to the specific discharge geometry and the initial conditions.

By controlling the initial conditions of the discharge, for instance by a double voltage pulse, the breakdown behaviour of the discharge can be influenced. For a better control of the initial conditions, it is necessary to have a better understanding of the afterglow phase of the discharge, since this sets the initial conditions for the next discharge. Further studies, both experimental and modelling are needed to obtain a better understanding of the processes involved in this phase. Once the afterglow phase is properly understood, a better control of the breakdown phase is possible by manipulating the initial breakdown conditions by for instance, voltage pulse shape or repetition frequency and by discharge geometry.

The electric field diagnostic that has been developed has proven to be very valuable for studying plasma breakdown phenomena. It offers the possibility to perform time- and space-resolved, direct, local measurements of the driving force of the breakdown process; the electric field. These investigations showed that the ionization front observed during breakdown, is sustained by a spatially narrow, rapidly moving region of strong electric field. It caused an enhancement of the electric field in the gap of about 50%, relative to the applied electric field. Furthermore, it was concluded that the breakdown process continued to develop after the ionization front crossed the gap; the potential distribution in the gap was not fully modified by the ionization front.

The use of the electric field diagnostic is not limited to breakdown studies in low-pressure xenon discharges. Application of the technique to different discharge geometries is straightforward, for instance breakdown studies in fluorescent lamps. By using xenon as trace gas, also investigations in different gases, for instance argon or air, are possible. Finally, an extension of the technique to higher pressures could open up the possibility to study electric fields in streamer discharges and dielectric barrier discharges. Breakdown phenomena under these conditions are only poorly understood, and quantitative measurements are needed to further develop the understanding of these breakdown processes.
References


Chapter 9. General discussion and conclusions


Natural gas discharges like lightning and polar light are spectacular phenomena that have impressed and fascinated people for a long time. During the last two centuries, people have learned how to create their own gas discharges and how to make use of them. Nowadays, man-made gas discharges are commonly used in many applications. Well-known examples include fluorescent lamps, plasma televisions and high intensity lamps used in data projectors. Also in industry, gas discharges are often part of the production process, for instance in the fabrication of computer chips, cleaning of exhaust gases, coating of fabrics and production of solar cells.

A gas discharge, or plasma, consists of a variety of different particles like electrons, ions, atoms and molecules in various excited states. These particles not only interact with each other, but also with electric and magnetic fields and with electrode and wall surfaces in the discharge volume. This wide variety of particles and interactions makes a gas discharge a complex system, which is difficult to understand completely and control accurately.

To create a gas discharge one generally needs to apply a sufficiently large voltage across a volume of neutral gas. There are many options how to do this, resulting in many different types of gas discharges. Nevertheless, the basic principles involved are the same for all these situations. The electrically insulating, neutral gas is transformed into a conducting, (partially) ionized state by the applied voltage. This evolution from a neutral gas to a self-sustaining discharge is known as plasma breakdown, or plasma ignition, and is the subject of this thesis.

Plasma breakdown is a fundamental process in gas discharges; it is a highly transient process that involves particles drifting in electric fields, charge multiplication in electron avalanches and moving ionization fronts. The driving force for these processes is the electric field in the discharge volume. The research in this thesis was aimed at obtaining a better understanding of the fundamental processes involved in plasma breakdown in low-pressure discharges by experimental investigations.

Two types of discharges were studied; a pulsed discharge between parabolic, metal electrodes and a parallel-plate, low-pressure dielectric barrier discharge. The breakdown
phases of these discharges were investigated using various experimental techniques.

Breakdown processes in the low-pressure dielectric barrier discharge were investigated by studying the light emission from the discharge in a spatially, temporally and spectrally resolved way. Additionally, electrode voltages and discharge currents were measured. These investigations, together with the results from a two-dimensional fluid model, showed that the breakdown process in this discharge followed a Townsend-like mechanism in which the effects of the dielectric plates were limited.

