Multi-fluid modeling of transient plasmas: a case study in the generation and guiding of light

Citation for published version (APA):

DOI:
10.6100/IR610194

Document status and date:
Published: 01/01/2006

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:
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Multi-fluid Modeling of Transient Plasmas:
a Case Study in the Generation and Guiding of Light

PROEFSCHRIFT

ter verkrijging van de graad van doctor
aan de Technische Universiteit Eindhoven,
op gezag van de Rector Magnificus, prof.dr.ir. C.J. van Duijn,
voor een commissie aangewezen door het College voor Promoties
in het openbaar te verdedigen
op woensdag 5 juli 2006 om 16.00 uur

door

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geboren te Breda
This research was sponsored by the Foundation for Fundamental Research of Matter (FOM)

CIP-DATA LIBRARY TECHNISCHE UNIVERSITEIT EINDHOVEN

Broks, B.H.P.

ISBN-10: 90-386-2052-7
NUR 928
Trefwoorden: numerieke plasmafysica / numerieke simulatie / plasmaversnelling/ waterstofplasma / hoogspanningsschakelaars / plasmaspectroscopie / argonplasma
Subject headings: plasma simulation / plasma filled waveguides / plasma switches / spark gap / spectroscopic light sources / plasma sources / arc discharges

Printed by PrintPartners Ispkamp, Enschede, The Netherlands
Cover design: D.A.H. Broks
Cover art: Image based on data obtained as part of the INT Photometric Hα Survey of the Northern Galactic Plane: prepared by Nick Wright, University College London
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Chapter 1

Introduction

In this work a framework is presented, in which several transient plasmas are modeled using a general multi-fluid approach. These plasmas exhibit one unifying characteristic: they are used in the creation and guiding of light. The aim of the study is to provide a description of these plasmas that is as uniform as possible given the differences between them, and to use these descriptions to predict and understand the plasma behavior with an emphasis on their application.

1.1 The plasma state

A plasma is essentially a gas, in which the fraction of charged particles is so large that the behavior of the gas is significantly altered. In particular, a plasma conducts electricity, and couples strongly to electromagnetic fields. Furthermore, the electric forces that exist between charged particles allow long-range interaction, leading to a strongly collective and self-organizing behavior. The presence of ions and electrons also means that the plasma has a considerable energy density. This energy is the driving force behind many processes that may occur in a plasma, such as heating, the formation of electromagnetic fields, the creation and destruction of molecules, the excitation or de-excitation of atoms, ionization and recombination, and the creation, absorption and refraction of light.

The plasma state is common in the universe; Over 99% of the visible matter in the universe is in the plasma state, in for instance stars and nebulae. The sun, a star, is a huge plasma, emitting so much light that it can heat the Earth to a comfortable temperature at a distance of 150 million kilometers. This plasma is maintained by hydrogen fusion at a temperature of roughly 13 million K and densities of $10^5$ kg m$^{-3}$. Nebulae, on the other hand, are clouds of low-density gas that are often turned into the plasma state by means of light.
On Earth, the natural plasma state is more rare. Examples include lightning, St. Elmo’s fire and the corona Borealis and Australis. However, there are many man-made plasmas, which have diverse applications in for instance welding, lamps, lithography, waste treatment, speakers, decontamination, high-voltage switches, displays, nuclear fusion, and many others.

In all of these plasmas, light plays a role. Some are created to produce light, such as displays and lamps, some are produced by light, such as interstellar nebulae and laser-produced fusion plasmas, and others just emit light. In this thesis, plasmas that interact with light are described. This interaction can be the guiding or the production of light, in which the light may even create the plasma.

The plasmas mentioned above, and the plasmas described in this thesis, vary vastly in composition, density, size, energy source, energy density, temperature and the presence or absence of flow. Treating even a fraction of this enormous parameter space demands a flexible, and preferably modular, approach. A modular approach, in which relevant parts of a description of one plasma can be used in the description of another plasma that is in some respects similar allows the reuse of work. This greatly speeds up research. Furthermore, the repeated testing of modules under different circumstances offer possibilities for thorough validation.

1.2 Plasma modeling

A model is a simplified representation of some aspect of reality. Because a good model contains the essential features of a system, it should predict a behavior of the system that is consistent with experimental observations. The predictive power and insight offered by models are essential for the progress of science.

Analytical models have been used in science for centuries, and have been responsible for breakthroughs such as the prediction of the orbits of planets. Analytical modeling can give useful results, especially when good approximations can be made that reduce the complexity of the model. However, many realistic systems are too complex to be described accurately using analytical methods. A plasma, in which electromagnetic fields, interaction between many different types of atoms and molecules, the emission and absorption of light and flow all play a role, is an example of a system for which analytical modeling only has limited success.

With the availability of computers, a new tool for modeling became available, allowing accurate description of systems that could not feasibly be treated analytically. In computer modeling, the lab is replaced by a computer and the knowledge of the basic physics that define a system. Broadly speaking, a scientific problem is reduced to the basic equations, parameters and coefficients that describe it. These equations are
1.3 The codes used in this thesis

In this thesis, two codes, that have been developed at Eindhoven University of Technology, are used to describe the plasmas under study. We will first give a very brief overview of the philosophy behind the modeling. Then, a description of both codes with references to relevant documentation and publications will be given. The applica-
In our modeling philosophy, which is described in detail in [1, 2], modularity, orthogonality and extendability play a key role. In an orthogonal, modular program, the code is organized in modules that are highly independent, and communicate exclusively through well-defined interfaces. This makes it possible to easily exchange one module for another, without making changes to any of the many other modules that comprise the model. This approach has two key advantages:

- The well-defined input and output make replacement of a module much easier. In order to create a better module, all one has to do is to make sure that the input produces the desired output. It is not necessary to make any changes to any other modules; indeed, it is not even necessary to know anything about the contents of any other modules, greatly reducing the complexity of development, and the possibility to introduce bugs.

- Validating the code becomes much easier, as test cases that stress just one module can be made. If a bug is found, it is known exactly in which part of the code the bug is, greatly reducing the time needed to solve it.

It is sensible to use the physics of the plasma as a guideline for the boundaries of individual modules, for instance, the temperature equations, the density equations, the flow equations, the electromagnetic problem, etc. However, other aspects of the code, such as the processing of input data, the output of the computation, and the equation solvers are quite suitable for modularization as well.

The modularity of the code makes it easy to extend it by replacing one module with another module. This extendability is crucial for practical applications, as they often require dedicated pieces of code to handle aspects that are not foreseen by the code developers. In practice, a modeler can use the validated modules that are already present in the code, select those needed, if necessary create modules, and combine them into a model that describes the problem. The reuse of large pieces of validated code greatly speeds up development of a model, while greatly reducing the effort needed to validate the code. We refer to such a general codebase, that can be used to construct different models, as a modeling platform.

1.3.1 PLASIMO

The PLAsma SImulation MOdel PLASIMO code [3] is the result of over a decade of concerted effort to create a general, flexible and user-friendly plasma modeling toolkit. A thorough description of the PLASIMO code is beyond the scope of this thesis; the interested reader is referred to the literature in this Section, in particular the PhD theses of
the people who contributed to the project. These present a fairly complete description of the code and its capabilities.

The work on the PLASIMO project was begun by Benoy [4], who produced a version of the code in the programming language C, and applied it to the simulation of spectrochemical inductively coupled plasmas for spectrochemical analysis. Janssen [5] extended the code, and used it to compute both expanding cascaded arc sources and microwave-produced plasma for material processing.

A major rewrite of the code was performed by Van Dijk [1], in which the foundation for the modern PLASIMO was created. The code was rewritten in C++, an object-oriented programming language, that is particularly suitable to obtaining fast, modular code using modern programming techniques. He applied the code to an inductively coupled plasma, a mercury plasma and an argon-mercury plasma.

PLASIMO was extended significantly by Hartgers [6], by adding the possibility to do time-dependent modeling and to model non-Maxwellian electron energy distribution functions. He used the code to create an accurate model of the tubular fluorescent lamp. Further improvements to in particular the flow solver and the matrix solvers have been made by Garloff.

PLASIMO was used to model the sulfur lamp by Van der Heijden [7–10] and Johnston [11]. For this, the computation of both radiation transport and collision integrals were improved.

The result of all these efforts is a modular, extendable code, which is validated both by testing of individual models and by the successful modeling of many different plasmas. The code is based on a quasineutral (no net charge density in a macroscopic volume) fluid approach [12], and can handle both non-LTE and LTE plasmas [13]. The computation of flows interacting with these plasmas using the Navier-Stokes equations is supported. Time-dependence and orthocurvilinear coordinates are supported on Cartesian, cylindrical and spherical grids. Because of the modular nature of the code, it is also possible to handle simple flow and heat transport problems.

This code, with some adjustments and extensions, is used for most of the modeling in this thesis, namely the systems in Chapters 5 to 9 and Chapter 12. Using this unified approach, the deeper similarities that exist between various plasmas can be exploited to reduce duplicate effort and to cross-validate the code.

1.3.2 MD2D

The MicroDischarge 2-Dimensional (MD2D) code was initially created by Hagelaar [14] for the simulation of the microplasmas used in display technologies such as PALC [15] and PDP [16].

A significant rewrite of the code was performed by Van Dijk and Brok [17], who
restructured the code, and integrated various PLASIMO (See Section 1.3.1) modules into the code, such as the matrix solvers, the input parser, and the graphical user interface. This is part of an ongoing effort to integrate PLASIMO and MD2D. The code was used by Brok to simulate plasma breakdown between parabolic electrodes [17], breakdown of argon in a configuration resembling a fluorescent lamp [18], and a plasma needle for biomedical applications [19].

The code is a two-dimensional fluid code, which is suitable for the simulation of plasmas that are far from thermal equilibrium. The electron energy balance and various particle density balances are solved. Poisson’s equation is solved, allowing the description of plasmas that are not quasi-neutral. However, a basic assumption is that the plasma neither heats the background gas, nor to cause a drop in background gas density by inelastic processes such as ionization and recombination. The code is also incapable of computing a bulk flow field, although the ability to include a precalculated bulk flow was added by Brok for the work in this thesis.

The MD2D code is used in this thesis in Chapters 10 and 11. The code is particularly suitable to the low-density plasma that is treated in these Chapters, because it allows for deviations for quasi-neutrality, which are important in these kinds of plasmas.

1.4 The topics of this thesis

In this thesis, a variety of plasmas is described, that have an even wider range of applications, ranging from revealing the mysteries of the cosmos to the contents of a person’s pockets in an airport security scanner. All these topics share a central feature: plasma. This thesis can be seen as an application of a general framework, implemented in codes such as PLASIMO (cf. Section 1.3.1) and MD2D (cf. Section 1.3.2), for the multi-fluid simulation of transient plasmas, that are used for the creation and guiding of light.

The first part of the thesis is theoretical in nature. The derivation of the fluid equations from the Boltzmann Transport Equation is described, and the concept of a multi-fluid plasma is briefly explained with references to literature. The use of code reuse and modularity a general framework in modeling is illustrated using the example of cross sections. Furthermore, the concept of disturbed Bilateral Relations is used to create a simple 0 D model, that trades accuracy for simplicity and complements the grand models.

The second part of this thesis deals with the application of two grand models, MD2D and PLASIMO to various practical systems. The results are discussed while considering the application, and compared to the—often scarce—experimental data, which means that many of the results are predictive in nature. The thesis ends with some concluding remarks.
1.4. The topics of this thesis

The remainder of this Chapter is devoted to a more detailed description of the contents, with an emphasis on the application and the place in contemporary scientific literature the work in this thesis fills.

1.4.1 The Boltzmann Transport Equation

The Boltzmann Transport Equation is a fundamental equation describing the transport of an ensemble of particles [20, 21]. By integrating moments of this equation, the fundamental conservation equations of a plasma can be derived, including the mass, momentum and energy equation. While this procedure is by no means new, it is difficult to find a concise, complete, and correct treatment in literature, that is focused on plasma physics. The procedure is described in Chapter 2. The results, in both their extensive and intensive form, are presented. Furthermore, the most important assumptions, and the interpretation of the terms and source terms in the equation is given.

These results, which are derived for a system containing only one type of particles, are generalized for a plasma, in which many particles play a role. The complexity of the resulting system is reduced by rewriting them in a formulation in which the mass and momentum conservation of the plasma as a whole, the energy conservation of electrons and the energy conservation of the other particles in the plasma play a leading role. Furthermore, we discuss two salient details of the numerical procedure, namely the treatment of transient plasmas with different time scales and multi-region systems with different length scales.

1.4.2 A modular cross section implementation method

In order to compute transport properties of the plasma, cross sections of the elastic interaction of species are needed. While accurate general expressions exists for some cases, in particular Coulomb interaction [22], it is normally necessary to supply cross sections for many other processes. While some cross sections were present in the codes, these were found to be inadequate for highly accurate plasma modeling. In Chapter 3 two new methods to implement cross sections are discussed, with modularity, extendability and code generality in mind.

1.4.3 Disturbed Bilateral Relations

Plasmas are characterized by the vast amount of internal interactions between the various constituents. The method of disturbed Bilateral Relations (dBR) offers a theoretical framework to characterize these interaction.
Individual processes, such as electron impact ionization, or transfer of heat from one species A to another B, have a corresponding inverse reaction, such as two-particle recombination in the former case and heat transfer from B to A in the latter. A balance between these reactions is called a proper balance. In the case of full Thermal Equilibrium, the system is entirely described by a set of these proper balances, which are all in equilibrium.

In a real plasma, transport fluxes will disturb these proper balances. It is noted that these fluxes might either be spatial, or temporal (the plasma properties change faster than the equilibria can follow). These terms are called improper terms, and in some cases, these improper terms are so large that they completely eclipse the proper terms. The dBR method structures interactions by grouping proper and improper balance terms, yielding insight in the degree of equilibrium departure. This is particularly useful, because most interactions are strongly determined by either the proper or the improper terms; only rarely, both terms are of roughly equal magnitude for one process. For further information, the reader is referred to [23].

Because the framework of dBR is sufficiently general to include many plasma interactions with various degrees of non-equilibrium, it is a suitable foundation for modeling. Previous application of this type of modeling include the modeling of plasma torches [23, 24] and water droplet plasmas [25].

In Chapter 4, the principle of dBR is used to create a simple zero-dimensional plasma model. Such a model can be used to find the degree of equilibrium departure and for extremely quick computations, without the numerical problems that are involved in using complex multi-dimensional grand models such as PLASIMO or MD2D. Such a program is also simple from a software engineering point of view, and could feasibly be "programmed" in a common office spread sheet, without the need to know any programming language. This simplicity and robustness makes it a useful tool as a complement to the grand models. Indeed, many grand models require a reasonable estimate of the solution as an input parameter, which the dBR model is able to provide [26]. However, the dBR framework is neither as accurate or as extendible as the grand plasma model, and hence is a complement to them rather than a replacement.

The advantage that using the dBR approach has over conventional approaches is that it is in many cases suitable for both large and small deviations from Local Thermal Equilibrium (LTE). This is due to the fact that both the proper and improper terms are included in all key equations, i.e. the equation for the electron temperature, the electron density, and the heavy particle temperature. This is unlike conventional LTE models [27, 28] and models for plasmas with a cold background gas, as presented in [29]. The strongly nonlinear set of equations that results from including all terms can be solved robustly and efficiently by a dedicated algorithm that is described in detail in Chapter 4.
1.4. The topics of this thesis

Figure 1.1: A schematic representation of the guiding of a laser beam through a convex index of refraction pattern, showing a few optical paths. The central laser beam moves slower because of the higher central index of refraction, while the off-axis beams move faster, but travel a longer distance. The net propagation speed is identical, in accordance with Fermat’s principle. This principle is more commonly used in graded-index fiberoptics.

1.4.4 Capillary discharge waveguides and their applications

The advent of compact femtosecond terawatt laser systems [30] has enabled novel research in chemistry and physics. In two important disciplines, femtochemistry [31] and attophysics [32, 33], the short pulse length plays a key role. For our applications, however, it is in particular the vast laser power that is of critical importance.

The power density of these lasers can be increased even further by focusing the laser into a spot. However, diffraction will cause the laser beam to rapidly diverge again, over a distance equal to the Rayleigh length [34]. The use of waveguides may prevent this. Constructing a suitable waveguide becomes more challenging because at the power densities involved (> $10^{18} \text{ W m}^{-2}$), any material the laser interacts with, gas or solid, will be turned into a plasma almost instantaneously while depleting laser power.

1.4.4.1 Laser guiding

For guiding terawatt lasers through a medium over lengths much longer than their Rayleigh length, a convex index of refraction pattern is needed [35]. Guiding of a laser in such a pattern is graphically illustrated in Figure 1.1. Such a pattern can be created using a preformed plasma channel waveguide and/or by using relativistic self-focusing.

A plasma channel waveguide should have the following properties:

- A concave electron density profile. Such a profile results in a convex index of refraction pattern, which is needed for laser guiding [36].

- Full ionization, i.e. (almost) every electron in the path of the laser should be free i.e. not bound to an atom. If the plasma is not fully ionized, the laser will ionize the plasma further. Because the laser is typically most intense in the center, and the ionization strongly increases with increasing laser power, the electron density
profile created by the laser has a tendency to become convex. This causes a concave index of refraction pattern and thus rapid divergence. This phenomenon is known as ionization induced defocusing [37].

For very large laser power densities (typically $10^{23}$ W m$^{-2}$), ionization induced defocusing is no longer an issue, in particular in a gas that is easily fully ionized, such as hydrogen or helium. For such power densities, the front of the laser pulse has already fully ionized the gas, creating an electron density profile that is essentially flat. This precludes ionization induced defocusing for the rest of the laser pulse. The convex index of refraction pattern is now created by a relativistic increase of the electron rest mass, due to the laser-induced electron acceleration [38–42].

While conceptually simple, as only a gas jet and a laser are needed, and certainly effective [43, 44], this so-called self-focusing scheme has two disadvantages:

- It requires exceptionally powerful and consequently expensive lasers. Faure et al. [43] remark that their laser, that generates 1 J in 30 fs, is at the lower limit for using this scheme.

- The laser and plasma properties are closely related, only a small part of the parameter space can be addressed.

Because of these limitations on the self-focusing scheme, plasma channel waveguides are used extensively for guiding of terawatt lasers. Sometimes, both methods are combined, i.e. a laser beam is guided that is powerful enough to cause relativistic self-focusing in a plasma channel waveguide [45–47].

There are several schemes by which plasma channel waveguides are created. We will give a brief overview, to end with a more detailed description of the device under study: the pulsed capillary discharge waveguide.

One scheme uses a hydrogen jet, in which lasers create a plasma [34, 48]. This hot plasma expands outward in a few nanoseconds, creating a shock wave and a concave electron density profile, in which guiding is possible for several nanoseconds. The channel quality hence critically depends on both the laser power and the time between the channel creation and the interaction with the pulse that is to be guided.

An expanding shock in a plasma might also be created using a pinch plasma [49–54]. If a plasma is subjected to a pulse with very high current density (typically $>10^{13}$ A m$^{-2}$), the Lorentz force becomes so strong that the plasma is compressed radially. This collapse is followed by an expansion creating a shock and a hollow density profile. However, the strong magnetic fields do make the plasma susceptible to magneto-hydrodynamic instabilities [55].

These plasmas are typically constructed in prefilled capillaries, with a sub-millimeter diameter. While the hot plasma can ablate the capillary wall, the pinching reduces the
plasma density near the wall, reducing the damage. For many applications, wall ablation is undesirable, as it contaminates the plasma with elements which are difficult to fully ionize, causing ionization induced defocusing when the plasma interacts with a laser. Furthermore, the ablation of wall material negatively impacts the lifetime of the device.

The material that is ablated from the wall can also be used as the (sole) source of particles for the plasma [56–59]. If a field is applied over a capillary that is sufficient for dielectric breakdown of the walls to occur, material is ejected into the capillary, and a channel with a concave density may form. Polymers are popular wall materials for this purpose, as the hydrogen they contain can be ionized fully at typical plasma conditions, and the carbon can be excited to states that are high enough to prevent serious ionization induced defocusing. A controlled amount of impurities can be released into the plasma by including them in the walls as dopant [60, 61]. High currents and pinching induce magneto-hydrodynamic instabilities, which may have a significant influence on channel behavior [62].

The slow pulsed capillary discharge waveguide is the newest type of gas-filled capillary plasma channel waveguide [63–67]. The high, short current pulse that causes heating and pinching is replaced by a longer, less intense pulse, which is sufficient to heat and ionize the plasma but not strong enough to cause pinching. Rather than relying on the pinch effect, the concave electron density profile in these waveguides is formed by the temperature gradient that exists between the cold wall and the hot central plasma, in effect creating a wall-stabilized arc plasma. In an ideal gas with a uniform pressure, conditions which are approximated in slow pulsed capillary discharge, this convex temperature gradient causes a concave electron density profile.

The main advantages are the long device lifetime, as wall ablation may be prevented, and the fact that the guiding channel that is formed [63, 68–72] is stable for a long time. Because of these advantages, their simple construction, and proven suitability in guiding terawatt laser pulses [63, 65], slow pulsed capillary discharge waveguides are very promising devices for the applications that require the guiding of terawatt lasers. They are studied in detail in this thesis in Chapters 5 to 8.

### 1.4.4.2 Applications of plasma waveguides

There are several applications in which the interaction of terawatt laser beams with matter plays a key role. These include high-harmonics generation, soft X-ray lasing, and laser wakefield acceleration. For these applications, a long interaction length between laser and matter is of critical importance. Hence, investigation of guiding schemes, both theoretical and experimental, is of great value.

High-harmonics generation is a promising method of creating radiation with a short
wavelength. When an atom or ion is subjected to a very strong laser field, it may respond in a strongly nonlinear way by emitting photons corresponding to a higher harmonic of a certain fundamental optical transition, i.e. these photons have a frequency that is an integer multiple of a frequency matching an optical transition in the atom. Using this method, up to the 300th harmonic has been generated [73–77]. Hence, starting from optical wavelengths, it becomes possible to release photons that are in the vacuum ultraviolet (VUV) (100-300 nm), or even in the soft X-ray range, where wavelengths of 13.4 nm may be used for next-generation lithography [78, 79]) and wavelengths between 4.4 to 2.3 nm that may be used to perform microscopy in the so-called “water window”. At this where water is transparent to radiation, making this a promising way of studying biological samples [80, 81].

The wavelength that can be reached efficiently by higher harmonics generation strongly shortens with increasing input field strength. Hence, terawatt lasers are very promising sources for the generation of short-wavelength high-harmonics. By using plasma waveguides, the interaction length between the laser and the plasma may be increases, increasing the output. It is even possible to obtain lasing [79].

There is another scheme by which plasma-laser interaction might be used to generate soft X-rays. The ionization and excitation energy of an atom rise with increasing ionization state. Pulsed capillary discharges, in particular of the ablative pinch type, can generate a plasma that is so hot that highly ionized states of gases such as carbon, tin, xenon and argon are formed, which have transitions that may generate soft X-rays [51, 60, 62, 64]. If population inversion is achieved, for instance by recombination, lasing is possible. It is possible to use this scheme to generate soft X-rays using just the atoms that are excited by the discharge [51, 60, 62], but it is also possible to guide a laser beam through the plasma waveguide to boost the power density and create a higher output [64, 82].

The application that the research at Eindhoven University of Technology focuses on is laser-wakefield acceleration. The concept of using plasma-based acceleration, of which laser-wakefield is an example, was first publiced by Tajima and Dawson in 1979 [83]. The chief advantage of plasma-based acceleration is the high acceleration gradient, which can reach values of typically 100 GV m$^{-1}$ [84, 85]. This is three orders of magnitude higher than in conventional RF linear accelerators, where the acceleration gradient is limited by the breakdown at the metal surfaces.

One particularly interesting use of these accelerators is as drivers for a free-electron laser, that can be used to generate tunable infrared light with a wavelength of around 7 µm. This allows surgery with unmatched precision [86]. A more compact electron source may make this application more practically feasible. Electron bunches can furthermore be made to interact with a metal, generating intense terahertz radiation [87].

Before the availability of lasers that were powerful enough to excite the laser fields
necessary for acceleration directly, plasma beatwave acceleration was the most popular method of laser-plasma acceleration [88–90]. Briefly, two lasers with a slightly different frequency are sent through the plasma. This slight frequency difference manifests as mode beating in the resultant electric field, and this mode beating is tuned to the plasma frequency, resonantly exciting an electric field [85].

Interest in plasma beatwave acceleration waned with the increased commercial availability of table-top terawatt lasers, which make laser-wakefield acceleration possible [85]. This scheme is schematically shown in Figure 1.2. The ponderomotive force of the laser, which can reach gigabars of pressure [91], pushes away some of the electrons in the plasma, temporarily breaking quasineutrality. After the laser pulse passes, strong Coulomb forces will pull the electrons back, causing an oscillation in the plasma with a very large electric field in the wake of the laser. This traveling electric field can trap electrons, and accelerate them.

This scheme has been successfully used to generate intense electron bunches with energies of around 100 MeV and excellent beam quality [43, 44, 48]. A plasma channel that guides the laser was essential in these experiments [93].

One of the strengths of laser-wakefield acceleration is that both the electrons and the
laser pulse move at a speed close to \( c \), the speed of light. However, the optical density of the plasma slows down the laser somewhat, causing the electrons to gradually overtake the laser [85, 92, 94]. This makes low plasma densities desirable, as this reduces the optical density of the plasma and hence the velocity difference. However, lower plasma densities may reduce the focusing power of the plasma and increase the area of the laser beam in the channel, which leads to an undesired lowering of the laser intensity [68, 72]. Furthermore, a lower density results in a lower accelerating field [95]. This, and several other considerations, such as relativistic self trapping [45–47, 95], underscore the critical dependence of the laser-wakefield mechanism on plasma properties.

### 1.4.4.3 Slow pulsed capillary discharge waveguides in this thesis

We have investigated a slow pulsed capillary discharge waveguide in this thesis, with its application in laser-wakefield acceleration in mind. The present body of research on the slow capillary discharge is small. The channel was developed at Oxford University, and first introduced by Spence and Hooker in 2001 [63]. Guiding over many Rayleigh lengths was demonstrated [96]. The first theoretical investigation was presented by Bobrova et al. in 2002 [68]. In this investigation, a magneto-hydrodynamic (MHD) two-temperature LTE model was used to model the plasma. While the model matched satisfactorily with experiments, the use of a different current than used in the experiment, the inadequate description of the capillary wall, and the deviations from the Saha equation that are to be expected in such a strongly dynamic system cast doubts on the accuracy of the model [97]. Furthermore, the chemistry model is based on a two-temperature Saha equation that is demonstrably wrong [98, 99]. In the same year, guiding of terawatt laser pulses was experimentally demonstrated by Butler et al. [66]. After this basic analysis, an MHD stability analysis was presented by Wang et al. [55]. It was found that the potential MHD instabilities develop at timescales that are much longer than the time needed for the laser to travel through the plasma, and can hence be avoided. A detailed review of the slow pulsed capillary discharge waveguide was presented in [64]. Quite recently, the slow pulsed capillary discharge proved to be instrumental of breaking the 1 GV/m barrier in laser-wakefield acceleration [100], further underscoring its importance in the progress of physics.

In this thesis, Chapters 5 to 8 are dedicated to the slow pulsed capillary discharge waveguide, providing a thorough numerical investigation of this device. In these Chapters, a non-Local Thermal Equilibrium (non-LTE) model of the waveguide is presented. This model addresses the issues [100] that were raised with regards to the work of Bobrova et al. [68]. It consists of a one or two-dimensional, two-temperature multi-region plasma/wall model, that can track the great dynamic variations easily because of the powerful non-LTE approach. In order to accurately and efficiently simulate the plasma,
a multi-region method is used for the heat transport, which happens at very different time and length scales in plasma and wall, was added. Furthermore, the ability to parse a wide variety of current shapes was added to the code to be able to simulate realistic current profiles. Efficiency was further boosted by adding a more fine-grained control of the simulation time steps. Accuracy was improved by implementing a novel (viz. Chapter 12) theory of plasma nonideality [101], allowing for a better prediction of the transport properties, and by the addition of a model of the hydrogen chemistry that uses recent experimental data. This model is used to simulate an actual capillary [63]. Furthermore, a parameter study of the discharge current was performed in this Chapter.

In Chapter 6, the model in Chapter 5 has been used to perform a parameter study, in which initial plasma density and capillary radius were varied. A general scaling law is presented, which can be used to predict guiding properties for a wide range of capillary discharge waveguides.

In Chapter 7, an investigation of a capillary discharge waveguide with a modulated radius is presented. Such a modulation could potentially be used to locally tailor the plasma and guiding properties. It may also provide insight in the influence of surface roughness.

Chapter 8 describes the results of numerical modeling of a rectangular pulsed capillary discharge waveguide. This device was developed at Oxford University to be able to do interferometry measurements of the electron density profile perpendicular to the axis of the channel [100]. This method is more accurate than the earlier electron density profile measurements along the channel axis [63]. The numerical analysis is used to quantify the measurement errors induced by wall heating, to investigate the difference in plasma behavior between the rectangular and the more common round channels, and to translate the one-dimensional measurements to a two-dimensional electron density profile. The numerical results are compared to experimental results that were obtained by A. J. Gonsalves, T. Rowlands-Rees and S. M. Hooker of Oxford University [100].

### 1.4.5 The photoconductively switched spark gap discharges and its applications

Spark gaps basically consist of two conducting high-voltage electrodes separated by a gas-filled gap. If this gas breaks down, either spontaneously because of a sufficiently high voltage, or because of an external trigger, a conductive path between the electrodes is formed, closing the electric circuit. This allows for the rapid switching of high voltages, and the production of intense sparks.

The technological use of spark gaps is over a century old. It was a key component of early radio transmitters, as it transmits broadband radio waves. Common modern applications include overvoltage protection, where a spark gap with a certain break
down voltage is used to shunt voltages that are higher than this breakdown voltage away from vulnerable equipment, in ignition, such as the spark plugs in a car engine, and in the rapid switching of high currents.

Rapid switching of a voltage that is below the breakdown voltage of a spark gap can be achieved using an external trigger. A common trigger is a third electrode, located in an electrically insulated orifice in one of the main electrodes. An example of this principle is found in the so-called trigatron [102], a device used in early radars. In a trigatron, a short high-voltage pulse on the third electrode causes a streamer to form, closing the gap [103, 104]. The rise time of these gaps is typically in the tens of nanosecond range, and they are used to switch voltages ranging from several kilovolts to several megavolt. Typically, triggered spark gaps can still only be switched when the voltage over them is close to the breakdown voltage.

The principle of using a laser to trigger a spark gap was first described by Guenther and Griffin in 1965 [105]. This allows for shorter breakdown times and jitter as the stochastic avalanches and streamers are bypassed [106]. Furthermore, the trigger is electrically isolated from the high-voltage circuit, which removes this safety hazard. There are various ways of triggering a spark gap by means of a laser. The earliest idea was to simply illuminate a part of the inter-electrode gap by creating a point focus with a perpendicular laser beam, producing a plasma which provides seed electrons for the discharge. An alternative triggering technique is focusing the laser on the electrode surface [107], releasing a plume of ablated electrode material which triggers the gap.

Faster, more stable switching can be achieved by ionizing the entire interelectrode gap by creating an axial laser focus by a laser which passes through an orifice in the electrodes [108]. Using this technique, switching of voltages between 1 and 500 kV is possible with jitter and rise time in the sub-nanosecond regime [109, 110].

The novel compact femtosecond, terawatt laser systems [30], such as the Ti:Sapphire laser, make it relatively easy to obtain field strengths that are so high that tunnel ionization of a gas becomes possible [111–113]. This allows for the fast (sub-picosecond) production of well-ionized plasma between the electrodes. This is unlike a conventional laser-triggered spark gap, where the laser produces seed electrons, which multiply due to plasma processes, and allow for a well-conducting channel to form. By closing the gap with this plasma, ultrafast switching with rise time and jitter that is an order of magnitude shorter than conventional laser-triggered spark gaps becomes possible [114].

Recently, an experimental characterization of such a device, dubbed a photoconduc-tively switched spark gap, was presented [115, 116]. The reported rise time was better than 100 ps, while the jitter was better than 15 ps. Furthermore, unlike conventional spark gaps, the photoconductively switches spark gap is able to switch voltages which are only a fraction of the breakdown voltage. The primary application of this spark gap is in the generation of ultrashort high-voltage pulses for use in compact DC electron
1.4. The topics of this thesis

accelerators [117]. Other applications include bioelectrics, where higher fields can be applied by virtue of the shorter pulse duration [118], and the generation of terahertz radiation, which is very promising as an alternative for X-rays in medical and security applications [119, 120].

In spite of the experimental success, the details of the spark gap plasma were not well understood. In particular, it was observed that there was a significant voltage drop over the spark gap, especially for lower output voltages. While conventional laser-triggered spark gap plasmas have been studied theoretically and are well understood [121], the much shorter time scales and higher initial electron densities in the photoconductively switched spark gap limit the applicability of these studies. In Chapter 9, the first theoretical study of the plasma in the gap is presented, with an emphasis on the switching behavior. Two possible causes of the voltage loss observed in switching are investigated.

Cold cathodes may need a cathode fall of several hundreds of Volts to emit electrons (cf. Chapter 10 and 11). This is a possible cause of the observed voltage loss, and is investigated by means of an analytical model.

Even a well-ionized plasma has a finite conductivity. For singly ionized plasmas with temperatures in the eV range, this conductivity is of the order of $10^4 \Omega^{-1} m^{-1}$ [71, 116]. For the typical size of the laser-produced plasma filament, it has been estimated that this leads to a significant voltage drop over the spark gap. This proposal has been investigated using a numerical model of the plasma between the electrodes. This model is based on the model presented in Chapters 5 to 8 of this thesis. An important improvement made to PLASIMO was the inclusion of a correct way of treating dissociative recombination, which is a key process in this discharge. Furthermore, the hydrogen chemistry has been replaced by a nitrogen chemistry, adapted to the very short time scale of the discharge ($\approx 2$ ns). Owing to the modular structure of the PLASIMO code, implementing a new chemistry is quite straightforward, underscoring the advantages of our modeling philosophy. A critical analysis of the used nitrogen chemistry is presented. The model not only allows a theoretical prediction of the switching behavior, which is verified by the experiments performed by J. Hendriks and G. J. H. Brussaard of Eindhoven University of Technology [115, 116], but also provides insight in the fundamental plasma processes, and their relative importance related to the laser-induced processes. It is the first step in obtaining a quantitative understanding of the novel, intense and extremely short time scale plasma that exists in the spark gap, and demonstrates useful techniques for short time scale fluid modeling.
1.4.6 The pulsed discharge nozzle and its applications

In 1921, Heger [122] discovered absorption features in the light from distant stars that did not seem to be subject to a Doppler shift, even though the stars had a high radial velocity. From this, it was concluded that the absorption occurred in the interstellar medium, rather than in the stars themselves [123]. Because of this, and because the lines were somewhat more diffuse than atomic lines, the term Diffuse Interstellar Bands (DIB) was coined. Currently, a few hundreds of these structures are characterized [124].

There have been many suggestions about the origin of these bands, such as silica grains [125], metal oxides [126], and organic molecules [127]. It took until the late 90s to firmly entrench the latter theory as the current explanation of the DIBs [128–130].

The organic molecules most likely responsible for the DIBs are Polycyclic Aromatic Hydrocarbons, or PAHs, and their cations [131–133]. In order to identify the PAHs responsible for particular DIBs, a setup was constructed at NASA Ames. This setup is designed to emulate the environment in which the PAHs and their cations exist in space—cold, high vacuum, few collisions between particles, and excited by VUV photons.

The Pulsed Discharge Nozzle (PDN) consists of a slit-like nozzle, through which a plume of noble gas expands hypersonically into a high-vacuum chamber [134–136], reaching temperatures of a few tens of K. A glow discharge plasma that is created through the gas jet creates electrons, ions, and excited noble gas atoms. It is thought that these excited noble gas ions are capable of ionizing PAHs with limited fragmentation [134], essentially playing the role of the VUV photons in the interstellar medium.

Using a slightly modified version of the MD2D code [18], we present the first model of the plasma that exists in the gas jet in Chapter 10. This modification consist of the inclusion of an external flow on the transport properties, something easily accomplished owing to the modular structure of the MD2D code. The results of this model have been used to gain insight of the plasma properties that exist in the discharge, and are instrumental in understanding the excitation mechanism of PAHs in the plasma.

The same model that is used in Chapter 10 is used in Chapter 11 to perform a parameter study on the anode-cathode spacing in the PDN. By analyzing the impact of the anode-cathode spacing on the plasma properties, we have confirmed the conclusions from the previous analysis of the PDN in Chapter 10 and [137, 138], and assessed the usefulness of lengthening the anode-cathode spacing to increase the effectiveness of the PDN at producing PAH cations, without the need for an overhaul of the experimental apparatus.
1.4.7 The cascaded arc and its applications

The cascaded arc was invented by Maecker [139] in 1956 as an intense light source. A cascaded arc essentially consists of a bore in a stack of metal plates, that are electrically isolated by isolating washers. The arc is stabilized by the presence of the metal plates, which are good conductors of heat and can withstand the intense thermal loads that are generated by the arc plasma. The arc plates are electrically isolated and floating. This allows for a gradual drop in voltage over the arc, which greatly enhances the stability.

Flowing cascaded arcs are used as sources of chemically excited particles such as ions and radicals. In this application, a jet of plasma from the arc is directed at a substrate, which may be mixed with gases either in the arc, or when traveling to the substrate. The remote operation of the arc allows for an independent optimization of the plasma generation and the interaction with the substrate. The high plasma densities generated by the arc allow for fast treatment [140], which is desirable for industrial applications. Applications include the deposition of silicon [141], carbon [142], and etching in the semiconductor industry [143].

Stationary cascaded arcs are used for their original purpose as light sources. The cascaded arc channel contains a dense plasma, which produces a strong broadband emission by Bremsstrahlung and free-bound radiation, ranging from the VUV [144] to the infrared [145]. The solid angle over which this light is emitted is small due to the geometry of the arc. This is favorable for imaging in many spectroscopic applications [146]. Furthermore, the high stability of the cascaded arc allows it to be used as a radiation standard in the VUV [147, 148].

In Chapter 12, a cascaded arc with a geometrically constricted arc channel is presented. This geometrical constriction was shown by Burm et al. [149–151] to increase the efflux of excited species in a flowing cascaded arc. In Chapter 12, we investigate to which extent such a constriction enhances the light output of the cascaded arc.

In order to accurately model the cascaded arc, care has been taken to use recent and accurate cross section data for argon, greatly benefitting from the work presented in Chapter 3. Furthermore, a correction to the standard Coulomb logarithm is used to account for the weak plasma nonideality that is present in dense, cool plasmas [101]. This treatment of plasma nonideality is used throughout this work for all the PLASIMO simulations, as all these plasmas are in the weakly nonideal regime. By comparing the simulated results of the cascaded arc with measurement, in particular the arc voltage, the validity of our treatment of plasma nonideality can be assessed.

It is noted that this cascaded arc plasma, unlike the other plasmas in this thesis, is stationary. This does not preclude the use of our general framework, as the stationary state can simply be seen as the special case of an infinitely long transition time.
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Chapter 2

Multi-fluid modeling of transient plasmas: the method

Abstract—In this thesis, a multi-fluid method is used for the modeling of transient plasmas. This method is based on equations which describe the conservation of particles, momentum and energy. In this chapter, we will derive these equations from the fundamental Boltzmann Transport Equation. The assumptions made and the physical interpretation of the various terms will be discussed. The energy equation for the electrons, which plays a crucial role in the plasmas in this thesis, is treated in detail. Furthermore, salient features of the numerical method used are discussed.

2.1 Introduction

Modeling a physical system requires a firm mathematical description of the physics underlying its behavior. For a plasma, this description should describe the interaction of the many different species—atoms, ions, electrons, and possibly molecules—with electromagnetic fields, gravitation, radiation, and external particle sources.

In this Chapter, we will give an outline of the theory that is used for the modeling of the plasmas described in this thesis. This theory should be general enough to handle the wide variety of transient plasmas, but should also be simple enough to simulate the plasmas in a manageable amount of computer time.

The description we have chosen is a multi-fluid description. In this description, we greatly reduce the number of equations that describe the system by assuming that many species have an almost identical temperature and average velocity. In this context, a species is defined as an electron, or an atom, ion or molecule with a certain internal structure; hence, atoms, ions or molecules with a different excitation state are considered to be different species.

This multi-fluid description is implemented in the codes that are used in the Eind-
hoven modeling group, PLASIMO and MD2D. Key features of these codes are described in [1–6] and [7, 8], respectively.

In this Chapter, we will start by presenting the fundamental conservation equations for a single species in Section 2.2. A detailed derivation, including a critical discussion of the assumptions, is presented in Appendix 2.A. These fundamental conservation equations, known as the particle balance, the momentum balance, and the energy balance, are used to arrive at the multi-fluid description, as presented in Section 2.3. In this Section, we will start by treating the fundamental difference between the behavior of the electrons and the other particles in the plasma, which is of fundamental importance in the multi-fluid approach. The implementation of this multi-fluid approach in PLASIMO is presented in Section 2.3.1, with an emphasis on the treatment of diffusion and the existence of multiple temperatures. The implementation of these equations in MD2D is described and contrasted with PLASIMO. The solution procedure of the equations that describe the plasma is discussed in Section 2.4, and discussion of multi-region modeling in Section 2.5. Conclusions are presented in Section 2.6.

2.2 The conservation equations

A plasma consists of many species, which are described using the multi-fluid approach. In this Section, we will present the building blocks of this multi-fluid approach, namely the conservation equations of mass, momentum and energy for one single species.

Each particle of a single species can be considered to occupy a position in a six-dimensional phase space of spatial position $\vec{r}$ and velocity $\vec{w}$. If we could track the phase space coordinates of all the particles as a function of time we would have (more than) enough information to follow the evolution of the plasma as a whole. Generally, however, we are not so much interested in the behavior of the individual particles, but rather in the macroscopic properties of the plasma. This is certainly the case for the plasmas presented in this work, in which the density is typically $10^{24} \text{ m}^{-3}$. It is therefore much more convenient to study the phase space distribution function $f(\vec{r}, \vec{w}, t)$, with $t$ the time. $f(\vec{r}, \vec{w}, t)$ is defined such that $f(\vec{r}, \vec{w}, t) \, d^3r \, d^3w$ equals the number of particles in a volume element $d^3r \, d^3w$ in the phase space at time $t$. The temporal behavior of the species can then be described by tracking the evolution of the distribution function $f(\vec{r}, \vec{w}, t)$ which can be done using the Boltzmann Transport Equation [9, 10]:

$$\left( \frac{\partial}{\partial t} + \vec{w} \cdot \nabla + \frac{\vec{F}}{m} \cdot \nabla_w \right) f(\vec{r}, \vec{w}, t) = \left( \frac{\partial f(\vec{r}, \vec{w}, t)}{\partial t} \right)_c .$$  \hspace{1cm} (2.1)

Here, $m$ denotes the mass of the particle, $\vec{F}$ denotes external forces, and the subscript $c$ denotes the effect of collisions of this species with other species. $\nabla_w$ denotes the nabla-
operator in velocity rather than normal space.

The conservation equations of mass, momentum and energy for a single species may be obtained by the method of moments. In this method, $f(\vec{r}, \vec{w}, t)$ is multiplied with a function $g(\vec{w})$ of the velocity and integrated over the entire velocity space. For the case that $g(\vec{w}) = m$, we obtain the mass conservation equation, if $g(\vec{w}) = m\vec{w}$ we obtain the momentum conservation equation, and if $g(\vec{w}) = \frac{1}{2}m\vec{w}^2$, with $w$ the magnitude of $m\vec{w}$, we obtain the energy conservation equation. The derivation of these equations is somewhat complicated, and is presented in Appendix 2.A. A detailed discussion of the assumptions made is presented in Appendix 2.A as well. In this Section (2.2), we will merely present the results, and discuss on the physical interpretation of the various terms.

### 2.2.1 The mass conservation equation

The mass conservation equation is given by

$$\frac{\partial mn}{\partial t} + \vec{\nabla} \cdot (mn\vec{v}) = \left( \frac{\partial mn}{\partial t} \right)_c. \quad (2.2)$$

Here, $\bar{v}$ denotes the average velocity $n$ denotes the particle number density of this species.

The first term of the mass conservation equation (2.2) denotes the accumulation of the specific mass. This term is nonzero in a transient system, such as the systems treated here. The second term is the transport of mass by the average velocity. The plasmas simulated in this work exhibit significant density gradients, making the common simplification of moving $nmn$ out of the divergence inapplicable. The term on the right-hand side denotes the mass formation caused by interactions with other species. This can for instance be the creation of a certain species by a chemical reaction, thus increasing the total amount of the specific mass.

### 2.2.2 The momentum conservation equation

In Appendix 2.A, two forms of the momentum conservation equation are derived. In (2.75), we have the extensive form, in which the momentum $mn\vec{v}$ appears. In (2.77), we present an intensive form, in which the velocity rather than the momentum is the variable. Here, we discuss the extensive form, which is given by:

$$\frac{\partial nm\bar{v}}{\partial t} + \vec{\nabla} \cdot (mn\bar{v}\vec{v}) + \vec{\nabla} \cdot \Pi + \nabla p - n\vec{F} = \int m\vec{w} \left( \frac{\partial f}{\partial t} \right)_c d^3w, \quad (2.3)$$
were \( \Pi \) denotes the viscosity, \( p \) denotes the pressure, and \( \vec{F} \) the specific external forces exerted on the species.

The first term of (2.3) represents the accumulation of the specific momentum, which is generally nonzero in a transient system. The second term denotes the momentum transport caused by the flow. The third term represents the viscous forces, which are caused the friction the species exerts on itself. The fourth term is the pressure gradient. For many flowing systems, including the plasmas treated in this work, this is the driving force that causes the various plasma species to flow. The fifth term represents the external forces, thus the combined action of the electric force, the Lorentz force and gravity. Tight-hand side term represents the momentum gained and lost through collisions with other species. This may include the transfer of momentum from other species, or the creation of species with nonzero momentum.

### 2.2.3 The energy conservation equation

We have derived various forms of the energy conservation equation in Appendix 2.A: an extrinsic form, taking into account both the thermal and the directed kinetic energy in (2.90), an intrinsic form (not containing directed kinetic energy) in terms of pressure in (2.93), and an intrinsic form in terms of the specific temperature (2.99). In the multi-fluid description, (2.99) is used with Fourier’s law for the thermal conductivity (2.103) is substituted in (2.99). This gives:

\[
\frac{3}{2} \frac{\partial n k_B T}{\partial t} + \frac{3}{2} \vec{\nabla} \cdot (n k_B T \vec{v}) + p \vec{\nabla} \cdot \vec{v} + (\vec{\nabla} \vec{v}) : \Pi - \vec{\nabla} \cdot \left( \lambda \vec{\nabla} T \right) + \int E^T \left( \frac{\partial f}{\partial t} \right) \text{d}^3w
\]

(2.4)

with \( k_B \) Boltzmann’s Constant, \( \lambda \) the thermal conductivity and \( E^T \) the thermal energy. By assuming the existence of a temperature \( T \) for the species, we implicitly assume Maxwell-Boltzmann equilibrium. However, (2.4) can readily be rewritten in terms of average particle energies if deviations from Maxwell-Boltzmann equilibrium are relevant. This is for instance the case in Chapters 10 and 11.