The pulsed discharge between parabolic, metal electrodes, was firstly studied in a low-pressure argon environment by light emission imaging with an intensified charge-coupled device (ICCD) camera. This relatively simple diagnostic provided time- and space-resolved information on the characteristic features of the breakdown process. Different phases in the breakdown process were identified. Firstly, the build-up of a light emission region in the discharge gap in front of the anode, followed by a light front crossing the electrode gap from anode to cathode and finally, a stable discharge covering the cathode surface. These features were in qualitative agreement with the breakdown process observed in parallel-plate discharges at low pressure, which are accurately described by Townsend’s breakdown theory.

The ICCD imaging experiments also showed that before the main breakdown process started, a weak flash of light could be observed around the anode. This pre-breakdown light emission occurred during the rise of the applied voltage, but before the breakdown voltage was reached. The origin of this feature was found to be electron avalanches seeded by volume charges left over from previous discharges in combination with the specific discharge geometry used in our experiments. It was concluded that the initial conditions of the discharge influenced the breakdown process. Although the qualitative behaviour of the main breakdown phase did not change for a wide variety of discharge conditions, the details of the process, especially the timing of the different phases were strongly influenced by the initial conditions.

Finally, a new diagnostic was developed to measure electric field distributions during the breakdown phase of a discharge. With this diagnostic, electric field strengths were determined by measuring Stark effects in xenon atoms using laser-induced fluorescence-dip spectroscopy. Stark shifts of up to $4.8 \text{ cm}^{-1}$ (160 pm) were observed for ns and nd Rydberg states, with principal quantum numbers ranging from 12 to 18, as a result of electric fields between 250 and 4000 V/cm. Additionally, a theoretical calculation method, based on solving the Schrödinger equation for a perturbed Hamiltonian by matrix diagonalization, proved to be very accurate for describing the observed Stark effects in nd Rydberg levels.

With this diagnostic we performed measurements of the electric field distribution during the breakdown phase of the discharge between parabolic electrodes in xenon. For the first
time, quantitative, direct measurements of the evolution of electric field during breakdown were obtained. Electric fields between 0 and 1600 V/cm were measured with a resolution of 200–400 V/cm, depending on the magnitude of the electric field. These experiments showed that the ionization front, already observed in the ICCD imaging experiments, is sustained by a spatially narrow, rapidly moving region of strong electric field. Additionally, this ionization front did not completely modify the potential distribution in the discharge gap; the discharge continued developing towards a steady-state after the ionization front crossed the gap.

In conclusion, the investigations in this thesis show that the fundamental processes involved in plasma breakdown are well understood. However, the details of the process, especially timing of events, depend strongly on the specific discharge geometry and the initial discharge conditions. The electric field diagnostic has proven to be very useful for breakdown studies, identifying a narrow, moving region of high electric field as the cause of the moving ionization fronts. This diagnostic offers the possibility to obtain quantitative, direct information on local electric field strengths with both spatial and temporal resolution. In future research, such electric field measurements can be very valuable not only for investigations on low-pressure breakdown but also for studies on breakdown processes at higher pressures, which are only poorly understood at the moment.
Samenvatting

Natuurlijke gasontladingen zoals bliksem en het noorderlicht zijn spectaculaire fenomenen die de mens al jarenlang imponeren en fascineren. Gedurende de afgelopen tweehonderd jaar hebben mensen geleerd hoe ze zelf gasontladingen kunnen maken en hoe ze deze kunnen gebruiken. Tegenwoordig worden gasontladingen veelvuldig gebruikt in verscheidene toepassingen. Bekende voorbeelden hiervan zijn zijn TL lampen, plasma televisies en hoge intensiteit lampen gebruikt in beamers. Ook in een industriële omgeving worden gasontladingen veel gebruikt als onderdeel van het productieproces, bijvoorbeeld bij het maken van computerchips, het reinigen van afvalgassen, het aanbrengen van beschermende lagen op textiel en de productie van zonnecellen.

Een gasontlading, vaak ook plasma genoemd, bevat meerdere verschillende deeltjes zoals elektronen, ionen, moleculen en atomen in verschillende aangeslagen toestanden. Deze deeltjes hebben niet alleen een wisselwerking met elkaar, maar ook met elektrische en magnetische velden en de oppervlakken van elektrodes en wanden in het ontladingsvolume. Deze verscheidenheid in deeltjes en interacties maken gasontladingen tot complexe systemen, die moeilijk volledig te begrijpen en nauwkeurig te controleren zijn.