The first term on the left-hand side of (2.4) denotes the accumulation of thermal energy, and generally is nonzero in the transient systems treated in this work. The second term represents the convective transport of energy by means of the systematic velocity of the species. The third term represents the expansion work. The fourth term is the production of thermal energy by viscous dissipation, which is in fact the transfer of directed kinetic energy to random thermal energy in the species. The fifth term represents the diffusive heat transport (thermal conduction). The term on the right-hand side is
the accumulation of thermal energy by collisions. These collisions may either be elas-
tic, in which case the energy of the species changes as thermal energy of the particles
involved in the collisions changes, or inelastic, in which case particles with a nonzero
thermal energy might be created or destroyed.

Conspicuously absent from (2.4) is the energy generated by external forces. These
forces do not generate random energy, but rather directed energy. This directed en-
ergy is not part of (2.4); however, the randomization process that leads to conversion to
thermal energy is included in the source term of (2.4).

2.3 A multi-fluid plasma description

A plasma contains many different species (or particle types). While it is possible to treat
all species with separate mass (2.2), momentum (2.3), and energy (2.4) conservation
equations, this is quite laborious and often not necessary. For the plasmas treated in
this work, a multi-fluid two-temperature approach will be used, based on chemical non-
equilibrium.

This multi-fluid method is based on the fact that the electrons $e$ behave differently
from the atoms, molecules and ions. We will refer to the latter collectively as heavy
particles $h$. Electrons are typically between 3 and 5 orders of magnitude lighter than the
heavy particles, leading to distinctly different behavior:

- Due to the high mobility, most of he current in the plasma is carried by the elec-
  trons, and hence they receive most of the heat generated by Ohmic dissipation.

- The thermal exchange between electrons and heavy particles is inefficient as only
  a fraction of the thermal energy difference of the order of $\frac{m_e}{m_h}$ is exchanged per
  collision. This allows the electrons to have a different temperature $T_e$ than the
  heavy particles $T_h$.

- In contrast to the heavy particles, the electrons are not easily cooled by means of
  collisions with the wall. This has the potential to further increase the difference
  between $T_e$ and $T_h$.

- The more mobile and often hotter electrons often play a dominant role in the reac-
  tions in the plasma.

This difference in the behavior between electrons and heavy particles allows for a con-
siderable simplification of the description of the plasma, as we will see below.
2.3.1 The multi-fluid description in PLASIMO

PLASIMO is a flexible, modular plasma simulation platform. In this code, it is presently assumed that charge neutrality exists, meaning that there is little significant deviation between the positive and negative charge density in a macroscopic plasma volume. This is a particularly good assumption for the simulation of plasmas with a high electron density, as is the case for all plasmas treated in this Thesis, except from the Pulsed Discharge Nozzle described in Chapters 10 and 11.

Solving all transport equations for all species is avoided in PLASIMO by combining several equations in one or more general conservation equation. All these conservation equations have the following general form [3, 11]:

\[
\frac{\partial \phi}{\partial t} + \nabla \cdot \left( \vec{U} \phi \right) - \nabla \cdot \left( \Gamma \nabla \phi \right) = S_c + S_p \phi \tag{2.5}
\]

Here, \( \phi \) denotes a conserved quantity, and \( S_c \) and \( S_p \) denote the linearized source terms. The meaning of \( C \), \( \bar{U} \) and \( \Gamma \) depends on the particular conserved quantity, but they can be loosely identified with inertia, the convective velocity and diffusion coefficient, respectively. Using a single form for all the transport equation greatly simplifies the discretization and numerical solution procedure.

The set of equations describing the plasma are combined in two sets of equations. The individual mass (2.2) and momentum (2.3) conservation equations are summed to the Navier-Stokes equations, describing the barycentric velocities, while allowing for deviations from these equations due to drift and diffusion. The individual temperature equations (2.4) of the heavy particles are summed to obtain a single heavy-particle temperature equation, while the electron temperature is solved with a separate equation. This is described below.

2.3.1.1 The mass and momentum conservation equation

The elastic collisions between the particles of the plasma distribute the momentum these particles carry. This has a tendency of equalizing the average velocities of the various species. Therefore, it is advantageous to describe the mass and momentum conservation of a species in terms of the mass and momentum conservation of the plasma as a whole and deviations from this equilibrium to describe the exact behavior of the plasma. In this description, we follow the work of Hartgers [5, 12] and references therein, where a much more detailed analysis and rigorous derivation of the equations presented here may be found.

In the presence of other species, the mass and momentum conservation equation for a species \( i \) take the following form:
The total bulk force $\vec{F}$ and the effect of elastic collisions is split into the thermodiatomic force $\vec{R}_i^T$, the friction force $\vec{R}_i^F$ and the remaining forces $\vec{F}_R$. The friction force is given by

$$\vec{R}_i^F = \sum_j f_{ij} (\vec{v}_i - \vec{v}_j)$$

(2.8)

with the volumetric friction coefficient $f_{ij}$ given by

$$f_{ij} = m_{ij} \Omega_{ij}, \quad f_{ii} \equiv 0$$

(2.9)

with $\Omega_{ij}$ the rate coefficient for momentum transfer from species $i$ to species $j$ and $m_{ij}$ the reduced mass of the $i - j$ system. The friction a species encounters in the plasma is hence roughly proportional to the difference between the velocity of the species and the average velocity of the other species.

The bulk mass and momentum conservation equation now follow from the summation of (2.6) and (2.7). We define the mass density $\rho_b$ as

$$\rho_b = \sum_i n_i \rho_i$$

(2.10)

and the bulk velocity $\vec{v}_b$:

$$\vec{v}_b = \frac{\sum_i n_i m_i \vec{v}_i}{\rho_b}.$$  

(2.11)

The bulk mass conservation equation is obtained by summing (2.6) for all $i$:

$$\frac{\partial \rho_b}{\partial t} + \vec{\nabla} \cdot (\rho_b \vec{v}_b) = 0.$$  

(2.12)

The production terms at the right hand of the equation cancel, because the total mass is conserved in each individual reaction.

The bulk velocity conservation is given by

$$\frac{\partial \rho_b \vec{v}_b}{\partial t} + \vec{\nabla} \cdot (\rho_b \vec{v}_b \vec{v}_b) + \vec{\nabla} \cdot \Pi + \vec{\nabla} p = \sum_i n_i \vec{F}_R$$  

(2.13)
A detailed derivation of (2.13) is offered in [5, 12, 13]. The friction and thermoforetic force cancel due to Newton’s third law: the force species $i$ exerts on species $j$ causes an opposite and equal force from $j$ on $i$, and when all forces are summed, these cancel. Furthermore, the contributions due to reactions cancel because each individual reaction conserves momentum.

The deviations from a particular species from the average is treated in PLASIMO by a diffusion model. In this diffusion model, both drift and diffusive terms are included. PLASIMO offers two diffusion models: self-consistent diffusion and Fick diffusion.

The self-consistent diffusion treatment in PLASIMO is described in detail in [5, 12]; for the mathematical derivation, we refer to these works. Essentially, (2.7) is rewritten so that the friction becomes the leading term. This friction force inhibits diffusion, while the other forces, such as the thermoforetic force, the electric force, and the partial pressure gradient may cause it. The first two terms of (2.7) can generally be neglected, as they are typically much smaller than the friction and partial pressure gradient due to the modest diffusive velocities. Furthermore, we assume the only external force acting on the species is the electric force in a field $\vec{E}$, and viscosity is neglected. Note that this electric field includes the ambipolar field, which is caused by the fact that the electron mobility is higher than the ion mobility.

Substituting (2.8) in this simplified form, we obtain [5, 12]:

$$\sum_j f_{ij} (\vec{\chi}_i - \vec{\chi}_j) = -\nabla p_i + n_i q_i \vec{E} + n_i R^T_i$$

(2.14)

with $q_i$ the charge of the particle and $\vec{\chi}$ the diffusive velocities, which are given by

$$\vec{\chi}_i = \vec{v}_i - \vec{v}_b.$$  

(2.15)

the set of equations (2.14) can now be solved, given proper boundary conditions. In this solution procedure, one equation is eliminated by the constraint that

$$\sum_i n_i m_i \vec{\chi}_i = 0$$  

(2.16)

which is a mathematical statement that, due to Newton’s third law, the diffusive fluxes cannot cause net mass transfer; the net mass transfer is described in the bulk Navier-Stokes equation (2.13).

The self-consistent diffusion approach is quite general, but also fairly complicated and occasionally prone to numerical instabilities. For many problems, a simpler approach based on the empirical Fick’s law is in order. In this approach, it is assumed thermoforetic forces and temperature gradients are negligible, and it is furthermore assumed there is one dominant background gas, labeled $g$. In this case, the diffusive transport is dominated by the friction between the diffusing species and the background gas.
Because of the small number density of the diffusing species, the momentum transfer from the diffusing species to the background gas is small. Hence, the diffusive velocity of the background gas is small. This leads to

\[ n_i f_g \vec{\chi}_i = -\vec{\nabla} p + n_i q_i \vec{E} \]  

(2.17)

Using the ideal gas law and the assumption that temperature gradients are negligible, we obtain

\[ n_i n_g m_i \Omega_{ig} \vec{\chi}_i = -k_B T_i \vec{\nabla} n_i + n_i q_i \vec{E} \]  

(2.18)

Substituting the definition of the diffusion coefficient \( D_i \)

\[ D_i = \frac{k_B T_i}{m_i n_g \Omega_{ig}} \]  

(2.19)

and the mobility \( \mu_i \)

\[ \mu_i = \frac{q_i}{m_i n_g \Omega_{ig}} \]  

(2.20)

we obtain the familiar drift-diffusion equation:

\[ n_i \vec{\chi}_i = -D_i \vec{\nabla} n_i + \mu_i n_i \vec{E}. \]  

(2.21)

Here, \( z_i \) is the charge number of the particles. Note that \( \mu_i \) may be negative. The complexity of (2.21) can be reduced even further if \( \vec{E} \) is chiefly caused by the ambipolar field. This ambipolar field is the field that is generated in the plasma because the more mobile electrons diffuse faster than the ions. Quasineutrality demands that both fluxes are equal. In this case, it can be shown [14] that the diffusion coefficient \( D_i \) for an ion is modified to the ambipolar diffusion coefficient \( D_{amb} \)

\[ D_{amb} = D_i \left( 1 + \frac{T_e}{T_i} \right). \]  

(2.22)

Equation (2.22) is only valid in case only a single ionic species exist. The end result is the well-known expression of Fick’s law

\[ n_i \vec{\chi}_i = -D_{amb} \vec{\nabla} n_i. \]  

(2.23)

In the case multiple ionized species exist, the multi-ion diffusion [5, 12] model that is present in PLASIMO may be used.

Regardless of the diffusion model used, the total mass conservation equation for a species \( i \) reads:

\[ \frac{\partial m_i n_i}{\partial t} + \vec{\nabla} \cdot (m_i n_i \vec{v}_b) + \vec{\nabla} \cdot (m_i n_i \vec{\chi}_i) = \left( \frac{\partial m_i n_i}{\partial t} \right)_c. \]  

(2.24)
The equations describing $\vec{\chi}$ depend on either the density or the density gradient; this allows a splitting along these lines of this terms, leading to a form that is readily cast into a $\phi$-equation (2.5):

$$\frac{\partial m_i n_i}{\partial t} + \nabla \cdot (m_i n_i \vec{v}_b + \vec{\chi}'_i) - \nabla \cdot (m_i C_{\text{diff}} \hat{n}_i) = \left( \frac{\partial m_i n_i}{\partial t} \right)_c \quad (2.25)$$

with $\vec{\chi}'$ the part of the diffusive velocity that depends on $n_i$ and $C_{\text{diff}} \hat{n}_i$ the part of the diffusive velocity that depends on $\hat{n}_i$. As a simple example, we can consider (2.25) in case the ambipolar Fick's law is used:

$$\frac{\partial m_i n_i}{\partial t} + \nabla \cdot (m_i n_i \vec{v}_b) - \nabla \cdot (m_i D_{\text{amb}} \hat{n}_i) = \left( \frac{\partial m_i n_i}{\partial t} \right)_c \quad (2.26)$$

In the strongly transient plasmas treated in this work (the pulsed capillary discharge and the spark gap), the dominant terms in 2.25 are $\frac{\partial m_i n_i}{\partial t}$, $\nabla \cdot (m_i n_i \vec{v}_b)$ and $\left( \frac{\partial m_i n_i}{\partial t} \right)_c$. Generally, diffusion in these dense plasmas is negligible at the time scales of the discharge, which is $\approx 100$ ns for the capillary discharge and $\approx 1$ ns for the spark gap. Chapter 6 gives a quantitative justification of this claim. However, this makes (2.26) by no means trivial: the production and bulk flow terms vary so rapidly that the "lagging" of the system, represented by the first term in (2.26), leads to large deviations from equilibrium.

### 2.3.1.2 The energy conservation equations

As mentioned in Section 2.3, we allow for two different temperatures in the plasma: one temperature for the electrons $T_e$ and one for the heavy particles $T_h$. For electrons, the temperature equation becomes [3]:

$$\frac{3}{2} \frac{\partial n_e k_B T_e}{\partial t} + \frac{3}{2} \nabla \cdot (n_e k_B T_e (\vec{v}_B + \vec{\chi} e)) + p_e \vec{v}_B \cdot \vec{v}_B + (\nabla \cdot \vec{v}_B) : \Pi_e - \nabla \cdot \left( \lambda_e \nabla T_e \right) = \int E^T \left( \frac{\partial f_e}{\partial t} \right) c d^3w \quad (2.27)$$

It is noted that for the cases treated in this thesis, the first, fifth and sixth term are dominant.

The heavy particle temperature equation can be obtained by summing over all heavy particle equations:

$$\frac{3}{2} \frac{\partial n_h k_B T_h}{\partial t} + \frac{3}{2} \nabla \cdot (n_h k_B T_h \vec{v}_B) + p_h \vec{v}_B \cdot \vec{v}_B + (\nabla \cdot \vec{v}_B) : \Pi_B - \nabla \cdot \left( \lambda_h \nabla T_h \right) = \sum_i \int E^T_i \left( \frac{\partial f_i}{\partial t} \right) c d^3w \quad (2.28)$$
It is noted that the net diffusive velocity of all heavy particles summed must be small because of the mass ratio of electrons and ions. The right-hand terms correspond to all the collisions with other particles in which the thermal energy changes. Such an interaction can be seen as a discrete event, in which some particles appear or disappear at a position in velocity space. Because the integrals are performed over the thermal energy of the particles, this leads to a thermal energy source that is equal to the thermal energy corresponding to the position in phase space. This is schematically shown in Figure 2.1.

In the case of an elastic interaction, a particle merely changes place in velocity space, while in the case of inelastic interactions, it is also possible that new particles are added and removed to the velocity space of a species. In order to evaluate the energy source, the contribution of each elastic or inelastic particle interaction process has to be evaluated.

One important elastic process is elastic energy transfer from electrons to heavy particles. In this process, an electron and a heavy particle with a certain velocity collide, exchanging energy. These collisions are frequent, with typical frequencies of $10^{11}$ s$^{-1}$ for the plasmas treated with PLASIMO in this work. Hence, we do not describe each individual collision, but rather look to the average effects of all collisions. A highly
detailed derivation of the exchange of energy from electrons to heavy particles \( S_{el} \) is given in [14], with \( S_{el} \) the energy source term source for the heavy particles, and \(-S_{el}\) the energy source term for the electrons. Essentially, \( S_{el} \) is given by

\[
S_{el} = \sum_i k_i E_{i_{exch}}
\]  

(2.29)

with \( k_i \) the collision rate between electrons and heavy particles of species \( i \) and the average energy exchanged per collision between an electron and a particle of species \( i \). \( E_{i_{exch}} \) is given by [14]

\[
E_{i_{exch}} = 3 \frac{m_i m_e}{(m_i + m_e)^2} k_B (T_e - T_h) \approx 3 \frac{m_e m_i}{m_i} k_B (T_e - T_h) .
\]  

(2.30)

The collision rate is generally a function of temperature, and depends on the particles involved in the collision. The computation of rates in PLASIMO is discussed in detail in Chapter 3.

Another important process is Ohmic heating of the electrons. The electrons are accelerated in the electric field, gaining directed momentum in the opposite direction. When the electrons scatter, this thermal energy is isotropized. The combined effect of acceleration and scattering leads to an increase in \( \vec{u} \) and hence in thermal energy. This energy dissipation \( S_{Ohm} \) can be described using Ohms law

\[
S_{Ohm} = \sigma E^2
\]  

(2.31)

with \( \sigma \) the conductivity and \( E \) the magnitude of the external electric field. In this work, we use the Frost mixture rules [15] and hence account for the contribution of each heavy particle \( i \) to the scattering of electrons in the computation of \( \sigma \). The Frost mixture rules are the method of choice for obtaining \( \sigma \) for dense plasmas [16], such as the plasmas treated in this work.

In inelastic processes, particles of some species are destroyed, while particles of other species are created, in what is essentially a chemical reaction. A general reaction equation is schematically given by

\[
A + B + \ldots \rightarrow P + Q + \ldots
\]  

(2.32)

Here, the letters denote species that may be heavy particles or electrons. In order to evaluate the energy source that is caused by reactions, it is necessary to know the energy gain for both the electrons and heavy particles that is the result of a single reaction, and the rate of these reactions. We can distinguish between four different cases:

- **Only heavy particles in** (2.32): In this case, energy conservation dictates that the heavy particles gain an amount of kinetic energy that is equal to the difference
in formation energy between the particles on the right-hand side of (2.32) and the particles on the left-hand side. The rate is determined by the heavy particle temperature.

- **Electrons on both sides of** (2.32): In this case, the electrons determine the rate and receive the (positive or negative) energy source. This is a very important case because of the high velocity of electrons, and hence high reaction rates for electron reactions, in a typical plasma.

- **Electrons only on the left-hand side of** (2.32): In this case, the light and fast electrons generally determine the reaction rate. In case of an exothermal reaction, the energy difference between the reactants on the left and right-hand side is a (positive) source term for the heavy particles; in case of an endothermal reaction, the energy difference is a (negative) source term for the electrons.

- **Electrons only on the right-hand side of** (2.32): In this case, the heavy particles determine the reaction rate. If the average thermal energy of the heavy particles is not large compared to the reaction energy, and the reaction is endothermal, the reaction chiefly leads to a source term for the heavy particles. In case of an exothermal reaction, the distribution of the resulting kinetic energy over the particles on the right-hand side cannot be determined solely from considerations of simple momentum and energy conservation. There are no examples of reactions of this kind in this Thesis.

We see that the particle that determines the rate does not necessarily received the energy. A correct implementation of this is one of the improvements made to PLASIMO in this work.

The specifics of each reaction are obtained from literature. PLASIMO offers a wide variety of methods to implement various reaction rates, and automatically determines to whether to add the source term to the electrons or the ions. The reactions that are used in the specific models created with the PLASIMO are described in detail in the Chapters that deal with the applications.

If necessary, other processes, such as photoionization, two-particle recombination, emission, stimulated emission, Bremsstrahlung and absorption of photons can also be added to this general framework. This allows for a general two-temperature treatment of reactions in PLASIMO.

### 2.3.2 The multi-fluid description in MD2D

Where PLASIMO is quite suitable for the simulation of dense plasmas, the assumption of quasi-neutrality makes it unsuitable for the simulation of plasmas in which there are
significant deviations of quasi-neutrality, such as glow discharges. In this work, the MD2D code is used or the simulation of these discharges. The MD2D code is described in detail in [17, 18]. A brief overview of the code will be given here.

A central assumption in MD2D is that the plasma can be considered to exist in a neutral buffer gas. This buffer gas is assumed not to be heated by the plasma. Furthermore, the destruction of buffer gas atoms by reactions is neglected. It is, however, possible to supply externally-computed velocity and density fields to the buffer gas. These influence the other species in the plasma. However, the effect of the other species on the velocity and density of the buffer gas is neglected. These assumptions can only be valid if the degree of ionization is low. This precludes the use of MD2D for the simulation of the arc plasmas that are treated in this work, as gas heating, depletion of the neutral buffer gas and/or flow induced by plasma heating may play a key role in these systems.

The species conservation equations are solved in MD2D using the drift-diffusion approach. The species conservation equation is given by (2.6), while the drift-diffusion equation is given by

$$ n_i \vec{v}_i = -D_i \nabla n_i + \mu_i n_i \vec{E} + n_i \vec{v}_b. \quad (2.33) $$

In MD2D, it is assumed the heavy particles are not heated. Hence, the only energy equation that is solved is the electron energy equation, which is given by [19]

$$ \frac{\partial n_e \epsilon}{\partial t} + \nabla \cdot \left( \frac{5}{3} n_e \epsilon \vec{v}_e - \frac{5}{3} n_e D_e \nabla \epsilon \right) = -e \vec{v}_e \cdot \vec{E} - S_r \quad (2.34) $$

with $\epsilon$ the energy per electron and $S_r$ the energy source from reactions. The second term in the brackets is the result of thermal conduction. Note that there is no assumption of Maxwell equilibrium in MD2D; hence, it is not strictly possible to use temperatures. The transport coefficients and reactions are obtained using a Boltzmann solver.

The electric field is obtained by solving Poisson’s equation

$$ \nabla \cdot \left( \epsilon_p \vec{E} \right) = \rho_e \quad (2.35) $$

with $\epsilon_p$ the dielectric permittivity and $\rho_e$ the electric charge density.

The extendability of MD2D is more limited than that of PLASIMO. Furthermore, there is only a limited set of features that is only present in MD2D and not in PLASIMO. One of the focuses of the plasma modeling efforts at Eindhoven University of Technology is the further integration of features from MD2D in PLASIMO.

## 2.4 Time-dependent modeling

The modeling of transient plasmas requires time-dependent modeling. The subject of time-dependent modeling is an active branch of mathematics. A very short introduction to the numerical method used in this work is given in this Section; for details, we
2.4. Time-dependent modeling refer the reader to [11]. Rather, we will discuss some salient problems that are inherent to this type of modeling: non-linearity and multiple timescales. Then, we will discuss the numerical method used in PLASIMO to solve these equations, which is robust with respect to these problems.

2.4.1 Discretization

As mentioned, all conservation equations that are used to describe the plasmas in PLASIMOS can be cast in the form of a generalized equation, the \( \phi \)-equation (2.5). In steady state, i.e. ignoring the first term, this equation is discretized on a control volume grid. Essentially, this discretization means that the values of \( \phi \) are obtained in a point \( P \) that lies in the control volume, usually in the center, by solving an algebraic equation, in which the value of \( \phi \) is linked with the fluxes originating from the neighboring grid cells, and local source terms. The discretized equation [11] has the following form:

\[
a_P \phi_P = a_N \phi_N + a_E \phi_E + a_S \phi_S + a_W \phi_W + S_c V + S_p V \phi_P
\]

(2.36)

with \( a \) the discretization coefficients [11], which depend on the transport coefficients of the plasma, and \( V \) the volume of the grid cell. Thus, the value of \( \phi \) in a point \( P \) depends on its value neighboring points \( E, W, N \) and \( S \), and the source terms \( S_c \) and \( S_p \). The splitting of the source term in the negative terms \( S_p \) and the positive terms \( S_c \) allows rewriting of (2.36)

\[
(a_P - S_p V) \phi_P = a_N \phi_N + a_E \phi_E + a_S \phi_S + a_W \phi_W + S_c V
\]

(2.37)

This linearization increases the prefactor of \( \phi_P \). Hence, changes in the source terms and the neighbor points have less influence on \( \phi_P \), leading to more stable behavior. This can be seen by imagining a disturbance \( \Delta \) on the right hand side of (2.37): The larger the prefactor of \( \phi_P \), the smaller the influence of this disturbance becomes.