Om een gasontlading te creëren moet men een voldoende grote elektrische spanning aanbrengen over een volume met neutraal gas. Er zijn vele verschillende mogelijkheden om dit te doen, wat betekent dat er ook vele verschillende types gasontladingen zijn. Niettemin zijn de fundamentele processen voor al deze situaties dezelfde. De aangelegde spanning zet het elektrisch isolerende, neutrale gas om in een geleidende, (gedeeltelijk) geioniseerde toestand. Dit proces, de verandering van een neutraal gas naar een stabiele ontlading staat bekend als plasma ontsteking en is het onderwerp van dit proefschrift.

Het ontsteken van plasma’s is een fundamenteel proces in gasontladingen. Het is een zeer snel fenomeen dat gekenmerkt wordt door processen zoals bewegende deeltjes in een elektrisch veld, ladingsvermenigvuldiging in elektronenlawines en bewegende ionisatiefron ten. De drijvende kracht achter deze processen is het elektrisch veld in het ontladingsvolume. Het doel van het onderzoek in dit proefschrift was het beter begrijpen van de fundamentele processen die betrokken zijn bij het ontsteken van ontladingen. Dit is ge-
daan door middel van experimenteel onderzoek.

In dit proefschrift zijn twee verschillende ontladingen bestudeerd; een gepulste ontlading tussen parabolische, metalen elektrodes en een lage-druk, diëlektrische-barrière ontlading tussen parallelle, vlakke platen. De ontsteekfasen van deze ontladingen zijn onderzocht met behulp van verschillende experimentele technieken.

 Ontsteekprocessen in de lage-druk diëlektrische-barrière ontlading zijn onderzocht door middel van het bestuderen van het licht dat wordt uitgezonden door de ontlading. Dit werd zodanig gedaan dat ruimtelijke, tijds en spectrale resolutie verkregen kon worden. Ook de ontladingsstroom en aangelegde spanning werden gemeten. Deze onderzoeken, in combinatie met de resultaten van een twee-dimensionaal vloeistof model, tonen aan dat de ontsteekprocessen in deze ontlading een mechanisme volgen dat lijkt op ontsteking volgens de Townsend theorie. Deze theorie geeft een nauwkeurige beschrijving van het ontlsteekproces van een ontlading tussen metalen, vlakke elektrodes. De effecten van de diëlektrische materialen in onze ontlading waren beperkt tijdens de ontsteekfase.

De lichtemissie van de gepulste ontlading tussen parabolische, metalen elektrodes werd bestudeerd in een lage-druk argon omgeving met behulp van een versterkte CCD camera (ICCD). Deze relatief eenvoudige diagnostiek geeft ruimtelijk en tijdsopgeloste informatie over de kenmerkende eigenschappen van het ontsteekproces. Verschillende fasen in het ontsteekproces konden worden onderscheiden. Ten eerste, het ontwikkelen van een gebied met lichtemissie tussen de elektrodes, vlak voor de anode, vervolgens een lichtfront dat oversteekt van de anode naar de kathode en ten slotte, een stabiele ontlading die de kathode bedekt. Deze kenmerken zijn in kwalitatieve overeenstemming met de ontsteekprocessen in een ontlading tussen parallelle platen bij lage druk zoals die door de ontstekingstheorie van Townsend nauwkeurig worden beschreven.

De emissie experimenten met behulp van de ICCD camera toonden ook aan dat er een zwakke lichtflits te zien was in de buurt van de anode, voordat de echte ontsteking van de ontlading startte. Deze pre-ontstekings lichtemissie was te zien tijdens het stijgen van de aangelegde spanning, maar voordat de ontsteekspanning daadwerkelijk bereikt werd. Verdere emissiemetingen lieten zien dat dit fenomeen veroorzaakt wordt door geladen deeltjes die overgebleven zijn van vorige ontladingen, in combinatie met de specifieke geometrie van de ontlading in onze experimenten. We hebben geconcludeerd dat de begincondities van de ontlading het ontsteekproces beïnvloeden. Ondanks het feit dat het ontlsteekgedrag kwalitatief hetzelfde was voor een variëteit van ontladingscondities, werden de details van het ontsteekproces, in het bijzonder de timing van de verschillende fasen, sterk beïnvloedt door de begincondities.