It is noted that in (2.36) and (2.37), the value of \( \phi_P \) depends on its neighboring points, and these depends on their neighboring points, etc, meaning that an influence on one point is generally felt by all. Equation (2.37) is solved by casting it into a matrix-vector form.

2.4.2 Non-linearity

As mentioned, the various coefficients in the equations that describe the plasma are, generally, functions of plasma parameters; for instance, the heat transfer coefficient \( \lambda \) is a function of temperature and the density of the various species, and the reaction rates that determine the energy sources are strong functions of the temperature as well.
Because the results of one equation depend on the results of other equations, it is necessary to iteratively solve each equation, substituting the result of one equation in the next equation. Eventually, the results may converge to a self-consistent solution.

A problem with this method is the strong non-linearity of the source terms and the transport coefficients: small variations in the temperature may lead to large variations in the value of $S_p$ and $S_c$ and the discretization coefficients. In practice, these variations are particularly large for the reaction source terms, which depend superexponentially on the temperature for many important reactions such as ionization. These reaction source terms are particularly important in these highly transient plasmas, in which the composition may rapidly change.

A way of solving this is by using underrelaxation, in which the new solution of the equation is not the solution of (2.37), but rather a weighed average of the new solution and the solution of the previous iteration $\phi^*$:

\[
a^{-1} (a_P - S_P V) \phi_P = a_N \phi_N + a_E \phi_E + a_S \phi_S + a_W \phi_W + S_c V + (1 - \alpha) a^{-1} \phi^* \tag{2.38}
\]

The part of the solution determined at the current time step is called the underrelaxation factor $\alpha$, which should be between 0 and 1. (2.38) shows that the prefactor of $\phi$ becomes even larger, making the result again less susceptible to changes and the solution more robust. Occasionally, overrelaxation, in which $1 < \alpha < 2$, may be used, in which stability is sacrificed for a more rapid convergence. A disadvantage of underrelaxation is that the slower changes in $\phi_P$ mean that it takes more iterations to move from an intermediate solution to a final solution. By tuning the $\alpha$ of the various equation, stable, yet fairly rapid convergence may be obtained. This is one of the greatest practical challenges in the numerical method used in this work.

### 2.4.3 Multiple timescales

Numerical solution of the transient problems treated in this Thesis is done by solving the differential equations that describe it at various discrete time steps. The problems we treat exhibit vastly different timescales: the hydrodynamic flow times, the time it takes for reactions to change the contribution significantly, the time it takes for the temperatures to respond to changing plasma and external conditions, the total time of the discharge, etc. Problems in which very different timescales play a role are difficult to solve because they have to be solved at enough timesteps to resolve the shortest timescale, for a time which is determined by the longest timescale.

Problems of varying timescales should be treated with implicit numerical methods. In an implicit numerical method, the value of $\phi$ at the next time step $\phi^1$ is obtained using transport coefficients, sources and values of the neighboring points that are evaluated
at this next time step. This can be contrasted with an explicit method, in which only coefficients, sources and values of the neighboring points at the present time are used. Mathematically, we obtain the following discretized $\phi$-equation [11]:

$$\frac{CV}{\delta t} \left( \phi_1^1 - \phi_0^0 \right) = a_N^1 \phi_N^1 + a_E^1 \phi_E^1 + a_S^1 \phi_S^1 + a_W^1 \phi_W^1 - a_P\phi_P^1 + S_c^1 V + S_p^1 V \phi_P^1$$  \hspace{1cm} (2.39)

with $\delta t$ the time step. This scheme is known as an Implicit Euler scheme [5]. It can be rewritten to:

$$\left( \frac{CV}{\delta t} - S_p^1 V + a_P^1 \right) \phi_P^1 = a_N^1 \phi_N^1 + a_E^1 \phi_E^1 + a_S^1 \phi_S^1 + a_W^1 \phi_W^1 + S_c^1 V + \frac{CV}{\delta t} \phi_P^0$$  \hspace{1cm} (2.40)

It can be seen that this implementation of time-dependence does not change the structure of the equation at all; it merely changes the term in front of $\phi_P^1$ and adds an extra source term. It is also possible to underrelax (2.40):

$$\alpha^{-1} \left( \frac{CV}{\delta t} - S_p^1 V + a_P^1 \right) \phi_P^1 = a_N^1 \phi_N^1 + a_E^1 \phi_E^1 + a_S^1 \phi_S^1 + a_W^1 \phi_W^1 + S_c^1 V + \frac{CV}{\delta t} \phi_P^0 + (1 - \alpha) \alpha^{-1} \phi^*$$  \hspace{1cm} (2.41)

It can be seen that both time-dependence and underrelaxation have the effect of adding an additional source term on the right-hand side of the equation, and increasing the term $\phi_P^1$. Hence, they both improve the stability of the solution procedure.

A smaller timestep $\delta t$ will lead to a larger factor in front of $\phi_P^1$ and hence improved stability. Shorter timesteps are also more accurate, because the terms on the right-hand side are updated more frequently, reducing the time-discretization error. Of course, smaller time steps mean that the computation time will increase, because a converged solution of the set of discretized differential equations has to be found at each time step. This can be alleviated by reducing the underrelaxation factors. The reduction in stability this causes is compensated by the larger value of $\frac{CV}{\delta t}$ in front of $\phi_P^1$.

### 2.5 Multi-region modeling

In the computations on the pulsed capillary discharge waveguide (Chapters 5 to 8), the modeling is not limited to the modeling of the plasma. Instead, the heating of the wall is modeled as well, because it was found to be essential for an accurate description of the plasma, and because the wall properties are interesting themselves, e.g. for predicting ablation.
Mathematically, the wall temperature depends on the heavy particle temperature equation, as these exchange heat at the plasma-wall boundary. The differential equation that governs the wall temperature $T_{\text{wall}}$ is given by

$$\frac{\partial C_{\text{wall}} T_{\text{wall}}}{\partial t} - \nabla \cdot (\lambda_{\text{wall}} \nabla T_{\text{wall}}) = 0 \quad (2.42)$$

with $C_{\text{wall}}$ the wall thermal capacity and $\lambda_{\text{wall}}$ the wall thermal conductivity. Both $C_{\text{wall}}$ and $\lambda_{\text{wall}}$ are functions of $T_{\text{wall}}$, necessitating a numerical solution of (2.42).

An obvious solution would be to extend the grid at which the heavy particle temperature (2.28) is solved to encompass a fraction of the wall. The terms needed for the description of the wall in (2.42) are then a subset of the terms in (2.28); hence, by locally replacing the source terms and transport coefficients in (2.28) with those in (2.42), the temperature in the wall can be solved.

This method, while applicable to many different situations, and used in the modeling of pulsed capillary discharge waveguides [20], is in fact quite poorly suitable for accurate modeling of the wall temperature in this particular. The reason lies in the very different length scales in the plasma-wall system, a problem echoing the time scale problem discussed in Section 2.4.1. The plasma channel is typically in the order of 100 $\mu$m in radius, while the heat from the plasma significantly penetrates the walls by only about 1$\mu$m in the time scales of interest. Solving the plasma-wall system on a grid sufficiently fine to accurately describe the wall would require such a fine grid that the computation would become unmanageably slow. The technique of grid stretching [11], in which the size of the cells is locally decreased, could partially alleviate this problem, but at the expense of very large discretization errors [11].

A better solution would be to solve (2.28) in the plasma, and (2.42) in the wall, and to impose boundary conditions on both stating continuity of thermal flux and temperature. In this scheme, (2.28) is solved, using the wall temperature predicted by (2.42). This produces a heat flux into the wall. This heat flux is then used as a boundary condition to (2.28), which is solved, leading to a new boundary condition for the temperature, etc.

The advantage of this scheme is that the equations can be solved on grids with very different grid cell sizes, greatly reducing the computational cost. The numerical instabilities that may result from introducing an artificial boundary can be reduced by underrelaxing the boundary conditions. In this case, the temperature or temperature gradient is not updated to its newly computed value, but rather to a blend of the value at the previous iteration and the new solution, analogous to the technique in (2.4.2). In the implementation of this method, it is possible to set either underrelaxation factor independently; practice has taught us that setting either, but not both, to a small underrelaxation ($\alpha \approx 0.9$) produces fast and stable convergence. Furthermore, it was found that the underrelaxation of the plasma-wall coupling has a strong effect on convergence.
The combination of the techniques in this Section and Section 2.4 have allowed us to greatly enhance the model in terms of computational speed. While the first models of the pulsed capillary discharge waveguide that were created had computation times in the order of one week, the current model for one-dimensional channels requires a computational time of about four hours.

## 2.6 Conclusions

In this Chapter, we have used the Boltzmann Transport Equation to derive the well-known specific conservation equations for particles, momentum and energy. Particular attention is payed to the assumptions that are made to arrive at the resulting equations. The physical interpretation of the most critical assumptions is discussed.

For the momentum equation, an intensive and an intensive form have been derived, the former describing momentum conservation and the latter describing the velocity. Furthermore, an extrinsic and an intrinsic energy equation have been derived. The source terms of the intrinsic energy equation are written in a form that has a clear physical interpretation.

These equations for one species have been used to construct a multi-fluid description of a plasma consisting of several species, in which the light weight and consequently different behavior of the electrons is used to reduce the complexity. Equations for the bulk transport and diffusion in \textsc{Plasimo} are presented. Furthermore, the two-temperature approximation is discussed, with an emphasis on the correct implementation of the source terms. A rudimentary description of the MD2D code is given, with the extensions to the equations that are the result of the addition of bulk transport to the MD2D code for the modeling in this work.

A short discussion on the practical aspects of time-dependent modeling is given, in particular the consequence of having multiple time scales in the model. It is concluded that accurate, stable and relatively fast convergence can be obtained by using small timesteps combined with little underrelaxation.

In the applications of this framework, it was found necessary to predict the thermal properties of walls surrounding the plasma. Two methods are discussed; the common method of extending the plasma grid, and a multi-domain method in which the wall and plasma are on separate grids. It is concluded that for this particular application, the latter is better. This method does lead to an artificial boundary condition in the plasma, and the numerical method to handle this is discussed.
2.A Derivation of the fluid conservation equations from the Boltzmann Transport Equation

In this Appendix, we will derive the fluid conservation equations of mass, momentum and energy for a single species from the Boltzmann transport equation. The extension of this result to a plasma is detailed in Section 2.3 of this Chapter. This derivation roughly follows the derivations given in [9, 10]

2.A.1 The distribution function

The position of particles in the six-dimensional\(^1\) phase space is described by a distribution function \(f(\vec{r}, \vec{w}, t)\). Here, \(\vec{r}\) denotes the spatial coordinate, while \(\vec{w}\) denotes the velocity coordinate. This distribution function is defined in such a way that \(f(\vec{r}, \vec{w}, t)d^3rd^3w\) is equal to the number of particles in a volume element \(d^3rd^3w\) in the phase space on time \(t\).

In the fluid approach, \(d^3rd^3w\) is not infinitesimally small in the mathematical sense of the word; Instead, it represents a volume that is much smaller than the typical dimensions of the system, yet contain a large number of particles, large enough to have meaningful statistics. The existence of such volumes is a necessary condition for the validity of a fluid description. Because the volume elements are very small, the summation over the elements can be approximated by an integral:

\[\sum f(\vec{r}, \vec{w}, t)d^3rd^3w \approx \int f(\vec{r}, \vec{w}, t)d^3rd^3w.\]  \hspace{1cm} (2.43)

The distribution function \(f(\vec{r}, \vec{w}, t)\) can be normalized by acknowledging that there are \(N\) particles in the system:

\[\int f(\vec{r}, \vec{w}, t)d^3rd^3w = N.\]  \hspace{1cm} (2.44)

In order to find the equation of motion for the distribution function, we investigate how it evolves after an infinitesimal timestep \(\delta t\). In the case there is no collision in this timeframe, the movement of particles in phase space is restricted to a position change from \(\vec{r}\) to \(\vec{r} + \vec{w}\delta t\), and a position in velocity space change from \(\vec{w}\) to \(\vec{w} + \vec{F}m\delta t\), with \(\vec{F}\) the external forces and \(m\) the particle mass. This gives the equality

\[f(\vec{r} + \vec{w}\delta t, \vec{w} + \vec{F}m\delta t, t + \delta t)d^3rd^3w = f(\vec{r}, \vec{w}, t)d^3rd^3w.\]  \hspace{1cm} (2.45)

\(^1\)three spatial and three velocity dimensions
As shown in [10], the volume of $d^3r'd^3w'$ is the same as the volume in $d^3rd^3w$. This means we can simplify (2.45) to
\[
f(\vec{r} + \vec{w}\delta t, \vec{w} + \vec{F}_m \delta t, \vec{w}, t + \delta t) = f(\vec{r}, \vec{w}, t).
\] (2.46)

In a plasma, the collisions are generally important. This introduces an additional term, which represents the change in $f$ due to collisions:
\[
f(\vec{r} + \vec{w}\delta t, \vec{w} + \vec{F}_m \delta t, \vec{w}, t + \delta t) = f(\vec{r}, \vec{w}, t) + \left(\frac{\partial f(\vec{r}, \vec{w}, t)}{\partial t}\right)_c \delta t.
\] (2.47)

If we use a Taylor expansion on the left hand side we obtain:
\[
f(\vec{r}, \vec{w}, t) + \left(\delta t \vec{w} \cdot \vec{\nabla}_r + \delta t \frac{\vec{F}_m}{m} \cdot \vec{\nabla}_w + \delta t \frac{\partial}{\partial t}\right) f(\vec{r}, \vec{w}, t) =
\]
\[
f(\vec{r}, \vec{w}, t) + \left(\frac{\partial f(\vec{r}, \vec{w}, t)}{\partial t}\right)_c \delta t.
\] (2.48)

This can be simplified to:
\[
\left(\frac{\partial}{\partial t} + \vec{w} \cdot \vec{\nabla}_r + \frac{\vec{F}_m}{m} \cdot \vec{\nabla}_w\right) f(\vec{r}, \vec{w}, t) = \left(\frac{\partial f(\vec{r}, \vec{w}, t)}{\partial t}\right)_c.
\] (2.1)

Equation 2.1 is generally known as the Boltzmann Transport Equation. This equation will be the foundation of the derivation of the fluid conservation equations.

### 2.A.2 The moments of the Boltzmann Transport Equation

In this section, we outline a method to obtain the mass conservation equation, the momentum conservation law and the energy conservation law from the Boltzmann transport equation. For this, we multiply the Boltzmann Transport Equation with a function $g(\vec{w})$ of the velocity and integrate over the whole velocity space. This function is equal to $m|\vec{w}|^0$, or $m$, for the zeroth moment, $m\vec{w}$ for the first moment, and $1/2m|\vec{w}|^2$ for the second moment. By evaluating
\[
\int g(\vec{w}) \frac{\partial f}{\partial t} d^3w + \int g(\vec{w}) \vec{w} \cdot \vec{\nabla}_w f d^3w + \int g(\vec{w}) \frac{\vec{F}_m}{m} \cdot \vec{\nabla}_w f d^3w =
\]
\[
\int g(\vec{w}) \left(\frac{\partial f}{\partial t}\right)_c d^3w
\] (2.49)

---

2The ()c denotes the effects of collisions
3The subscripts on the nabla-operator denote the variable to which is differentiated
for each of the three moments, we can obtain the mass conservation equation from the zeroth moment, the momentum conservation equation from the first moment and the energy conservation equation from the second moment. This shall be carried out in the next sections. These equations are derived in a spatially differential formulation. By integrating over a volume element, an integral formulation may be obtained.

2.A.3 The zeroth moment: the mass conservation law

The equation for the zeroth moment is

\[
\int m \frac{\partial f}{\partial t} \, d^3w + \int m \bar{w} \cdot \nabla_r f \, d^3w + \int \vec{F} \cdot \nabla_{\vec{w}} f \, d^3w = \int m \left( \frac{\partial f}{\partial t} \right) c \, d^3w. \tag{2.50}
\]

We want to rewrite this equation in more convenient terms. We will carry this out term by term. For the first term, we note that the volume of the integration does not change. This means the sequence of differentiation and integration can be changed. Using this, and (2.44), we obtain:

\[
\int m \frac{\partial f}{\partial t} \, d^3w = m \frac{\partial n}{\partial t}, \tag{2.51}
\]

with \( n \) the particle density.

For the second term, we use that \( \bar{w} \) does not depend on explicitly on \( \vec{r} \). This means that we can change the order of differentiating and integrating, and obtain:

\[
\int m \bar{w} \cdot \nabla_r f \, d^3w = \int \sum_{i=1}^{3} m w_i \frac{\partial}{\partial r_i} f \, d^3w = \sum_{i=1}^{3} \frac{\partial}{\partial r_i} \int m w_i f \, d^3w = m \nabla_r \cdot \int \bar{w} f \, d^3w. \tag{2.52}
\]

We define the average of a quantity \( x \), denoted by \( \langle x \rangle \) as

\[
\langle x \rangle = \frac{\int x f \, d^3w}{\int f \, d^3w}. \tag{2.53}
\]

Hence, we can split the velocity \( \bar{w} \) in an average value of \( \bar{w} \), denoted with \( \bar{v} \):

\[
\frac{\int \bar{w} f \, d^3w}{\int f \, d^3w} = \bar{v}, \tag{2.54}
\]

\(^{4}\)A point that is as subtle as it is crucial. The integration is carried out over a six-dimensional space, thus, for each \( \vec{r} \), there is an integration over a whole three-dimensional space in velocity space. A dependence of \( \bar{w} \) on \( \vec{r} \) in the macroscopic system simply means that some parts of this cube are empty, and is thus expressed in \( f(\vec{r}, \bar{w}, t) \).
and in the deviations from this average are defined as $\vec{u}$:

$$\vec{w} - \vec{v} = \vec{u}. \quad (2.55)$$

Hence,

$$\langle \vec{u}\rangle = \langle \vec{w}\rangle - \langle \vec{v}\rangle = \vec{0}. \quad (2.56)$$

Physically, $\vec{v}$ is identified with a drift speed, while $\vec{u}$ is a thermal speed. This gives us as the expression for the second term:

$$\int \vec{w} \cdot \vec{\nabla} r f \, d^3w = \vec{\nabla} r \cdot \int \vec{f} d^3w = \vec{\nabla} r \cdot (n\vec{v}). \quad (2.57)$$

In the treatment of the third term, we apply partial integration over the terms of $\vec{w}$. In this partial integration, the staff term is zero, because there are no particles with infinite velocity. We obtain:

$$\int \vec{F} \cdot \vec{\nabla} w f \, d^3w = - \int f \cdot \vec{\nabla} w \vec{F} \, d^3w = 0. \quad (2.58)$$

This expression evaluates to if we only consider gravity, electric and Lorentz forces, because the forces, are not an explicit function of $\vec{v}$ parallel to the direction of the force [9].

The fourth term is treated similarly to the first term. This yields

$$\int m \left( \frac{\partial f}{\partial t} \right)_c \, d^3w = \left( m \frac{\partial}{\partial t} \int f \, d^3w \right)_c = \left( \frac{\partial mn}{\partial t} \right)_c. \quad (2.59)$$

Adding the terms, we obtain the mass conservation equation:

$$\frac{\partial mn}{\partial t} + \vec{\nabla} r \cdot (mn\vec{v}) = \left( \frac{\partial mn}{\partial t} \right)_c. \quad (2.2)$$

### 2.A.4 The first moment: the momentum conservation law

In order to obtain the momentum conservation law, we look at the first moment of the Boltzmann Transport Equation, which is obtained by setting $g(\vec{v})$ to $m\vec{w}$. Unlike the zeroth and second moments, the first moment yields a vector equation. This equation is given by:

$$\int m\vec{w} \frac{\partial f}{\partial t} \, d^3w + \int m\vec{w} \vec{v} \, d^3w + \int m\vec{w} \vec{F} \, d^3w = \int m\vec{w} \left( \frac{\partial f}{\partial t} \right)_c \, d^3w. \quad (2.60)$$
Again, this equation will be treated term by term.

The first term is treated similarly to the first term of (2.50). By noting that \( \vec{\omega} \) does not depend on time and that the integration volume is constant in time, we can put the time derivative in front of the integral, and obtain:

\[
\int m\vec{\omega} \frac{\partial f}{\partial t} d^3w = \frac{\partial}{\partial t} \int m\vec{\omega} f d^3w = \frac{\partial m\vec{\omega}}{\partial t}.
\] (2.61)

The second term differs significantly from that in (2.50) due to the dyadic product of \( \vec{\omega} \) in it. Noting again that \( \vec{\omega} \) is independent of \( r \), and using that the integration volume and \( m \) are independent of position, we obtain:

\[
\int m\vec{\omega} \vec{\omega} \cdot \vec{\nabla}_r f d^3w = m \int \sum_{i=1}^{3} \sum_{j=1}^{3} w_i \vec{e}_j w_i \frac{\partial}{\partial r_i} f d^3w =
\]

\[
m \sum_{i=1}^{3} \sum_{j=1}^{3} \frac{\partial}{\partial r_i} \int w_i \vec{e}_j w_i f d^3w
\] (2.62)

where \( \vec{e}_j \) is the unity vector. Thus, the result of (2.62) can be rewritten to

\[
m \sum_{i=1}^{3} \sum_{j=1}^{3} \frac{\partial}{\partial r_i} \int w_i \vec{e}_j w_i f d^3w = \sum_{i=1}^{3} \sum_{j=1}^{3} \frac{\partial}{\partial r_i} nm\langle w_i w_i \rangle \vec{e}_j.
\] (2.63)

We now split the terms of \( \vec{\omega} \) in the terms of \( \vec{v} \) and \( \vec{u} \), and obtain:

\[
\langle w_i w_j \rangle = \langle u_i u_j \rangle + \langle u_i v_j \rangle + \langle u_j v_i \rangle + \langle v_i v_j \rangle.
\] (2.64)

Using the fact that \( \vec{v} \) already is an average (cf. (2.54)), we obtain:

\[
\langle w_i w_j \rangle = \langle u_i u_j \rangle + v_j \langle u_i \rangle + v_i \langle u_j \rangle + v_i v_j.
\] (2.65)

Because \( \langle \vec{u} \rangle = 0 \) (cf. (2.56)), we obtain:

\[
\langle w_i w_j \rangle = \langle u_i u_j \rangle + v_i v_j.
\] (2.66)

Substitution this back into (2.62), we get:

\[
\int m\vec{\omega} \vec{\nabla}_r f d^3w = \sum_{i=1}^{3} \sum_{j=1}^{3} \frac{\partial}{\partial r_i} (mn\langle u_i u_j \rangle) \vec{e}_j + \sum_{i=1}^{3} \sum_{j=1}^{3} \frac{\partial}{\partial r_i} (mnv_i v_j) \vec{e}_j.
\] (2.67)

Now, we are left with two terms, one depending on the thermal speed and the other on the bulk velocity. The first term can be rewritten to a quantity with a physical interpretation that is more clear by defining the pressure tensor \( P \) as

\[
P = \sum_{i=1}^{3} \sum_{j=1}^{3} mn\langle u_i u_j \rangle.
\] (2.68)
In practice, it is convenient to split this tensor into the diagonal elements, that have the physical meaning of a pressure $p$, and the other terms, which are the viscous forces $\Pi$. For this, we define $p$ as

$$p = \frac{1}{3} \sum_{i=1}^{3} mn \langle u_i^2 \rangle. \quad (2.69)$$

Note that when assuming isotropy, all components of the thermal velocity are of equal size. Using this, and the definition of the viscous forces

$$\Pi = \sum_{i=1}^{3} \sum_{j \neq i} mn \langle u_i u_j \rangle \quad (2.70)$$

we obtain

$$P = \Pi + pI. \quad (2.71)$$

Here, $I$ is the unity tensor.