Tenslotte hebben we een nieuwe diagnostiek ontwikkeld voor het meten van elektrische velden tijdens de ontsteekfase van een ontlading. Deze diagnostiek bepaalt elek-
trische veldsterktes uit metingen van Stark effecten in xenon atomen, met behulp van laser-geïnduceerde fluorescentie-dip spectroscopie. We hebben Stark verschuivingen van maximaal 4.8 cm$^{-1}$ (160 pm) gemeten voor $ns$ en $nd$ Rydberg niveaus met hoofdquantum-getallen tussen 12 en 18, als gevolg van elektrische velden van 250 tot 4000 V/cm. Een theoretische berekeningsmethode bleek de geobserveerde Stark effecten in de $nd$ Rydberg niveaus nauwkeurig te kunnen beschrijven. Deze methode is gebaseerd op het oplossen van de Schrödinger vergelijking met een verstoorde Hamiltoniaan door middel van matrixdiagonalisatie.

Met deze diagnostiek hebben we metingen uitgevoerd van de elektrische velden tijdens de ontsteekfase van de ontlading tussen parabolische elektrodes. Voor het eerst hebben we kwantitatieve, directe metingen van de tijdsontwikkeling van het elektrisch veld tijdens ontsteking kunnen uitvoeren. Elektrische velden tussen 0 en 1600 V/cm zijn gemeten met een nauwkeurigheid van 200 tot 400 V/cm, afhankelijk van de grootte van het veld. Deze experimenten toonden aan dat het ionisatiefront, zoals gemeten in de ICCD emissie experimenten, veroorzaakt wordt door een smal, snel bewegend gebied van hoge elektrische veldsterkte. Verder hebben we aangetoond dat het ionisatiefront niet de volledige potentiële verdeling tussen de elektrodes veranderde van een lineaire verdeling naar die van een glimontlading. Het ontsteekproces van de ontlading werd vervolgd na het oversteken van het ionisatiefront.

De conclusie van dit proefschrift is dat de fundamentele processen die betrokken zijn bij plasma ontsteking goed begrepen zijn. Echter, de details van het ontsteekproces, in het bijzonder de timing van de verschillende fasen, worden sterk beïnvloed door de specifieke geometrie en de begincondities van de ontlading. De elektrische veld diagnostiek is erg waardevol gebleken voor onderzoek naar ontsteekgedrag van ontladingen. Een smal, bewegend gebied van hoge elektrische veldsterkte bleek de drijvende kracht te zijn achter de bewegende ionisatiefronten die in de ontlading te zien waren. Deze diagnostiek biedt de mogelijkheid om kwantitatieve, directe informatie te verkrijgen over lokale elektrische veldsterktes met ruimtelijke en tijdsresolutie. In verder onderzoek kunnen dergelijke elektrische veldmetingen zeer waardevol zijn, niet alleen voor de bestudering van lage-druk ontsteking, maar ook voor studies naar ontsteekprocessen bij hogere drukken. Deze zijn tot op heden nog niet volledig begrepen.
Related publications

Journal papers

*Investigations of Stark effects in xenon Rydberg states by laser-induced fluorescence-dip spectroscopy*
E. Wagenaars, G.M.W. Kroesen and M.D. Bowden

*Pre-breakdown light emission phenomena in low-pressure argon between parabolic electrodes*
E. Wagenaars, N.W.B. Perriëns, W.J.M. Brok, M.D. Bowden, E.M. van Veldhuizen and G.M.W. Kroesen

*Experimental and modelling investigations of a dielectric barrier discharge in low-pressure argon*
E. Wagenaars, R. Brandenburg, W.J.M. Brok, M.D. Bowden and H.-E. Wagner

*Measuring electric fields with laser-induced fluorescence-dip Stark spectroscopy*
E. Wagenaars, M.D. Bowden and G.M.W. Kroesen
High Temperature Material Processes, accepted for publication, 2006.