For the treatment of third term of (2.60), we split the vectors in them into their components. With some rearrangement, we obtain:

$$\int \vec{m} \vec{\tilde{a}} \cdot \nabla \vec{w} f d^3w = \int \sum_{j=1}^{3} \sum_{j \neq 1} \vec{w} \vec{\tilde{a}}_j F_1 \frac{\partial f}{\partial \vec{w}_1} d^3w \quad (2.72)$$

Next, we use integration by parts on the expression for a term $i = 1$ to obtain:

$$\int \sum_{j=1}^{3} \vec{w} \vec{\tilde{a}}_j F_1 \frac{\partial f}{\partial \vec{w}_1} d^3w = \int \sum_{j=1}^{3} \vec{w} \vec{\tilde{a}}_j F_1 f d\vec{w}_2 d\vec{w}_3 \bigg|_{-\infty}^{\infty} - \int f \vec{F}_1 \vec{\tilde{a}}_1 d^3w. \quad (2.73)$$

The staff term vanishes, because the velocity distribution goes superlinearly to 0. We have also assumed by moving the differential operator over the terms of $\vec{a}$ that $\frac{\partial f}{\partial \vec{w}_1} = 0$, which is the case for the acceleration induced by the electromagnetic and gravitational force. Carrying out the procedure in (2.73) *mutatis mutandis* and summing over all tree terms yields:

$$\int \vec{\tilde{w}} \vec{F} \cdot \nabla \vec{w} f d^3w = -n \vec{F}. \quad (2.74)$$

We have now treated all terms on the left-hand side of the equation. Using (2.54) on the first term, we obtain:

$$\frac{\partial mn \vec{\tilde{v}}}{\partial t} + \sum_{i=1}^{3} \sum_{j=1}^{3} \frac{\partial}{\partial \vec{r}_i} (mn \vec{v}_j \vec{\tilde{v}}_j) \vec{\tilde{e}}_j + \vec{\nabla}_r \cdot \Pi + \vec{\nabla}_r p - n \vec{F} = \int mn \vec{\tilde{v}} \left( \frac{\partial f}{\partial t} \right) \vec{c} d^3w.$$
For the first term, we have used that the average of $\bar{w}$ is $\bar{v}$. Equation (2.75) can be rewritten to an equation for the velocity by subtracting the mass conservation equation (2.2), multiplied with $m\bar{v}$:

$$\frac{\partial mn\bar{v}}{\partial t} - \frac{\partial m}{\partial t} + \sum_{i=1}^{3} \sum_{j=1}^{3} \frac{\partial}{\partial r_i} (mnv_i\bar{v}_j)\bar{e}_j - \sum_{i=1}^{3} \sum_{j=1}^{3} v_j \frac{\partial}{\partial r_i} (mnv_i)\bar{e}_j + \nabla_r \cdot \mathbf{I} + \nabla_r p - n\bar{F} = \int m\bar{w} \left( \frac{\partial f}{\partial t} \right)_c d^3w - m\bar{v} \left( \frac{\partial n}{\partial t} \right)_c .$$

(2.75)

By using the product rule on the first and third term, we obtain:

$$mn \frac{\partial \bar{v}}{\partial t} + \sum_{i=1}^{3} \sum_{j=1}^{3} mv_i \frac{\partial}{\partial r_i} (nv_i\bar{e}_j) + \nabla_r \cdot \mathbf{I} + \nabla_r p - n\bar{F} = \int m\bar{w} \left( \frac{\partial f}{\partial t} \right)_c d^3w - m\bar{v} \left( \frac{\partial n}{\partial t} \right)_c .$$

(2.76)

We can rewrite this into a more familiar form:

$$mn \frac{\partial \bar{v}}{\partial t} + nm(\bar{v} \cdot \nabla_r)\bar{v} + \nabla_r \cdot \mathbf{I} + \nabla_r p - nm\bar{a} = \int m\bar{w} \left( \frac{\partial f}{\partial t} \right)_c d^3w - m\bar{v} \left( \frac{\partial n}{\partial t} \right)_c .$$

(2.77)

2.4.5 The second moment: the energy conservation law

In a similar fashion, the energy conservation law can be derived from the second moment of the Boltzmann equation. It is noted that this equation is a scalar equation. For this, we multiply the Boltzmann Transport Equation with $1/2mw^2$, where $w$ is the absolute value of $\bar{w}$, to obtain:

$$\int \frac{1}{2} mw^2 \frac{\partial f}{\partial t} d^3w + \int \frac{1}{2} mw^2 \bar{w} \cdot \nabla_r f d^3w +$$

$$- \int \frac{1}{2} mw^2\dot{a} \cdot \nabla_r f d^3w = \int \frac{1}{2} mw^2 \left( \frac{\partial f}{\partial t} \right)_c d^3w .$$

(2.78)

Again, we will treat the terms one at at time.

In the first term, we again use the fact that $w$ does not depend on $t$ to change the order of differentiation and integration to obtain:

$$\int \frac{1}{2} w^2 \frac{\partial f}{\partial t} d^3w = \frac{1}{2} m \frac{\partial n\langle w^2 \rangle}{\partial t} = \frac{1}{2} m \frac{\partial n\langle u^2 \rangle}{\partial t} + \frac{1}{2} m \frac{\partial n v^2}{\partial t} .$$

(2.79)
For the splitting of $w$ in $u$ and $v$, we use that $\langle uv \rangle = 0$. Furthermore, we used that $v$ already is an average. By using (2.69), we can rewrite this to:

$$
m \int \frac{1}{2} w^2 \frac{\partial f}{\partial t} \, d^3 w = \frac{3}{2} \frac{\partial p}{\partial t} + \frac{1}{2} m \frac{\partial n v^2}{\partial t}.
$$

(2.80)

For the second term, we use that $w$ does not depend explicitly on $r$, to obtain:

$$
\int \frac{1}{2} m w^2 \vec{w} \cdot \vec{\nabla}_r f \, d^3 w = \int \sum_{j=1}^{3} \sum_{i=1}^{3} \frac{1}{2} m w_j^2 w_i \frac{\partial f}{\partial r_i} \, d^3 w = \sum_{j=1}^{3} \sum_{i=1}^{3} \frac{1}{2} m \frac{\partial n \langle w_j^2 w_i \rangle}{\partial r_i}.
$$

(2.81)

The next step is to split $w$ in $v$ and $u$:

$$
\langle w_j^2 w_i \rangle = v_j^2 v_i + \langle u_j^2 u_i \rangle + v_i \langle u_j^2 \rangle + \langle v_j^2 u_i \rangle + 2 v_i \langle u_j v_j \rangle + 2 \langle u_j v_i u_i \rangle.
$$

(2.82)

In this equation, the fourth and fifth term are zero because the average of $\vec{u}$ is zero. The second term of (2.82) is related to the heat flux vector $\vec{q}$:

$$
\sum_{j=1}^{3} \frac{1}{2} m \frac{\partial n \langle u_j^2 u_i \rangle}{\partial r_i} = \vec{\nabla} \cdot \left( \frac{m n \langle u^2 \vec{u} \rangle}{2} \right) = \vec{\nabla} \cdot \vec{q}
$$

(2.83)

The sixth term can be rearranged to:

$$
\sum_{i=1}^{3} \sum_{j=1}^{3} m \frac{\partial n \langle u_j v_i u_i \rangle}{\partial r_i} = \sum_{i=1}^{3} m \frac{\partial n \langle v_i u_i^2 \rangle}{\partial r_i} + \sum_{i=1}^{3} \sum_{j \neq 1}^{3} m \frac{\partial n \langle v_i u_i u_j \rangle}{\partial r_i} = \vec{\nabla} \cdot (p \vec{v}) + \vec{\nabla} \cdot (\vec{v} \cdot \vec{\Pi}).
$$

(2.84)

Finally, we use the assumption that the thermal velocity is isotropic on the third term of (2.82)

$$
\sum_{i=1}^{3} \sum_{j=1}^{3} \frac{1}{2} m \frac{\partial n v_i \langle u_j^2 \rangle}{\partial r_i} = \sum_{i=1}^{3} \frac{1}{2} m \frac{\partial n v_i \vec{e}_i \langle u_j^2 \rangle}{\partial r_i} = \frac{3}{2} \vec{\nabla} \cdot (p \vec{v}).
$$

(2.85)

Inserting all terms in (2.81) produces:

$$
\int \frac{1}{2} m w^2 \vec{w} \cdot \vec{\nabla}_r f \, d^3 w = \frac{1}{2} m \vec{\nabla}_r \cdot (n v^2 \vec{v}) + \frac{5}{2} \vec{\nabla}_r \cdot (p \vec{v}) + \vec{\nabla}_r \cdot (\vec{v} \cdot \vec{\Pi}) + \vec{\nabla}_r \cdot \vec{q}.
$$

(2.86)
For the third term of (2.78), we again use the same procedure used for the third term of the momentum equation:

\[
\int \frac{1}{2} w^2 \vec{F} \cdot \vec{\nabla} w f \, d^3 w = \sum_{i=1}^{3} \int \frac{1}{2} w^2 F_i \frac{\partial f}{\partial w_i} \, d^3 w. \tag{2.87}
\]

We again use integration by parts on one of the terms (the others are treated mutatis mutandis):

\[
\int w^2 \frac{1}{2} F_1 \frac{\partial f}{\partial w_1} \, d^3 w = \frac{1}{2} \int w^2 F_1 f \, dw_2 dw_3 \bigg|_{-\infty}^{\infty} - \int \frac{1}{2} f \frac{\partial w^2 F_1}{\partial w_1} \, d^3 w = n \frac{1}{2} \frac{\partial \langle w^2 \rangle F_1}{\partial w_1}. \tag{2.88}
\]

Summing over all three components, and using that \( \vec{F} \) does not depend on \( \vec{v} \), we obtain:

\[
\int \frac{1}{2} w^2 \vec{F} \cdot \vec{\nabla} w f \, d^3 w = -\frac{1}{2} n \vec{\nabla} w \cdot \langle \langle w^2 \rangle \vec{F} \rangle = -n \vec{F} \cdot \vec{\bar{v}}. \tag{2.89}
\]

We can add all terms together to obtain:

\[
\frac{3}{2} \frac{\partial p}{\partial t} + \frac{1}{2} m \frac{\partial n v^2}{\partial t} + \frac{1}{2} m \vec{\nabla} r \cdot (n v^2 \vec{v}) + \frac{5}{2} \vec{\nabla} r \cdot (p \vec{v}) + \vec{\nabla} r \cdot (\vec{\bar{v}} \cdot \vec{\Pi}) + \vec{\bar{v}} \cdot \vec{\bar{q}} - \quad \tag{2.90}
\]

\[
\frac{3}{2} \vec{\nabla} r \cdot \vec{\bar{v}} = \int \frac{1}{2} m w^2 \left( \frac{\partial f}{\partial t} \right) c \, d^3 w.
\]

This form is called the extrinsic form. We can write this equation in a slightly more convenient form by subtracting the equation of the zeroth moment, multiplied by \( \frac{1}{2} m n v^2 \):

\[
\frac{3}{2} \frac{\partial p}{\partial t} + \frac{1}{2} m \frac{\partial n v^2}{\partial t} - \frac{1}{2} m v^2 \frac{\partial n}{\partial t} + \vec{\nabla} r \cdot (\vec{\bar{v}} \cdot \vec{\Pi}) \quad \tag{2.91}
\]

\[
\frac{1}{2} m \vec{\nabla} r \cdot (n v^2 \vec{v}) - \frac{1}{2} m v^2 \vec{\nabla} r \cdot (n \vec{v}) + \frac{5}{2} \vec{\nabla} r \cdot (p \vec{v}) + \vec{\nabla} r \cdot \vec{\bar{q}} - \quad \tag{2.90}
\]

\[
\frac{1}{2} m v^2 \vec{\nabla} r \cdot \vec{\bar{v}} = \int \frac{1}{2} m w^2 \left( \frac{\partial f}{\partial t} \right) c \, d^3 w - \frac{1}{2} m v^2 \int \left( \frac{\partial f}{\partial t} \right) c \, d^3 w.
\]

We can simplify this by manipulating the second and fifth term. In both cases, we use the product rule, to obtain:

\[
\frac{3}{2} \frac{\partial p}{\partial t} + \frac{1}{2} m n \frac{\partial v^2}{\partial t} + \vec{\nabla} r \cdot (\vec{\bar{v}} \cdot \vec{\Pi}) + \frac{1}{2} m n \vec{\bar{v}} \cdot \vec{\nabla} r v^2 + \frac{5}{2} \vec{\nabla} r \cdot (p \vec{v}) + \quad \tag{2.92}
\]

\[
\vec{\nabla} r \cdot \vec{\bar{q}} - \frac{1}{2} m v^2 \left( \frac{\partial n}{\partial t} \right) c = \int \frac{1}{2} m w^2 \left( \frac{\partial f}{\partial t} \right) c \, d^3 w - \frac{1}{2} m v^2 \left( \frac{\partial n}{\partial t} \right) c.
\]
The dual inner product operator : between tensors is defined as:

\[ m \cdot \vec{A} = \sum_{i=1}^m \sum_{j=1}^m A_{ij} \]

This form of the extrinsic energy conservation equation will be used to obtain the intrinsic energy equation.

To obtain the intrinsic form, the inner product of momentum conservation equation and \( m\dot{\vec{v}} \) is subtracted from (2.91):

\[
\begin{align*}
\frac{3}{2} \frac{\partial p}{\partial t} + 1 \cdot \frac{\partial v^2}{\partial t} - nm\vec{v} \cdot \frac{\partial \vec{v}}{\partial t} + \nabla \cdot (\vec{v} \cdot \vec{\Pi}) - \vec{v} \cdot (\nabla \cdot \vec{\Pi}) + \\
\frac{1}{2} mn\vec{v} \cdot \nabla v^2 - nm\vec{v} \cdot ((\vec{v} \cdot \nabla)\vec{v}) + \frac{5}{2} \nabla \cdot (p\vec{v}) - \vec{v} \cdot \nabla p + \nabla \cdot \vec{q} - \\
n\vec{F} \cdot \vec{v} + \vec{V} \cdot n\vec{F} &= \int \frac{1}{2} mw^2 \left( \frac{\partial f}{\partial t} \right) d^3w - \frac{1}{2} mv^2 \left( \frac{\partial n}{\partial t} \right) + \\
&\quad mv^2 \left( \frac{\partial n}{\partial t} \right) - \vec{v} \cdot \int m\vec{w} \left( \frac{\partial f}{\partial t} \right) d^3w.
\end{align*}
\]  

We can use the product rule on the second term and seventh term and use obvious simplifications to obtain (2.94):

\[
\begin{align*}
\frac{3}{2} \frac{\partial p}{\partial t} + \nabla \cdot (\vec{v} \cdot \vec{\Pi}) - \vec{v} \cdot (\nabla \cdot \vec{\Pi}) + \frac{1}{2} mn\vec{v} \cdot \nabla v^2 - \\
mn\vec{v} \cdot ((\vec{v} \cdot \nabla)\vec{v}) + \frac{3}{2} \nabla \cdot (p\vec{v}) + p\nabla \cdot \vec{v} + \nabla \cdot \vec{q} = \\
\int \frac{1}{2} mw^2 \left( \frac{\partial f}{\partial t} \right) d^3w + \frac{1}{2} mv^2 \left( \frac{\partial n}{\partial t} \right) - \vec{v} \cdot \int n\vec{w} \left( \frac{\partial f}{\partial t} \right) d^3w.
\end{align*}
\]

The second and third term can be taken together to further simplify (2.94). For this the second term will be rewritten:

\[
\nabla \cdot (\vec{v} \cdot \vec{\Pi}) = \sum_{i=1}^3 \sum_{j=1}^3 \frac{\partial}{\partial r_i} (v_j \Pi_{ij}) = \sum_{i=1}^3 \sum_{j=1}^3 \left( v_j \frac{\partial \Pi_{ij}}{\partial r_i} + \frac{\partial v_j}{\partial r_i} \Pi_{ij} \right) = \\
\vec{v} \cdot (\nabla \cdot \vec{\Pi}) + (\nabla \cdot \vec{v}) : \vec{\Pi}.
\]

Here, \( \Pi_{ij} \) denotes the components of \( \vec{\Pi} \). We will now make a similar simplification in terms four and five of (2.94). We can rewrite the fourth term to obtain:

\[
\begin{align*}
\frac{1}{2} mn\vec{v} \cdot \nabla v^2 &= \frac{1}{2} mn \sum_{i=1}^3 \sum_{j=1}^3 v_i \frac{\partial}{\partial r_i} (v_j^2) = mn \sum_{i=1}^3 \sum_{j=1}^3 v_i v_j \frac{\partial v_j}{\partial r_i} = \\
&\quad nm\vec{v} \cdot ((\vec{v} \cdot \nabla)\vec{v}).
\end{align*}
\]

Thus, the fourth and fifth term of (2.93) cancel. This gives us for the energy conservation

\[(2.95)\text{The dual inner product operator : between tensors is defined as: } A : B = \sum_{i=1}^3 \sum_{j=1}^3 A_{ij}B_{ij} \]
equation:

\[
\frac{3}{2} \frac{\partial p}{\partial t} + \frac{3}{2} \nabla_r \cdot (p \vec{v}) + p \nabla_r \cdot \vec{v} + (\nabla_r \vec{v}) : \mathbf{I} + \nabla_r \cdot \vec{q} = \left( \frac{\partial f}{\partial t} \right)_c \quad (2.97)
\]

\[
\int \frac{1}{2} m w^2 \left( \frac{\partial f}{\partial t} \right)_c \; d^3 w + \frac{1}{2} m v^2 \left( \frac{\partial n}{\partial t} \right)_c - \vec{v} \cdot \int m \vec{w} \left( \frac{\partial f}{\partial t} \right)_c \; d^3 w.
\]

This equation is a valid, albeit unusual, statement of the intrinsic energy conservation equation. If a Maxwell-Boltzmann distribution of the thermal velocities is assumed, a more familiar form can be obtained using the ideal gas law

\[
p = n k_B T \quad (2.98)
\]

to rewrite the pressures to temperatures:

\[
\frac{3}{2} \frac{\partial n k_B T}{\partial t} + \frac{3}{2} \nabla_r \cdot (n k_B T \vec{v}) + p \nabla_r \cdot \vec{v} + (\nabla_r \vec{v}) : \mathbf{I} + \nabla_r \cdot \vec{q} = \left( \frac{\partial f}{\partial t} \right)_c \quad (2.99)
\]

\[
\int \frac{1}{2} m w^2 \left( \frac{\partial f}{\partial t} \right)_c \; d^3 w + \frac{1}{2} m v^2 \left( \frac{\partial n}{\partial t} \right)_c - \vec{v} \cdot \int m \vec{w} \left( \frac{\partial f}{\partial t} \right)_c \; d^3 w.
\]

The first three terms on the right hand side represent the energy gained by collisions. They can be given more significance by applying that \( \vec{w} = \vec{v} + \vec{u} \):

\[
\left( \frac{\partial f}{\partial t} \right)_c \quad (2.100)
\]

\[
\int \frac{1}{2} m w^2 \left( \frac{\partial f}{\partial t} \right)_c \; d^3 w + \frac{1}{2} m v^2 \left( \frac{\partial n}{\partial t} \right)_c - \vec{v} \cdot \int n \vec{w} \left( \frac{\partial f}{\partial t} \right)_c \; d^3 w = \int E_T \left( \frac{\partial f}{\partial t} \right)_c \; d^3 w.
\]

with \( E_T \) the thermal energy. Here, we have used the fact that the bulk velocity does not depend on \( \vec{w} \). Physically, this indicates that the thermal energy gain of the system is equal to the thermal energy a particle possesses when it is created; this is intuitively correct. Combining (2.99) and (2.100), we obtain:

\[
\frac{3}{2} \frac{\partial n k_B T}{\partial t} + \frac{3}{2} \nabla_r \cdot (n k_B T \vec{v}) + p \nabla_r \cdot \vec{v} + (\nabla_r \vec{v}) : \mathbf{I} + \nabla_r \cdot \vec{q} = \int E_T \left( \frac{\partial f}{\partial t} \right)_c \; d^3 w \quad (2.101)
\]

### 2.A.6 Closing the system

In order to be able to solve this set of equations, we need to close it. This cannot be done by adding more moments — each moment has a term that has a structure which
belongs to a moment which is one higher. Thus, we need a formula that can describe the these higher order terms in terms of lower moments. These formulas are based on experimental experience, and often only valid for a certain range of systems.

In order to close the system, we look at (2.99). The fourth and fifth term depend on the velocity to the third power, which is one higher than what we can solve using the second equation. For both of these terms, we will use an an experimental relation to reduce the order of the velocity that is in it.

2.A.6.1 Viscous forces

The fourth term of (2.99) represents the viscous forces. By assuming a Newtonian fluid, we can rewrite the viscosity term using [9]:

\[ \Pi_{ij} = \eta \frac{\partial u_i}{\partial y}. \]  

Here \( \eta \) is the dynamic viscosity. Using this, the order of the velocity is reduced by one. Thus, this term no longer precludes solving the system.

2.A.6.2 Thermal conductivity

For the fifth term of (2.99), we use Fourier’s law of heat conductivity [11]:

\[ \vec{q} = -\lambda \vec{\nabla} T. \]  

Because the temperature depends on the square of the thermal velocity, this equation reduces the highest order of the velocity again to two, eliminating the problem that this term posed in obtaining a solution for the system.

2.A.7 Assumptions

During the derivation of the various conservation equations, we have attempted to make as few assumptions as possible. The most critical assumptions will be discussed below.

2.A.7.1 Fluid assumption

The fluid assumption is formulated in section 2.A.1. It basically states that the system is so large, that it can be split into a large number of volumes, in which there are so many particles that we can have meaningful statistics. This is a rather vague statement. We can quantify this somewhat by considering the typical densities and gradient lengths of the plasmas in this work. In the spark gap plasma of Chapter 9 the typical density is
10^{25} \text{m}^3$, while the typical length scales over which significant differences in the plasma composition occur are of the order of $10^{-6} \text{m}$. Hence, there are $10^7$ particles in a cube with a size equal to this length scale, which is ample for meaningful statistics. For the other systems in this work, the larger geometrical size makes this number even larger.

2.A.7.2 Isotropy of the thermal velocity

This assumption is used in the definition of the pressure. If we cannot assume isotropy of the thermal velocity, the resulting equations become more complicated. Note however, that a drift velocity is not subject to this restriction, as it is not a part of the random thermal motion.

Bibliography


Multi-fluid modeling of transient plasmas: the method
A modular cross section implementation

Abstract—Interactions between particles can be described using the concept of cross sections. Accurate modeling of plasmas requires these cross sections to be supplied to the code. In this Chapter, we present two different methods that are used to supply cross sections from literature to the code. In the first, older method, the user fits analytical relations to the literature data, and supplies these analytical relations as plugins to the code. In the second method, newer method, plugins read look-up tables that are supplied to the code. The methods are compared, and advantages of each method are discussed.

3.1 Introduction

In Section 1, we have discussed the advantages of using a general, modular framework for modeling plasmas. In summary, using a modeling platform that consists of interchangeable modules with a well-defined input and output allows for code reuse and better code validation. In this Chapter, we will illustrate this principle by discussing two different ways of implementing elastic cross sections in one of the grand models used in this work, PLASIMO [1]. In Section 3.2, the problem will be briefly sketched. In Section 3.3, the technique of using plug-ins will be briefly discussed. In Section 3.4, a method of implementing cross sections using analytical functions is discussed. In Section 3.5, a general method of implementing cross sections using look-up tables is discussed. In Section 3.6, conclusions will be drawn.

3.2 Elastic cross sections in plasma physics

In plasmas, collisional interactions between species can be quantified using the concept of cross sections $\sigma$, which are generally a function of the thermal velocity difference be-
etween the species, which can be expressed in terms of an interaction energy \( E \). Computing the transport properties of a plasma, such as the thermal and electrical conductivity, heat transfer, and diffusion coefficients, generally involves computing the collision frequencies between the relevant particle pairs.

Generally, the thermal velocity of both species determines the collision frequency. In our multi-fluid approach, two distinctly dissimilar situations might arise: electrons and heavy particles might collide among themselves, or electrons might collide with heavy particles. In the latter cases, the thermal velocity of the electrons is so much higher that the heavy particles are essentially motionless. In the latter case, one might correct for the fact that both particles have a finite velocity by using a reduced thermal velocity, in which the particle mass \( m \) is replaced by a reduced mass:

\[
m = \frac{m_1 m_2}{m_1 + m_2}
\]

(3.1)

with \( m_1 \) and \( m_2 \) the masses of the colliding particles. We will now confine ourselves to the case in which electrons are the leading particles.

By integrating the energy-dependent cross section, multiplied with the velocity \( u \), over the energy distribution function \( f_T(E) \), a rate coefficient \( k(T) \) can be obtained:

\[
k(T) = \langle \sigma(E)u \rangle_T = \int_0^\infty \sigma(E)f_T(E)u dE
\]

(3.2)

with \( E \) an average electron energy, and the subscript \( T \) denoting that \( \langle \sigma(E)u \rangle \) and \( f \) depends on temperature.

While the approach in (3.2) is generally valid, it does involve the computation of an integral for the evaluation of each cross section. It may be convenient to use a description that avoids this repetitive task. We note that the exponential decay of the Maxwell-Boltzmann energy distribution function means that the contributions to (3.2) occur in a fairly small energy range. For many collision types\(^1\), \( \sigma(E) \) only depends only weakly on the energy. Hence, it is possible to split

\[
\langle \sigma(E)u \rangle_T \approx \langle \sigma(E) \rangle_T \langle u \rangle_T.
\]

(3.3)

In this case, it is possible to define an averaged cross section \( \sigma_{av}(T) \) as:

\[
\langle \sigma(E) \rangle_T = \sigma_{av}(T) = \int_0^\infty 2 \sqrt{\frac{E}{\pi (k_B T)^3}} \exp \left( \frac{-E}{k_B T} \right) \sigma(E) dE,
\]

(3.4)

\(^1\)A notable exception to this is are electron-noble gas collisions, where the Ramsauer-Townsend effect causes the cross section to vary by orders of magnitude in the typical energy range of electrons that are present in the plasma [2].
while the average thermal velocity $\langle u \rangle_T$ is given by

$$\langle u \rangle_T = \sqrt{\frac{8k_B T}{\pi m}}.$$  \hspace{1cm} (3.5)

It is noted that the accuracy of cross sections is typically poor, often having errors of tens of percents. The errors induced by 3.3 are typically not larger than this. Hence, 3.3 can be a useful approximation in practice. For cases in which this approximation causes unacceptable errors, or for transport coefficients that do not explicitly include a $k(T)$, the ordinary cross section $\sigma(E)$ should still be provided. A very important example of the latter case are the Frost mixture rules [3, 4], that are used for the modeling of the electron thermal and electrical conductivity.

Hence, PLASIMO requires both values of $\sigma(E)$ and $\sigma_{av}(T)$. For a non-trivial chemistry, there are many different possible interactions between particles. Fortunately, the interaction between charged particles can be described well using a Coulomb cross section [5], reducing the amount of cross sections that need to supplied considerably.\(^2\) Otherwise, it is necessary to consult literature, which may provide $\sigma(E)$ (and sometimes $\sigma_{av}(T)$), obtained from analytical approximations, theoretical computations, measurements, or a combination of these methods. Hence, including any new species involves a literature study of the cross section, and implementing the obtained cross sections in the code.

In this Chapter, we will discuss two different methods to implement cross sections in PLASIMO. Both make use of plugins to extend the core code. In the first method, analytical functions are used to approximate both cross sections, while the second method, look-up tables are used. Both methods are designed to avoid evaluations of (3.4) when possible to speed up the calculation. It is noted that it is possible for the user to use either method for any cross section, in any combination.

### 3.3 Plugins

A plugin can be defined as a piece of software that can be added to a larger piece of software, extending its functionality. These plugins only interact with the core code via well-defined interfaces. These interfaces basically defines what is implemented, be it a boundary condition, a matrix solver, or, in this case, $\sigma(E)$ or $\sigma_{av}(T)$. Hence, implementing cross sections requires at least two plugins, one for $\sigma(E)$ and one for $\sigma_{av}(T)$.

How it is implemented, however, is determined by the plugin. For instance, in the case of boundary conditions, depending on this implementation, a boundary condition

\(^2\)It is noted that PLASIMO is capable of automatically computing non-Coulomb cross sections as well. However, the results are often very poor, especially for relatively simple species, where quantum-mechanical effects cause large deviations from the classical hard-sphere and Langevin models that are used [1].
might be a Neumann boundary condition, a Dirichlet boundary condition, or something else, such as the multi-region wall boundary condition discussed in Section 2.5. It is possible to add as many plugins as desired without any modification of the core code. This allows for extendability. Furthermore, if an error is made in a new implementation, it is now contained to the plugin, making debugging far easier.

PLASIMO contains facilities to make many different aspects of a computation pluggable, including cross sections. Hence, both cross section implementations described in this Chapter are implemented as plugins. For further details of the use of cross sections in PLASIMO, we refer to [6, 7].

### 3.4 Analytical functions

Generally, $\sigma(E)$ and $\sigma_{av}(T)$ are smooth functions of $E$ and $T$, respectively. This makes an analytical function an obvious choice to implement them.

$\sigma(E)$ is sometimes already provided as an analytical function in literature, which means that implementing it in a plugin is trivial. However, $\sigma(E)$ may also be presented as a look-up table, and has to be fitted with an analytical function before it can be implemented in a plugin. While this is not difficult, it is not something that can be automated, as the analytical functions that can approximate the cross section are different in form for each cross section.

A function that gives $\sigma_{av}(T)$ can be derived by evaluating (3.4) for various values of $T$, and fitting a smooth, analytical curve through the data points thus obtained. This analytical curve is then implemented as a plugin. The resulting plugin structure is shown in Figure 3.2.

The most important disadvantage of this method is the duplicate user effort that is required for each cross section: It is necessary to carry out a fairly large number of integrations, and the curve fit is something that requires considerable user involvement, as each cross section has a different shape and hence needs a different function. Moreover, this procedure is very error-prone, as no standard, well-tested method can be used to generate a cross section. Another disadvantage is the fact that for a realistic interaction database, a vast number of cross sections are needed: two for each interaction. This is undesirable from a maintenance point of view, although the fact that these cross section implementations are plugins and are not part of the core code makes this problem less acute. This has prompted us to use an even more general method to implement cross sections.
3.5 Look-up tables

Rather than using an analytical approximation, it is possible to use a look-up table containing the cross section for various energies [8]. For common computer memory sizes, look-up tables of several thousands of entries are quite feasible. This is likely to result in an accuracy that is superior to that of an analytical function. Obtaining a look-up table from an analytical function is trivial, unlike the reverse procedure, as discussed in Section 3.4. The functionality of importing a look-up table and extracting a cross section is implemented using a plugin. This makes it possible to import $\sigma(E)$ into the core code.

Obtaining $\sigma_{av}(T)$ from $\sigma(E)$ using (3.4) is computationally expensive, and needs to be carried out for each transport coefficient that requires $\sigma_{av}(T)$, for each interaction, at each grid cell, and at each iteration, making it a significant if not dominant part of the computational cost of the entire computation. This cost can be reduced by using the Gauss-Laguerre integration algorithm, which requires that the integrand can be written as the product of a function and a decaying exponent, as is the case in (3.4).

An even larger reduction in computational cost can be achieved by evaluating (3.4) for various values of $T$ before the main computation starts, and storing the results in a look-up table of $\sigma_{av}(T)$. The size of this look-up table is specified by core code in the initialization. Computation of several thousand entries is usually more than sufficient. This amount is much smaller the total amount of cross section evaluations, which, for a
The cross section $\sigma(E)$ is read from a look-up table. This look-up table is also read by the averaged cross section plugin, integrated using (3.4), and stored in an internal look-up table, with a size that is determined by the core code. The averaged cross section $\sigma_{av}(T)$ are read from internal look-up tables that are generated at startup (adapted from [8]).

![Diagram](image)

**Figure 3.2:** The new plugin structure of the look-up table cross section implementation in PLASIMO. The cross section $\sigma(E)$ is read from a look-up table. This look-up table is also read by the averaged cross section plugin, integrated using (3.4), and stored in an internal look-up table, with a size that is determined by the core code. The averaged cross section $\sigma_{av}(T)$ are read from internal look-up tables that are generated at startup (adapted from [8]).

The complete plugin structure is depicted in Figure 3.2. A single look-up table is read by both the $\sigma(E)$ and the $\sigma_{av}(T)$ plugin. $\sigma_{av}(T)$ evaluates (3.4), creating an internal look-up table.

Using the look-up table method has additional advantages apart from computation speed and accuracy. Firstly, this method is general enough to handle any cross section. Rather than two plugins for each cross section, just two plugins are needed: one for all $\sigma(E)$ and one for all $\sigma_{av}(T)$. This leads to a vast reduction in the amount of code needed, inherently making the code less likely to contain bugs: code that does not exist cannot contain bugs, and the code that is used is run more often and hence tested more often. Secondly, it is much more user-friendly: instead of burdening the user with the repetitive and error-prone task of evaluating (3.4) and the subsequent curve-fitting, the computer performs this task. Thirdly, it can be used as a step toward further abstraction: while this example is restricted to the use of elastic cross sections, the concept can be used for inelastic processes and reaction rates as well.
3.6 Conclusions

It is noted that this implementation effectively eliminates \( \sigma_{\text{av}}(T) \) as an input parameter of the model from the user’s point of view, as the model now only requires \( \sigma(E) \). Should it be desired that the use of \( \sigma_{\text{av}}(T) \) to approximate cross sections is obsoleted, perhaps as a result of ever-increasing computer power, then this implementation already provides the correct interface awarded the user.

3.6 Conclusions

In this Chapter, we have shown two different methods of using a general formalism to implement elastic cross sections. In the first (old) method, analytical functions, that are derived by the user, are implemented as plugins. This method has the disadvantage that a considerable amount of effort is needed for each cross section. An even further generalization is obtained in the second (new) method, where the cross sections are provided as look-up tables. By using a reusable cross section plugin, coding needs to be done only once to be able to handle all cross sections, greatly reducing the effort needed to add new cross sections to PLASIMO and making the code less error-prone. This is a good example of the design philosophy behind the coding methods used in this thesis, and serves to illustrate the general framework that is used for the modeling of the very different systems in this work.

Acknowledgments

J.G. Keizer is gratefully acknowledged for his work on the implementation of the look-up table cross sections. J. van Dijk and M. L. Bekx are acknowledged for the fruitful discussions.

Bibliography


Abstract—Disturbed Bilateral Relations offer a way of categorizing plasma processes by their deviation from equilibrium. This method can be used to create a simple plasma model that solves key plasma parameters, namely the electron temperature, electron density and heavy particle temperature. An implementation of this method is presented, and the results validated against a detailed plasma simulation, for a wide range of parameters.

4.1 Introduction

Plasma modeling is an indispensable part of plasma physics research. It is a very broad field that has a very wide variety of different models, each appropriate for different plasmas and with their own drawbacks and strengths. This wide varsity is of course related to the breadth of plasma physics research itself. A useful distinction in modeling is the distinction between global models and grand models.

Global models are models that rely on a high number of assumptions to simplify a problem as much as is feasible, and then solve the simplified problem. They are usually zero or one-dimensional, can be analytical or numerical, and if they are numerical, the numerical cost is small. Because the basic physics is not clouded by numerical details, the successful use of global models for plasma simulation can give a good understanding of the fundamental physics involved in a problem.

This Chapter is based on B. H. P. Broks, J. J. A. M. van der Mullen, Accepted for publication in Journal of Physics: Conference Edition (2006)
Creating a global plasma model using disturbed Bilateral Relations

With the advent of cheap and powerful computers, grand models have become a popular method of describing plasmas. Using this method, it is possible to create a computer model that, given only elemental input such as reaction and collision cross sections, and external plasma parameters such as geometry, power incoupling and gas feeds, gives an accurate description of the plasma.

In this Chapter, we will discuss the creation of a global model that is based on the principle of disturbed Bilateral Relations (dBR). Briefly, this approach categorized various processes in the plasma based on whether they are in equilibrium or not. This powerful and flexible approach will be used to construct a model that is applicable over a wide parameter range. This allows one to use a single, simple model to browse a large part of the parameter space over which plasmas can exist. We will compare the results of a parameter study with this model with the results obtained with a validated grand model.

The method of disturbed Bilateral Relations is not new, having been described, having been introduced by Van der Mullen in 1990 [1], and has been used in the analysis of plasmas [2–4]. Quantitative numerical plasma modeling using disturbed Bilateral Relations was pioneered by the author of this Thesis [5].

4.2 Disturbed Bilateral Relations

In every laboratory or industrial plasma, a degree of nonequilibrium exists. A nonequilibrium is important from an application point of view, as a net production of a plasma product, be it light, heat, or chemically active species, is desired, and this cannot happen in perfect equilibrium. In an equilibrium situation between one state \( \alpha \) and a second state \( \beta \), in which there is a forward and backward process which are inverse processes and are in equilibrium, one can write

\[
\alpha \nu_f = \beta \nu_b, \tag{4.1}
\]

with \( n \) the densities, \( \nu_f \) the forward rate and \( \nu_b \) the backward rate. This balance is schematically represented in Fig.4.1(a).

Such an equilibrium can be disturbed by a transport term \( \beta \nu_t \), which without loss of generality can be considered as a loss term for state \( \beta \). This modifies Eq. (4.1) to

\[
\alpha \nu_f - \beta \nu_b = \beta \nu_t. \tag{4.2}
\]

In (4.2), we will refer to the second term as the proper term, while the third term is an improper term. This balance is schematically represented in Fig. 4.1(b). Because industrial and laboratory plasmas are designed to get something, such as radiation or excited species, out of them, this improper term might be quite large for many processes.
4.3 The model

In this section, we will describe a global model based on the principle of disturbed bilateral relations. We will, for the moment, restrict our discussion to a quasi-neutral atomic plasma, with a modest (<1%) degree of ionization. These assumptions are not fundamental, but will allow us to make the discussion more concrete.

In order to obtain the key plasma properties, three global balance equations are solved, namely the electron particle balance, the electron energy balance and the heavy particle energy balance. These give three key plasma properties, namely the electron temperature $T_e$, the electron density $n_e$, and the heavy particle temperature $T_h$, respectively.

The model is most accurate for plasmas with a low degree of ionization. For plasmas with a high degree of ionization, much simpler models that are based on a Local Thermal Equilibrium can be made.

4.3.1 The electron particle balance

In the plasmas under consideration, electron impact ionization is the most important source of ionization. In this process, a high-energy electron impacts on an atom, ionizing it and releasing a second electron. The proper inverse process for this is two-electron recombination, in which two electrons and an ion recombine to form an ion and an elec-
tron. An equilibrium between these processes is known as Saha equilibrium. We will, however, include a second, improper loss term for the electrons, namely ambipolar diffusion to the walls. This yields the following equation for the electron particle balance:

\[ n_e n_1 k_{\text{ion}} - n_e^2 n_+ k_{\text{rec}} = \frac{D_{\text{amb}} n_e}{\Lambda^2}. \]  

(4.3)

Here, \( n_1 \) is the ground state density, \( k_{\text{ion}} \) is the ionization rate, \( k_{\text{rec}} \) is the recombination rate, \( D_{\text{amb}} \) is the ambipolar diffusion coefficient and \( \Lambda \) is the typical length scale of the smallest dimension. The term on the left hand side of the equation represents the energy source from elastic transfer between electrons and heavy particles, while the second term on the right hand side represents the energy source caused by inelastic processes.

(4.3) can be rewritten to:

\[ k_{\text{ion}} = \frac{D_{\text{amb}}}{n_1 \Lambda^2} + \frac{n_e n_+}{n_1} k_{\text{rec}}. \]  

(4.4)

Because \( k_{\text{ion}} \) is strongly dependent on \( T_e \), the particle balance in fact gives us an equation for \( T_e \).

### 4.3.2 The electron energy balance

The electrons in a plasma undergo elastic collisions with the neutrals in the plasma. Due to the high mass ratio between electrons and ions, the energy transfer between them is quite inefficient, causing two different temperatures to exist, one for the electrons and one for all the heavy particles.

The electron energy balance has the following form:

\[ n_e n_\sigma k_{\text{heat}} (k_B T_e - k_B T_h) = \epsilon - (n_e n_\sigma k_{\text{ion}} - n_e^2 n_+ k_{\text{rec}}) E_{\text{ion}}, \]  

(4.5)

where \( n_\sigma \) is the heavy particle density, \( k_{\text{heat}} \) is the heat transfer coefficient, \( \epsilon \) the dissipated power and \( E_{\text{ion}} \) the ionization energy.

All terms in (4.5), except \( \epsilon \), are proportional to \( n_e \). This means, that (4.5) can be used to find the density of \( n_e \), when the coefficients and \( \epsilon \) are known.

### 4.3.3 The heavy particle energy balance

The heavy particles are heated by elastic collisions with the electrons, and lose their heat via heat conduction to the wall:

\[ n_e n_\sigma k_{\text{heat}} (k_B T_e - k_B T_h) = \lambda_h \frac{T_h - T_{\text{wall}}}{\Lambda^2}, \]  

(4.6)

with \( k_{\text{cond}} \) the coefficient of heat conduction and \( T_{\text{wall}} \) the wall temperature. Eq. (4.6) can be used to obtain a value for \( T_h \).
4.3.4 The coefficients

The three balance Equations (4.4), (4.5) and (4.6) give a values for \( T_e, n_e \) and \( T_i \), provided that the coefficients \( k_{\text{ion}}, k_{\text{rec}}, D_{\text{amb}}, k_{\text{heat}} \) and \( k_{\text{cond}} \) are known. These coefficients depend on \( T_e \) and \( T_i \), and on the physical properties gas in which the plasma exists.

The ionization rate coefficient \( k_{\text{ion}} \) can be approximated by an Arrhenius rate. This rate has the following general form:

\[
k_{\text{ion}} = k_{\text{rate}} T_e^q \exp \left( \frac{-I}{k_B T_e} \right) \tag{4.7}
\]

The factor \( q \), the energy threshold for the reaction \( I \), and the constant \( k_{\text{rate}} \) depend on the gas [6].

The most efficient ionization process in a plasma is not necessarily direct ionization, as stepwise ionization may be a more important mechanism. The rate that is implemented should reflect this, i.e. if direct ionization is dominant, the \( q, I \) and \( k_{\text{rate}} \) of the direct process should be used, and if stepwise ionization is dominant, the \( q, I \) and \( k_{\text{rate}} \) of excitation to the first excited state should be used. Ref. [2] gives a more thorough explanation of this.

From (4.4), it follows that a value of \( k_{\text{ion}} \) is necessary to compensate for the losses. Because \( k_{\text{ion}} \) depends on \( T_e \), this gives the value for \( T_e \). The recombination rate coefficient \( k_{\text{rec}} \) can be obtained using the principle of detailed balancing [6] from \( k_{\text{ion}} \):

\[
k_{\text{rec}} = \left( \frac{h}{\sqrt{2\pi m_e k_B T_e}} \right)^3 \frac{k_{\text{rate}} T_e^q}{2G} \exp \left( \frac{E_{\text{ion}} - I}{k_B T_e} \right), \tag{4.8}
\]

where \( G \) is the ratio of the degeneracy of the ion state and the ground state. The factor 2 in the denominator factors in the degeneracy of the electron.

The ambipolar diffusion coefficient \( D_{\text{amb}} \) consists of the ion diffusion coefficient multiplied with a factor to take into account the ambipolar field, and is given by [7]:

\[
D_{\text{amb}} = \left( 1 + \frac{T_e}{T_i} \right) \frac{2}{3n_a \sigma_{ia}} \sqrt{\frac{k_B T_i}{\pi M}} \tag{4.9}
\]

where \( \sigma_{ia} \) is the \( T_i \)-dependent ion-atom collision cross section and \( M \) is the ion mass. The heat transfer coefficient \( k_{\text{heat}} \) is the product of the reduced collision frequency (i.e. the collision frequency divided by \( n_a \) and \( n_e \)) and the heat transferred in one such collision, divided by the temperature difference \( T_e - T_i \), which is explicitly accounted for in Equations (4.5) and (4.6). This gives the following expression for \( k_{\text{heat}} \) [7]:

\[
n_e n_p \sigma_{ea} \sqrt{\frac{8k_B T_e}{\pi m_e} \frac{3m_e}{M}} (k_B T_e - k_B T_i) = n_e n_p (k_B T_e - k_B T_i) k_{\text{heat}}, \tag{4.10}
\]
Figure 4.2: A schematic flow chart of the algorithm used to solve the equations that form the model. Basically, a two-stage approach is used: First, $T_e$ is solved, and this typically rather accurate estimate is used in the second stage, which is the full model.

where $m_e$ is the electron mass and $\sigma_{ea}$ the electron-atom momentum transfer cross section. We can solve $k_{\text{heat}}$ from (4.10) to obtain [6]

$$k_{\text{heat}} = \frac{3\sigma_{ea}}{M} \sqrt{\frac{8k_B T_e m_e}{\pi}}. \quad (4.11)$$

The thermal conductivity coefficient $\lambda_h$ for the heavy particles is given by [7]

$$\lambda_h = \frac{\sqrt{2k_B}}{\sigma_{aa}} \sqrt{\frac{8k_B T_h}{\pi M}}, \quad (4.12)$$

with $\sigma_{aa}$ the neutral-neutral collision cross section, which generally depends on $T_h$.

### 4.4 The algorithm

The three balance equations and the five equations for the coefficients in those equations are in principle sufficient to solve for the eight variables in the system. This is not trivial, especially because of the highly nonlinear nature of many of the equation. In this subsection, an iterative algorithm that can reliably solve the equations will be presented. In this algorithm, we use the fact that the ionization degree is low to approximate $n_1$ with $n_a$. The solution procedure for the equations is shown in Figure 4.2.