*Plasma emission imaging of a low-pressure argon breakdown*
E. Wagenaars, M.D. Bowden and G.M.W. Kroesen

*Plasma emission imaging of electrical breakdown in low-pressure argon*
E. Wagenaars, M.D. Bowden and G.M.W. Kroesen
Measurements of electric field strengths in ionization fronts during plasma breakdown
E. Wagenaars, M.D. Bowden and G.M.W. Kroesen

Diagnostics of electric fields in plasma using Stark spectroscopy of krypton and xenon atoms
T. Jiang, M.D. Bowden, E. Wagenaars, E. Stoffels and G.M.W. Kroesen

Emission imaging of a PDP-like microdischarge
T. Jiang, M.D. Bowden, E. Wagenaars and G.M.W. Kroesen

Measurements of plasma breakdown
M.D. Bowden, E. Wagenaars, T. Jiang, W.J.M. Brok and M.F. Gendre

A study of Stark effects of Rydberg p states of noble gas atoms
T. Jiang, M.D. Bowden, E. Wagenaars, E. Stoffels and G.M.W. Kroesen

Numerical description of pulsed breakdown between parabolic electrodes
W.J.M. Brok, E. Wagenaars, J. van Dijk and J.J.A.M. van der Mullen

Conference contributions

Electric field measurements in moving ionization fronts during plasma breakdown
E. Wagenaars, M.D. Bowden and G.M.W. Kroesen
59th Gaseous Electronics Conference, Columbus (United States), 10-13 October 2006.

Measuring electric fields in low-pressure xenon plasmas by fluorescence-dip Stark spectroscopy
E. Wagenaars, M.D. Bowden and G.M.W. Kroesen
High Technology Plasma Processes 9, St. Petersburg (Russia), 27 May-4 June 2006.
Electric field measurements by fluorescence-dip Stark spectroscopy
E. Wagenaars, M.D. Bowden and G.M.W. Kroesen
58th Gaseous Electronics Conference, San Jose (United States), 16-20 October 2005.

Experimental study of breakdown in low-pressure argon between parabolic electrodes
E. Wagenaars, N.W.B. Perriëns, M.D. Bowden and G.M.W. Kroesen
58th Gaseous Electronics Conference, San Jose (United States), 16-20 October 2005.

Measurement of electric field strengths by fluorescence-dip Stark spectroscopy
E. Wagenaars, M.D. Bowden and G.M.W. Kroesen

Low-pressure dielectric barrier discharges in argon: experimental investigations and modelling
E. Wagenaars, R. Brandenburg, W.J.M. Brok, M.D. Bowden and H.-E. Wagner

Electric field measurements in a xenon discharge using Stark spectroscopy
E. Wagenaars, M.D. Bowden and G.M.W. Kroesen

Optical imaging of breakdown in a low-pressure argon discharge
E. Wagenaars, M.D. Bowden and G.M.W. Kroesen

Study of plasma breakdown by Stark spectroscopy
E. Wagenaars, M.D. Bowden and G.M.W. Kroesen
XXVI International Conference on Phenomena in Ionized Gases, Greifswald (Germany), 15-20 July 2003.

Experimental arrangement for the study of plasma breakdown
E. Wagenaars, M.D. Bowden and G.M.W. Kroesen
Frontiers in Low Temperature Plasma Diagnostics V, Villaggio Cardigliano (Italy), 3-7 April 2003.
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Erik Wagenaars,
Oss, 22 september 2006.
Curriculum Vitae


1991–1997  Pre-university education (atheneum) at Mondriaan College, Oss.


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Traineeship at the European Laboratory for Particle Physics (CERN) in Geneva, Switzerland. Subject: *The CsI evaporation unit for the HMPID photocathodes*, supervised by dr. E. Schyns and prof.dr. M.J.A. de Voigt.

Master thesis project at the Elementary Processes in Gas Discharges group of the Department of Applied Physics, Eindhoven University of Technology. Subject: *Analysis of the effects of plasma treatment on bone tissue*, supervised by prof.dr.ir. G.M.W. Kroesen.


Research visit to the Institut für Physik, Ernst-Moritz-Arndt Universität in Greifswald, Germany. Subject: *Experimental investigations of a dielectric barrier discharge in low-pressure argon.*