The first part of the algorithm consists of obtaining a good initial for $T_e$, that is to be used in the main iteration. For this, (4.4) and the coefficients therein are solved iteratively.

The iteration is started by choosing starting values, denoted with the superscript $^i$, which should be reasonably close to an actual plasma condition. This was found to be not critical, and the hard-coded default values of $T_e^i = 10000$ K, $T_h^i = 1000$ K, $n_e = 10^{12}$ m$^{-3}$.
performed well for all simulations in this work. Next, the right-hand terms of (4.4) are calculated using (4.9) and (4.8) and substituted. This gives a value for $k_{ion}$. $T_e$ cannot be solved directly from (4.7), as (4.7) is a transcendental equation. It is solved by substituting $T_{e1}$ in the $T_{Te}$-term, and solving for the $T_e$ in the exponential term.

This new value of $T_e$, which we will call $T_{e1}$, can now be used to compute new values for the right-hand terms in (4.4), and hence, a new value for $k_{ion}$ is obtained. By substituting $T_{e1}$ in the exponential term, (4.7) can be solved for a new value of $T_e$. This procedure is repeated until convergence is achieved.

With a good estimate for $T_e$, we now turn our attention to $T_h$ and $n_e$. First, $k_{heat}$ is computed using (4.11) and the new value of $T_e$ and $\lambda_h$ is computed using (4.12) and $T_{h1}$. At this point, all coefficients are known. To solve $n_e$, we substitute (4.3) in (4.5):

$$n_e n_a k_{heat}(T_e) - n_e n_a k_{heat}(T_h) = e - \left( \frac{D_{amb} n_e}{\Lambda^2} \right) E_{ion}. \tag{4.13}$$

We can now solve $n_e$ from Eq. (4.13):

$$n_e = \frac{e}{n_a k_{heat} (T_e - T_h) + \left( \frac{D_{amb}}{\Lambda^2} \right) E_{ion}} \tag{4.14}$$

and by substituting the coefficients and $T_e$ and $T_{h1}$, obtain a value for $n_e$.

The last property to be determined is $T_h$. It can be determined by solving (4.6) for $T_h$:

$$T_h = \left( T_e + T_{wall} \frac{\lambda_h}{k_{heat} \Lambda^2} \right) \left( 1 + \frac{\lambda_h}{k_{heat} \Lambda^2} \right)^{-1} \tag{4.15}$$

We now have obtained values for all coefficients and $T_e$, $T_h$ and $n_e$. This makes it possible to start the main iterative loop. In this loop, we first solve Equations (4.4), (4.13) and (4.15) to obtain $T_e$, $n_e$ and $T_h$, respectively. These plasma properties can now be used to obtain new values for the coefficients $k_{rec}$, $D_{amb}$, $k_{heat}$, and $\lambda_h$ using Eqs. (4.8), (4.9), (4.11) and (4.12), respectively. By iterating, the equations can be solved to a degree of precision that is in principle only limited by machine accuracy.

The equations for the transport coefficients presented in this Section are only the simplest realistic approximation of the true transport coefficients of a plasma. It is, of course, possible, and in fact quite easy to use other, more accurate and complicated expressions for the transport coefficients.

### 4.5 Comparison with a grand model

In order to demonstrate the viability of the dBR global model, we will compare the result this approach gives with the results of a grand model. For this, we will use a very
simple case, namely an Ar plasma in a cylindrical vessel with radius $\Lambda$, that is assumed to be infinitely long. By varying $n_a$, $\Lambda$ and $P$, the latter denoting the power per unit length, we can easily browse a large part of the parameters space.

The grand model used is the PLASIMO code. This is a well-validated code, that has been used successfully in the modeling of a wide variety of plasmas [8, 9].

The necessary input data for the model has been obtained from literature. For $\sigma_{aa}$, the hard-sphere cross section is used. For $\sigma_{ea}$, [10] is used, while for $\sigma_{ia}$ [11] is used. The ionization rate is obtained from [2].

We have carried out a parameter study by varying $n_a$, $\Lambda$ and $P$, in both the dBR simulation and the PLASIMO simulation. The default values are $n_a = 10^{24}$ m$^{-3}$, 1 mm and 100 W/m, respectively. The wall of the plasma is cooled to a temperature of 300 K.

Figure 4.3 shows that for increasing $P$, the electron density increases. Furthermore, the increased electron density leads to an increase in electron/heavy particle collision frequency and hence to background gas heating. The match between the dBR and PLASIMO is generally excellent. However, dBR does not take into account the fact that gas heating and ionization redistributes the background gas, and this leads to a discrepancy between the dBR and PLASIMO results.

Figure 4.4 shows that for decreasing $\Lambda$, $n_e$ increases. This is due to the increase in power density. Furthermore, decreasing $\Lambda$ leads to more diffusive losses and hence a higher $T_e$. The match between dBR and PLASIMO is again very good.

Figure 4.5 shows two competing trends for $n_e$ as a function of $n_a$. For low $n_a$, the
4.6 Conclusions

A zero-dimensional plasma model that uses disturbed Bilateral Relations to obtain $n_e$, $T_e$ and $T_h$ for a given set of plasma parameters is presented. In its present form, the model can simulate a wide range of atomic, quasineutral plasmas, and because of the conceptual simplicity and easy implementation of such a method, it is easily expanded to handle other plasmas. Such extensions could include the effect of external flow fields, causing extra transport losses, and the inclusion of a buffer gas, modifying heat and particle transport.

The model has been compared with a well-validated grand plasma model, in this case the PLASIMO code, in a parameter study, to see if the dBR model correctly reproduces the trends. The agreement between both models is excellent, and the results can easily be explained with elementary understanding of plasma physics, which leads to the conclusion that the dBR model produces physically relevant results over a wide parameter range, making it suitable for the global modeling of plasmas.
Figure 4.5: A study of the impact of variation of $n_d$ on $n_e$, $T_e$ and $T_h$, obtained with dBR and PLASIMO.

Bibliography


Chapter 5

A nonlocal-thermal-equilibrium model of a pulsed capillary discharge waveguide

Abstract—Slow pulsed capillary discharges are under investigation for use as plasma channel waveguides in laser-wakefield acceleration. In this study, we present a non-local-thermal equilibrium (non-LTE) plasma model with a model for the wall temperature coupled to it. This model is used to describe an example of a slow pulsed capillary discharge, and the results are compared with experimental results. The agreement is satisfactory, indicating suitability of our model. Significant deviations from LTE are found during the formation of the plasma channel. The model is also used to study the influence of the discharge current on the guiding properties. It was found that this influence is small over most of the current range that was investigated.

5.1 Introduction

Laser-wakefield acceleration is an acceleration technique, that makes it possible to build relatively compact particle accelerators by the virtue of the large acceleration gradients that are created [1]. For this application, it is critical that the laser is focused to a small spot size and kept as a narrow bundle over a length of several cm. This requirement means that the laser must not diffract over many Rayleigh diffraction lengths [2]. This can be achieved by channeling the laser beam through a suitably formed plasma. For a Gaussian beam, the channeling is optimal when the electron density of the plasma has a hollow parabolic shape, with the lowest density in the center. In this arrangement,
the refractive index of the plasma provides a guiding that is similar to that in gradient index fiberoptics [3].

It is also important that the plasma is fully ionized, meaning that every atom is stripped of all its electrons, in the parts where it will interact with the laser. Not fully ionized particles will be further ionized by the laser, consuming laser power and disturbing the electron density profile.

One of the methods used for creating a suitable plasma is the capillary discharge. There are several types of these discharges in use.

Discharge-ablated capillaries operate by ablating and subsequently ionizing wall material [4]. This obviously limits the lifetime of the capillary. Furthermore, the relatively low temperature causes less than full ionization.

An alternative is the fast gas-filled capillary discharge. The gas used should be easy to fully ionize, such as hydrogen. A current pulse, typically several tens of kA during tens of nanoseconds, creates a pinch plasma. This current produces a plasma that is suitable as a waveguide [5–7]. However, the azimuthal magnetic field not only produces the pinch, but also makes the channel susceptible to magneto-hydrodynamic (MHD) instabilities [3].

Using a lower current for a longer time reduces the MHD instabilities significantly. A working waveguide based on this principle, called a slow capillary discharge, has been demonstrated [8]. These discharges have a diameter of a few hundred microns, are filled with several kPa of hydrogen, and are ionized by a current pulse of a few hundred A lasting a few hundred ns. They are wall-stabilized. The typical current densities of $10^9$ A m$^{-2}$ are much more than conventional steady-state arcs, which operate at about $10^6$ A m$^{-2}$, but much less than pinch plasmas, where the current density can reach values of $10^{11}$ A m$^{-2}$.

In this study, a model will be presented that is valid for these slow capillary discharges. This model will be used to simulate an actual waveguide, and the simulated results will be compared with experimental results. The physics behind the formation of the channel and the suitability for guiding of the found electron density profile will be discussed. Our model will be compared with an earlier model given in Bobrova et al. [9] where the same device is modeled. Our model differs on three key points, namely in the treatment of the wall, the treatment of the nonequilibrium aspects and the composition calculation. It will be shown that nonequilibrium aspects have an important impact on the formation of the hollow electron density profile. The model is then used to investigate the influence of the discharge current on the plasma guiding properties.
5.2 The physical model

Based on the type of discharge, which is basically a pulsed wall-stabilized high-current hydrogen arc discharge, we will create a physical model to describe the system, and justify the assumptions used to arrive at this model.

The system consists of two regions, the discharge plasma and the wall. They are described by different models, coupled by the boundary conditions.

5.2.1 The model of the plasma

The basis of the plasma model is formed by a two-temperature non-LTE quasineutral fluid model, which is used extensively in the description of low-temperature plasmas and has a wide range of validity. This model is described by the following equations:

- The bulk continuity equation,
  \[
  \frac{\partial \rho}{\partial t} + \vec{\nabla} \cdot (\rho \vec{v}) = 0. \tag{5.1}
  \]
  Here, \( \rho \) is the bulk density, \( t \) is the time and \( \vec{v} \) is the bulk velocity.

- The bulk momentum conservation equation,
  \[
  \frac{\partial \rho \vec{v}}{\partial t} + \vec{\nabla} \cdot (\rho \vec{v} \vec{v}) = -\vec{\nabla} p + \vec{F}_{\text{Lor}} - \vec{\nabla} \cdot (\mu \vec{v} \vec{v}). \tag{5.2}
  \]
  Here, \( p \) is the bulk pressure, \( \vec{F}_{\text{Lor}} \) is the Lorentz force, and \( \mu \) is the viscosity.

- The species conservation equation,
  \[
  \frac{\partial n_i}{\partial t} + \vec{\nabla} \cdot (n_i \vec{v}) = \sum_j S_j R_{ji}. \tag{5.3}
  \]
  Here, \( n_i \) is the density of species \( i \), \( S_j \) is the reaction rate for reaction \( j \) and \( R_{ji} \) is the stoichiometric constant of the production of particle \( i \) by reaction \( j \).

- The electron energy equation,
  \[
  \frac{\partial C_e T_e}{\partial t} + \vec{\nabla} \cdot [(C_e T_e + p_e) \vec{v}] - \vec{\nabla} \cdot \vec{p}_e - \vec{\nabla} \cdot (\lambda_e \vec{\nabla} T_e) =
  \]
  \[
  -k_{\text{heat}}(T_e - T_h) + \frac{J^2}{\sigma} - \sum_j \Delta E_j S_j. \tag{5.4}
  \]
  Here, \( C_e \) is the electron heat capacity, \( T_e \) is the electron temperature, \( p_e \) is the electron pressure, \( \lambda_e \) is the electron heat conductivity, \( k_{\text{heat}} \) is elastic electron-heavy particle heat transfer rate, \( T_h \) is the heavy particle temperature, \( J \) is the current density, \( \sigma \) is the conductivity, and \( \Delta E_j \) is the reaction energy for reaction \( j \).
The heavy particle energy equation,

$$\frac{\partial C_h T_h}{\partial t} + \nabla \cdot [(C_h T_h + p_h) \vec{v}] - \nabla \cdot \nabla p_h - \nabla \cdot (\lambda_h \nabla T_h) + \bar{\tau} : \nabla \vec{v} = k_{\text{heat}}(T_e - T_h).$$

(5.5)

Here, $C_h$ is the heavy particle heat capacity, $p_h$ is the heavy particle pressure, $\lambda_h$ is the heavy particle heat conductivity, and $\bar{\tau}$ is the viscosity tensor.

Equations (5.1) and (5.2) describe the bulk behavior of the system. From these equations, the velocity field and pressure are computed. The non-LTE chemistry, which can contain as many chemical species and reactions between these species as necessary, is solved with a set of equations (5.3). In this non-LTE approach, it is assumed that all the heavy particles share a common temperature $T_h$ and the electrons have a possibly different temperature $T_e$. We use Eq. (5.5) to compute the former and Eq. (5.4) to compute the latter. Because we assume that all the Ohmic heating is done by the electrons and that all the reactions are due to electron kinetics, these source terms are solely in the electron energy balance and not in the heavy particle balance.

In principle, the capillary discharge is two-dimensional, having a radial symmetry. However, since the length of the capillary is typically two orders of magnitude larger than its radius, we expect that the discharge can be described well by assuming it only depends on the radial coordinate $r$ and the time $t$.

5.2.2 Source terms and transport coefficients

We will now discuss the meaning and validity of these equations, and present the formulas for the transport coefficients and source terms.

Equation (5.1) is the continuity equation, which does not contain source terms or transport coefficients.

Equation (5.2) is a special case of the Navier-Stokes equation with only one bulk force, $\vec{F}_{\text{Lor}}$. This force is caused by the current in the axial $z$ direction generating a magnetic field $B$ in the $\phi$ direction. This results in $\vec{F}_{\text{Lor}}$ (in the negative $r$ direction) of which the magnitude $F_{\text{Lor}}$ is given by

$$F_{\text{Lor}} = JB.$$  (5.6)

The magnetic field is obtained by using Ampère’s law [10]:

$$B = \frac{\int_0^r \int_0^{2\pi} r' dr' d\phi'}{2\pi r},$$  (5.7)

where $r'$ and $\phi'$ are integration variables. The magnetic permeability $\mu$ is assumed to be equal to the magnetic permeability of vacuum $\mu_0 = 4\pi \times 10^7$ F m$^{-1}$. The viscosity in the
last term of Eq. (5.2) is computed based on the description in [11]. However, we correct the Coulomb logarithm $\lambda_C$ for the effects of plasma nonideality as outlined in Ref. [12]:

$$\lambda_C = \ln(0.6 \Lambda_C)$$  \hfill (5.8)

with $\Lambda_C$ the ratio of the Debye length and the impact parameter [11]. We use this corrected Coulomb logarithm throughout this work. The difference between the corrected and uncorrected Coulomb logarithm is at most 30%. In this plasma, the ion Hall parameter is always much smaller than 1, so the magnetization of the ions has a negligible influence on the transport parameters.

In our model, the following four species are used: the heavy particles H$_2$, H, H$^+$ and the electrons e. We note that the actual number of species in the plasma is much larger than the four species we take into account: the plasma also contains among others excited hydrogen molecules and atoms, and hydrogen molecular ions. The small energy distance between these excited states and the ion state, combined with the relatively high electron temperature, causes the destruction of these species to be rapid. The formation on the other hand is slower due to the larger energy gap between the ground state and the excited states. Therefore, their density is low and we do not take them into account explicitly. The influence of molecular ions [13, 14] is negligible due to the high electron density, which leads to rapid dissociative recombination.

To obtain the non-LTE chemical composition, the species conservation equation (5.3) is solved for two species, the electron density is obtained from quasineutrality, and the last species follows from the bulk pressure obtained from Eq. (5.1) and Eq. (5.2) [15].

The right-hand side of Eq. (5.3) represents the sources from the reactions. We take into account three reactions and their inverse reactions.

- Electron impact dissociation,

$$\mathrm{H}_2 + e \rightarrow 2\mathrm{H} + e.$$  \hfill (5.9)

The rate coefficient $k_{\text{ediss}}$ for this reaction is determined using the cross sections in Ref [16], noting that the high electron temperature causes the direct process to be faster than the stepwise process. We found that the rate coefficient between an electron temperature of 1.5 eV and 3.5 eV can be approximated satisfactorily by a modified Arrhenius rate $k$,

$$k = c \left( \frac{T_e}{\text{eV}} \right)^q \exp \left( \frac{-E}{k_B T_e} \right)$$  \hfill (5.10)

with $q=2$, $c=1.41 \times 10^{-15} \text{ m}^3 \text{ s}^{-1}$ and $E$ the reaction energy of 4.5 eV. While the temperature in the plasma will rise beyond this range, the dissociation of H$_2$ is complete at that time, so the poor description of this reaction for these high temperatures is not important. We neglect dissociation by heavy particles, as the heavy
particle temperature does not rise high enough to significantly dissociate hydrogen before virtually all hydrogen is dissociated by electron impact dissociation.

- **Direct electron impact ionization**

\[ \text{H} + \text{e} \rightarrow \text{H}^+ + 2\text{e}. \]  

The rate coefficient for this reaction is obtained from [17], and fitted with an Arrhenius rate Eq. (5.10) with \( c = 7.1 \times 10^{-15} \text{ m}^3 \text{ s}^{-1}, \quad q = 0.4 \) and \( E = 13.6 \text{ eV} \).

- **Stepwise electron impact ionization.** Ionization can also take place from an excited state to the ion state. This reaction is generally a lot faster than ionization from the ground state. In this case the rate-determining step becomes excitation from the ground state to the first excited state [18] (this process is of lesser importance in lower-density plasmas, as the radiative decay of the excited state may prevent ionization). Using the cross sections from Ref. [17], the excitation rate from the ground state to the first excited state, and thus the rate of stepwise ionization, can be approximated by Eq. (5.10) with \( c = 1.24 \times 10^{-14} \text{ m}^3 \text{ s}^{-1}, \quad q = 0.3 \) and \( E = 10.2 \text{ eV} \). A similar process may occur for excitation to and subsequent ionization from the second or even higher excited state However, as the cross sections are smaller and the energy gaps are bigger for excitation to higher excited states, these processes are slower (typically by orders of magnitude) than stepwise ionization via the first excited state. Stepwise processes to higher excited states are therefore not taken into account.

For all processes, the reverse process rates are computed using detailed balancing.

Note that in Eq. (5.3) diffusion losses are not taken into account. In order to verify the assumption that diffusion is negligible, we will estimate the diffusion frequency with

\[ \nu_{\text{diff}} = \frac{D_{\text{amb}}}{\Lambda^2}, \]  

where \( D_{\text{amb}} \) is the diffusion coefficient and \( \Lambda \) a typical length scale. The typical diffusion coefficients are in the order of 0.1 m\(^2\) s\(^{-1}\), while the typical diffusion length can be approximated by the radius of the channel. Thus, we obtain a typical loss frequency in the order of 10\(^7\) Hz. This is small compared to the typical frequencies at which species are formed and destroyed, which are in the order of 10\(^8\)-10\(^9\) Hz. Hence, we can ignore diffusion and still expect reasonable results.

Equation (5.4) is used to compute the electron temperature field. The first term of this equation describes the temporal behavior of the heat content of the electron gas, in which we used

\[ C_e = \frac{3}{2}k_B n_e. \]
with $k_B$ Boltzmann’s constant. This assumes that the electrons behave as a perfect gas. The electron heat conductivity $\lambda_e$ is obtained from the Frost mixture rules [19]. These are valid provided the electron Hall parameter is much smaller than unity. This condition is satisfied during most of the discharge, albeit only marginally during the first 10 ns. The heat transfer rate $k_{\text{heat}}$ is determined using Ref. [11], ignoring the effect of the inelastic collisions of electrons with molecular hydrogen. The conductivity $\sigma$ is computed using the Frost mixture rules.

The last term in Eq. (5.4) is the contribution to the electron heat by reactions. It is assumed that the electrons provide and receive the energy involved in all reactions.

In Eq. (5.4), bound-bound, free-bound, and free-free radiation losses are neglected. Bound-bound radiation is of minor importance, as electron impact bound-bound transitions are much faster than the radiative bound-bound processes in this plasma. Free-bound and free-free radiation typically radiate less than 0.01% of the dissipated power.

In Eq. (5.5), the heavy particle temperature is computed. The first term of this equation describes the temporal behavior of the thermal energy of this gas, in which we use

$$C_h = \frac{3}{2} k_B \left( n_H + n_{H_2} + n_{H^+} \right).$$ (5.14)

The impact of heat capacity due to ro-vibrational excitation of hydrogen is negligible, as virtually all hydrogen is dissociated before the heavy particles have heated up significantly.

Ignoring the ro-vibrational excitation of molecular hydrogen is justified given that the dissociation is much faster than the excitation in this particular plasma. The heat conductivity $\lambda_h$ is computed with the mixture rules in Ref. [11]. The last term on the left-hand side represents the viscous dissipation. Under the assumption that the plasma is Newtonian, and using the simplification that only velocity components in the radial direction are involved, we can write

$$\bar{\tau} : \vec{\nabla} \vec{u_r} = \frac{4}{3} \mu \left[ \left( \frac{\partial u_r}{\partial r} \right)^2 - \frac{\partial u_r}{\partial r} \frac{v_r}{r} + \left( \frac{v_r}{r} \right)^2 \right].$$ (5.15)

### 5.2.3 Boundary conditions

For solving these differential equations, it is necessary to have boundary conditions.

The code we use is a two-dimensional (2D) code, which is used to solve a one-dimensional (1D) problem. This can be accomplished by setting the boundary conditions in the $z$ direction to homogeneous Neumann conditions. This is not an efficient method, but as the computational time of our simulation was quite manageable (about 1 day on a high-end PC), this is not an important issue.
While most of the other boundary conditions used are quite straightforward, the wall boundary conditions for the temperature are not. We will offer a detailed description of these boundary conditions below.

The wall material is an electrical isolator; hence, the amount of free electrons in this material will be negligible. This means that the plasma electrons do not directly transfer their energy to the wall. The presence of a virtually electron-free plasma sheath further hampers the heat transport by electrons to the wall. We therefore take a homogeneous Neumann boundary condition for the electrons at the wall, that is

\[
\left. \frac{\partial T_e}{\partial r} \right|_{r_0} = 0. \tag{5.16}
\]

The heavy particles do transfer their heat to the wall. This means that two conditions, namely the continuity of the temperature and of the heat flux, must be simultaneously satisfied,

\[
T_h \big|_{r_0} = T_{\text{wall}} \big|_{r_0} \tag{5.17}
\]

and

\[
\lambda_h \left. \frac{\partial T_h}{\partial r} \right|_{r_0} = \lambda_{\text{wall}} \left. \frac{\partial T_{\text{wall}}}{\partial r} \right|_{r_0}, \tag{5.18}
\]

with \(r_0\) the radius of the channel, \(T_{\text{wall}}\) the wall temperature and \(\lambda_{\text{wall}}\) the heat conductivity of the wall. In order to obtain a usable boundary condition from Eq. (5.17) and Eq. (5.18), we have to create a model of the heating of the wall to obtain \(T_{\text{wall}}\) and \(\lambda_{\text{wall}}\).

The heating of the wall is described in a one-dimensional model. We only take into account the temperature changes,

\[
\frac{\partial C_{\text{wall}} T_{\text{wall}}}{\partial t} - \nabla \cdot (\lambda_{\text{wall}} \nabla T_{\text{wall}}) = 0, \tag{5.19}
\]

where \(C_{\text{wall}}\) is the heat capacity of the wall material per cubic meter. It is not possible to solve Eq. (5.19) analytically, as \(C_{\text{wall}}\) and \(\lambda_{\text{wall}}\) are functions of temperature; therefore, we solve Eq. (5.19) numerically.

The value of \(C_{\text{wall}}\) was obtained by fitting Einstein’s formula [20] for the specific heat of a solid to measured specific heats of alumina from Ref. [21] in the temperature range from 300 to 600 K:

\[
C_{\text{wall}} = 3n_{\text{wall}} k_B \left[ \left( \frac{\theta_E}{T_{\text{wall}}} \right) \frac{\exp \left( \frac{\theta_E}{2T_{\text{wall}}} \right)}{\exp \left( \frac{\theta_E}{T_{\text{wall}}} \right) - 1} \right]^2, \tag{5.20}
\]

with \(n_{\text{wall}}=1.17 \times 10^{29} \text{ m}^{-3}\) the density of alumina in particles per cubic meter and \(\theta_E=690\) K the Einstein temperature. The fit was accurate to within 2 %. 
The value of $\lambda_{\text{wall}}$ was obtained by fitting the theoretical $1/T$ relation for the thermal conductivity at high temperatures [22, 23] to measured thermal conductivities of alumina from [21] in the temperature range between 273 K and 973 K:

$$\lambda_{\text{wall}} = \frac{9.5 \times 10^3 \text{K}}{T_{\text{wall}}} \text{Wm}^{-1} \text{K}^{-1}. \quad (5.21)$$

This expression is expected to be accurate to within 20%.

We do not solve Eq. 5.19 for the whole wall. Instead, we estimate which part of the wall will be heated significantly by the plasma. This is the innermost 500 nm. As the rest of the material does not heat up significantly, we assume a Dirichlet boundary condition with a value equal to the initial temperature for outer boundary condition of Eq. 5.19.

The system of plasma and wall temperature equation is solved by a sequential substitution procedure, which will be sketched. In this procedure, we first solve the plasma equations (5.1)–(5.4). Equation (5.5) is then solved with a Dirichlet boundary condition for the heavy particle temperature. This boundary condition follows from the value of $T_{\text{wall}}$ and Eq. (5.17). From the obtained $T_{\text{p}}$-field a heat flux into the wall is computed. Using Eq. (5.18), we get a boundary condition for Eq. (5.19). Solving this equation produces a $T_{\text{wall}}$ field. This procedure is repeated until convergence is reached.

### 5.2.4 Adsorption of hydrogen on the wall

Alumina can adsorb hydrogen. This hydrogen can be released when the wall is subjected to heating and ion bombardment from the discharge. We do not describe this process in detail; rather, we assume the hydrogen of the wall to be present at startup, evenly distributed throughout the discharge. Provided the hydrogen on the wall is a small fraction of the total hydrogen content, ignoring the release mechanism is not expected to influence the results greatly.

The number of surface sites $A_s$ on the alumina wall can be estimated as

$$A_s = \frac{2}{\sqrt{3}} n_{\text{wall}} \quad (5.22)$$

if the surface were a perfectly flat plane. However, this is very likely not to be the case. We multiply the number of surface sites with a factor of 3 to compensate for this, and for the possibility of hydrogen desorbing from deeper layers of the alumina. Assuming every surface site holds one hydrogen atom, the number of hydrogen atoms adsorbed to the capillary wall $N_{\text{wall}}$ given by

$$N_{\text{wall}} = 2\pi r_0 l A_s. \quad (5.23)$$
As mentioned, in the model, we distribute this hydrogen through the channel at startup. This leads to an additional hydrogen density due to wall desorption \( n_{\text{surf}} \) of

\[
n_{\text{surf}} = \frac{2A_s}{r_0}.
\]  

(5.24)

5.2.5 The code

For the simulations, we use the PLASIMO code. This code is described in detail in [15, 24–27]. It is a modeling platform that can handle LTE and non-LTE plasmas, currently in two dimensions. Furthermore, its modular structure allows for easy expansion of the code. It has been applied to simulate a wide variety of plasma as described in Refs. [28–33]. Salient additions for this work include the wall heat transport module and algorithmic changes to improve the treatment of plasmas with very high ionization degree.

5.3 The discharge parameters

The model we have created is suitable for a wide range of pulsed capillary discharges. In order to validate our model, we will use the specific discharge parameters of an existing pulsed capillary discharge, namely the discharge that is described in Ref. [8]. This discharge was simulated earlier by Bobrova et al. [9] using the MHD equations and a generalized Saha equation. The main difference between this approach and our non-LTE approach is that in our approach the finite reaction speed is taken into account.

The capillary under investigation has a radius of 150 \( \mu \)m. The current \( I(t) \) is given by:

\[
I(t) = I_0 \sin \left( \frac{\pi t}{200 \text{ ns}} \right), \quad 0 \text{ ns} < t < 200 \text{ ns}
\]  

(5.25)

with \( I_0 \) equal to 300 A for the main study. We will also make a parameter study in which \( I_0 \) is varied.

There are two contributions to the hydrogen density. The first contribution is the hydrogen prefilling. For this discharge, the initial \( \text{H}_2 \) density was \( 1.68 \times 10^{24} \text{ m}^{-3} \). The second contribution is the hydrogen that is liberated from the wall. Using Eq. (5.24), we obtain that the amount of \( \text{H}_2 \) from this source is \( 4.8 \times 10^{23} \text{ m}^{-3} \). The total amount of \( \text{H}_2 \) is thus \( 2.16 \times 10^{24} \text{ m}^{-3} \).

In order to initiate the discharge, a small preionization is necessary. The following composition of the initial plasma is assumed: \( n_{\text{H}_2} = 2.0 \times 10^{24} \text{ m}^{-3}, n_{\text{H}} = 2.8 \times 10^{23} \text{ m}^{-3}, n_{\text{H}^+} = n_e = 4 \times 10^{21} \text{ m}^{-3} \). The initial electron temperature is 1 eV, the initial heavy particle temperature is 25.9 meV (corresponding to 298 K). The plasma is initially at rest and homogeneously distributed in the channel.
5.4 Results and discussion

5.4.1 General results and discussion

The pulsed capillary discharge is simulated using the physical model described in Sec. 5.2 for the parameters in Sec. 5.3. Graphs of the electron density, electron temperature, the atomic hydrogen density, and heavy particle temperature as functions of time and radial position are presented in Fig. 5.1, Fig. 5.2 and Fig. 5.3, respectively. The discharge is strongly dynamic, and based on the plasma parameters, three phases can clearly be distinguished.

- The ionization phase. Lasting from the startup to about 30 ns, during this phase an almost homogeneous ionization and dissociation of hydrogen takes place.

- The formation phase. During this phase, the effect of wall cooling, amplified by the redistribution of current, forms the guiding channel with a hollow electron density profile. This phase lasts from 30 to 60 ns.

- The guiding phase. The guiding channel remains relatively stable during this phase, lasting from 60 ns to the end of the simulation.

We will discuss these phases in more detail.

In the ionization phase, during the first few nanoseconds, the electrons heat up. This causes a dramatic rise in the ionization rate and consequently of \( n_e \). The high temperature and increasing electron density rapidly dissociates the molecular hydrogen. The
Figure 5.2: The electron temperature $T_e$ as a function of time and radial position. The isolines indicate differences of 0.5 eV.

Figure 5.3: The heavy particle temperature $T_h$ as a function of time and radial position. The isolines indicate differences of 0.5 eV.
electron density during this phase, from 5 to 30 ns, increases far more rapidly than the power, which implies that the electron temperature drops slightly, from around 3 eV at 5 ns to 1.7 eV at 30 ns. This behavior is a result of the finite reaction speed which is used in a non-LTE approach; in a 2-temperature Saha approach, the electron temperature rises during this phase [9]. In our model, the rise of the heavy particle temperature is initially quite slow, as electron-neutral collisions are not very effective in transferring heat from the electrons to the heavy particles.

In the formation phase, wall cooling shapes the channel. The heavy particles are directly cooled at the wall. However, heat transfer from the electrons to the heavy particles also cools the electrons here. This causes the ionization to be reduced locally. Furthermore, the reduced ionization means that the local electrical conductivity is lower, causing less current to flow which leads to a reduced Ohmic dissipation in the wall region. This further amplifies the temperature drop at the wall.

The higher $T_h$, $T_e$, and $n_e$ in the center all cause the pressure there to be much higher than at the wall. This causes a bulk flow from the center to the wall. Maximum velocities are reached in the order of $2 \times 10^3$ m s$^{-1}$. Comparing this with the thermal velocity leads to a Mach number which can reach values of up to $M=0.15$, meaning that the flow remains subsonic. During this phase, the ionization degree keeps rising. The central ionization degree is well above 99% at 60 ns. $T_e$ starts rising again, especially when the plasma is nearly fully ionized, as no more energy is consumed by further ionization. $T_h$ also rises due to the higher collision frequency between electrons and ions. It becomes almost equal to the electron temperature at 40 ns, except near the wall. From this time onward, $n_e$ is nearly equal to the value predicted by the Saha equation with the temperature given by $T_e$ [34].

The lower temperature at the wall has two opposing effects on $n_e$. The total particle density, and therefore the electron density, becomes higher because the pressure remains roughly equal in the channel. However, the lower temperature at the wall means the plasma is not fully ionized near the wall.

The creation of pressure differences is amplified by redistribution of current. The higher temperature in the center causes higher conductivity there, which causes a higher local current density. The net effect of this is an increased Ohmic dissipation density, causing further increases in central temperature and pressure.

During the stable phase, the plasma parameters remain roughly stable over tens of nanoseconds. The ionization degree is well over 99%, except in the outer 10 µm of the channel, where the ionization degree drops rapidly, to a few percent near the wall. As long as the laser does not come close to the walls, this is not expected to be detrimental, as the center is fully ionized. The total average ionization degree is around 56% at 60 ns.
5.4.2 Error analysis and comparison with experiments

We will consider the most important sources of error in the model and discuss their impact on the plasma parameters. Furthermore, we will compare our model results with the experiment.

The key plasma parameters $n_e$, $T_e$, and $T_h$ are remarkably robust with respect to variations in the typically rather inaccurate reaction rates and transport coefficients.

With respect to the reaction rates, we note that the equilibrium composition is not influenced by the reaction rates, as backward and forward rate are linked via detailed balancing. The reaction rate would impact the electron temperature and density during startup.

The most important transport coefficients for the formation of the channel are the electrical conductivity, and the thermal conductivity. Because there is a strong positive correlation between temperature and the loss term (thermal conductivity) and a strong negative correlation to the the production term (ohmic dissipation), errors in the transport coefficients cause only small errors in the temperature fields.

There are two aspects of the model which could be improved:

- Diffusion is not included. While the typical diffusion time scales are slower than the other typical time scales of the discharge, it is expected to have an effect on the plasma parameters, especially near the wall.

- A better model of the hydrogen desorbing from the wall could reduce the uncertainty in the amount of hydrogen which is in the discharge.

Given these uncertainties, we do expect the model to predict electron densities with an accuracy of about 30%, and the temperatures with an accuracy of about 10%.

In order to verify the validity of our model, we compare the modeling results with the experimental results published in Ref. [8] in Fig. 5.4. Given that the error in the measurements are claimed to be 12%, the quantitative agreement between the electron density in experiment and model is satisfactory. The sharp peaks in the electron density near the wall, a result of the lower temperature near the wall, are absent in the measurements.

5.4.3 Suitability for guiding

For the guiding of a laser, the electron density profile is the key parameter. A graph of the electron density at various times as a function of the radius is given in Fig. (5.4). As can be seen, during the guiding phase, the electron density profile only superficially resembles a parabola; it is much flatter in the center. This is caused by the heat conductivity, which is much higher in the center than near the wall. This causes the
temperature gradients to be smaller in the center and hence the temperature profile to be flatter. This, in turn, makes the electron density profile flatter. A graph of $\lambda_h$ and $\lambda_e$ at 100 ns is shown in Fig. 5.5, while a graph of $T_e$ and $T_h$ at 100 ns is shown in Fig. 5.6.

While the electron density over the whole channel does not resemble a parabola, the electron density in the central region of the channel does. For $r < 90 \, \mu m$ and $t > 80 \, ns$, the central density $n'_e$ can be described well by:

$$n'_e = \left( 0.85 \left( \frac{r}{r_0} \right)^2 + 1.86 \right) \times 10^{-24} \, m^3. \quad (5.26)$$

For a Gaussian beam, the matched spot size $W_m$ is given by [9]

$$W_m = \left( 0.5\pi r_e \frac{\partial^2 n_e}{\partial r^2} \right)^{-1/4} \quad (5.27)$$

with $r_e$ the classical electron radius which has a numerical value of $2.817 \times 10^{-15} \, m$. Combining Eq. (5.26) and Eq. (5.27) gives a matched spot size of $42 \, \mu m$. Because the matched spot size is much smaller than the part of the discharge in which the electron density is approximately parabolic, the fact that the electron density is not parabolic over the whole channel is of little importance for guiding.
Figure 5.5: The electron thermal conductivity $\lambda_e$ and the heavy particle heat conductivity $\lambda_h$ as a function of the radius at 100 ns. In most of the plasma, the lighter electrons cause most of the heat transport. However, near the wall, the lower electron density and higher heavy particle density cause the $\lambda_h$ to be larger than $\lambda_e$. 

Excessive heating of the wall during the discharge might cause the practical problem of loss of wall material, thus reducing capillary lifetime and polluting the plasma with elements that cannot be fully stripped of electrons by the plasma. Furthermore, it will lead to cooling of the plasma. The temperature of the wall-plasma interface, which is the hottest point of the wall, reaches its highest value of 1780 K at $t=153$ ns. This is still well below the melting point of alumina. Furthermore, the low electron temperature near the wall means that ions impact the wall with little energy [35], meaning there is little sputtering. Hence, we expect very long capillary lifetimes, which is also found in the experiments [8]. This is important for practical applications.

5.4.4 The effect of non-LTE

As discussed, one of the main differences between our work and the earlier work of Bobrova et al.[9] is our adoption of a non-LTE model as opposed to a two-temperature magneto-hydrodynamic (MHD) system where the free energy is minimized. We will explore two non-LTE effects: the effect of the finite reaction speed on composition and the temperature difference between electrons and ions.

One of the effects of the finite reaction speed is an electron density which differs
from the Saha density. We present the overpopulation of the electron density and the Saha density $n_{\text{Saha}}$ as a function of time for various radial positions in Fig. 5.7. This figure shows that during the startup phase, $n_e$ is much lower than $n_{\text{Saha}}$. It takes about 60 ns for the electron density to reach the Saha density. This is in line with the estimated reaction speeds, which are in the order of $10^8$ Hz.

The finite heat transfer rate between electrons and ions allows the electrons to have a different temperature than the ions. We present the difference between the electron and ion temperature as a function of time for various radial positions in Fig. 5.8. As can be seen, the difference between $T_e$ and $T_h$ rises sharply during the first few nanoseconds, as $T_e$ rises while $T_h$ remains almost constant. After this phase, $T_e$ remains roughly constant until about 30 ns, while $T_h$ rises due to the heat flow from the electrons. While the rising ionization degree increases the collision frequency and hence $k_{\text{heat}}$, the decreasing difference between $T_e$ and $T_h$ roughly compensates for this, causing a roughly linear decrease of $T_e-T_h$ until the difference becomes negligible. The bump of $T_e-T_h$ at the center of the channel at about 60 ns is the result of the redistribution of current to the center.
**Figure 5.7:** The ratio of the electron density and the Saha density as a function of time, at three different radial positions.

**Figure 5.8:** The difference between $T_e$ and $T_h$ as a function of time, at three different radial positions.
5.4. Results and discussion

Figure 5.9: The computed electron density \( n_e \) as a function of radius for three different currents. Furthermore, a parabolic approximation in the central part of the discharge is given.

5.4.5 A parameter study with varying current

In the previous part of the discussion, a particular example of the pulsed capillary discharge has been simulated, and the results discussed and explained in great detail. Having established that our model produces physically relevant results for this case, we will now use this model to perform a parameter study by varying \( I_0 \) between 200 and 500 A.

We will restrict ourselves to the discussion of the result that is the most relevant for the guiding properties: \( n_e \) as a function of \( r \), at \( t=80 \text{ ns} \). At this time, all channels that were under investigation reached the stable phase. Graphs of \( n_e \) as a function of \( r \) for selected currents (200 A, 350 A and 500 A) are given in Fig. 5.9.

For all the values of the current that were investigated, a parabolic approximation of \( n_e^c \), described by

\[
 n_e^c = a + \frac{1}{2} b r^2, \tag{5.28}
\]

with \( a \) the on-axis density and \( b \) the second derivative of the central density, was fitted to the results. These curves are also drawn in Fig. 5.9. For all investigated currents, the values of \( a \) and \( b \) are given in Table 5.1.

From Fig. 5.9, we see that the match between the parabolic fit and the computed values of \( n_e \) is good for \( r < 90 \mu\text{m} \). As discussed, this means that the channel is wide enough for laser guiding. It can be clearly seen that the maximum of the electron density
shifts outwards and reaches a higher value for higher current. This is caused by the higher temperature near the wall, which causes a higher degree of ionization near the wall. Because a hydrogen ion plus a hydrogen electron produce more pressure for a given temperature than a hydrogen atom, the density near the wall is lower for a higher current, causing the central density to be higher. This trend is clear in the values of $a$ in Table 5.1.

Generally, the curvature of the channel $b$ did not depend strongly on the current, as can be seen in Table 5.1, with the exception of the lowest value of the current, 200 A. For this current, a significant part of the channel is not fully ionized. The relative insensitivity of $b$ to the value of the current $I$ makes $I$ a poor choice for tuning the guiding behavior of the channel, especially given that $W_m$ is proportional to $b^{-1/4}$ as described in Eq. (5.27).

A practical problem when increasing the current is the heating of the wall. While the wall temperature stays below the melting point of alumina of 2326 K [21] for the default current of 300 A, the wall material may melt for higher values of the current. Note that this does not influence the results presented here, as the results are taken at $t=80$ ns, well before the wall temperature reaches the melting point of alumina. At temperatures above the melting point, the wall model cannot accurately predict the temperatures anymore, as Eq. (5.20) and Eq. (5.21) are valid for solids only. A precise prediction of the current at which evaporation of wall material becomes significant is beyond the scope of this study.

### Table 5.1: The parameters $a$ and $b$ of a the parabolic approximation of the electron density in the central part of the channel, for various currents. $a$ and $b$ are defined in Eq. (5.28).

<table>
<thead>
<tr>
<th>Current (A)</th>
<th>$a$ ($10^{24}$ m$^{-3}$)</th>
<th>$b$ ($10^{31}$ m$^{-5}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>200</td>
<td>1.59</td>
<td>4.81</td>
</tr>
<tr>
<td>250</td>
<td>1.62</td>
<td>6.44</td>
</tr>
<tr>
<td>300</td>
<td>1.74</td>
<td>6.8</td>
</tr>
<tr>
<td>350</td>
<td>1.87</td>
<td>7.15</td>
</tr>
<tr>
<td>400</td>
<td>2.00</td>
<td>7.37</td>
</tr>
<tr>
<td>450</td>
<td>2.13</td>
<td>7.51</td>
</tr>
<tr>
<td>500</td>
<td>2.26</td>
<td>7.65</td>
</tr>
</tbody>
</table>
5.5 Conclusions

We have presented a theoretical model suitable for simulating a slow capillary discharge in hydrogen. This model has been used to simulate the discharge described in Ref. [8].

The model uses a non-LTE description, which has not yet been used for the description of these types of discharges to our knowledge. Previous models [9] used a two-temperature approach in which the free energy was minimized. The advantage of the current approach is that the effect of the finite reaction speed can be included. The heating of the wall is also numerically described, and the plasma and wall models are coupled.

The model predicts three distinctive phases in the discharge:

- The ionization phase. During this phase, the plasma ionizes roughly homogeneously. It lasts about 30 ns for the discharge under study. The finite reaction speed causes the electron temperature to drop, rather than rise, from 5 to 30 ns.

- The formation phase. Heat conductivity to the wall, becoming more significant with rising ionization and temperatures, starts to create significant temperature and pressure gradients. Bulk flow, which remains subsonic, counteracts this, causing a bulk density gradient, and, given the almost homogeneous full ionization, a hollow electron density profile. This phase lasts up to 60 ns.

- The stable phase. With the Ohmic dissipation and heat conductivity roughly balancing, and the plasma close to Saha equilibrium, the plasma enters a phase during which the plasma parameters are roughly stable over many tens of nanoseconds. The heating of the wall and the changing current cause slow variations in the plasma parameters. Given that a stable, fully ionized channel with a hollow electron density profile is formed for much longer than the sub-ns time it takes for the laser to pass, we expect this phase to be most suitable for guiding.

The model is expected to predict the electron density with an accuracy of 30%. Comparison of the model results with measurements [8] gives satisfactory agreement. This indicates that the used model is indeed suitable for the computation of pulsed capillary discharges.

The wall of the plasma heats up significantly, up to a temperature of 1780 K. This means however that the wall will not melt, which is desirable from a practical point of view. Furthermore, the magnitude of the wall heating demands that a robust and accurate model is used to describe it, such as the one used in this work.

The fact that the thermal conductivity is much higher in the center than near the wall causes the temperature, and thus the electron density profile, to be much flatter in the center than near the wall. This means that the that is not truly parabolic, but more
block-like. However, as the central part of the profile is nearly parabolic, this is not expected to be detrimental to guiding.

A parameter study in which the current was varied has been carried out. It was found that the influence of the current on the key guiding parameters is small, especially for currents above 250 A. Excessive wall heating might be detrimental to the practical use of currents above 300 A.

Acknowledgments

The authors would like to thank Zahoor Ahmad, Wang Chunmei, and Elena Toma for the fruitful discussions. The PLASIMO team members, current and former, are acknowledged for their contributions to the code. This work was performed as part of the research program of ‘Stichting voor Fundamenteel Onderzoek der Materie’ (FOM) with financial support from NWO, within FOM program 55 on Laser Wakefield Accelerators.

Bibliography


Abstract–
Slow pulsed capillary discharges are currently under investigation for use as plasma channel waveguides in laser-wakefield acceleration and XUV generation. In this work, a parameter study is performed on this device using a combination of two models, namely a non-local thermal equilibrium (non-LTE) plasma model and a wall temperature model that is coupled to it. This model has been validated against experiments. In the present study, two parameters are varied, the initial density and the channel radius. These parameters have a strong influence on the guiding properties. The results of this parameter study can be summarized in a single, empiric formula describing the matched spot size as a function of the initial density and the channel radius. This formula is expected to give a good prediction of the matched spot size, provided that no wall ablation occurs, diffusion is limited, and that the current pulse is sufficient in amplitude and duration for a well-ionized, stable plasma to form. This has been verified for the parameter range studied here.

6.1 Introduction

Pulsed capillary discharges are plasmas that are characterized by a very high power density and ionization degree, combined with a small geometric size. Various types exist, such as discharges created in ablated wall material [1, 2] and fast [3, 4] and slow [5, 6] capillary discharges in a prefilled channel. Their field of application currently includes laser-wakefield acceleration [7, 8] and XUV generation [9, 10].

This Chapter, in slightly altered form, has been accepted for publication in *Journal of Physics D: Applied Physics* by the authors B. H. P. Broks, W. van Dijk and J. J. A.M. van der Mullen.
The slow capillary discharge consists of a capillary that is made of a material that is resistant to thermal shock, such as alumina, sapphire or quartz. This capillary is filled with a gas that is easily fully ionized, such as helium or hydrogen, with a pressure typically between $10^2$ and $10^4$ Pa. A current pulse of a few hundred ampere that lasts for a few hundred nanoseconds is then sent through the capillary. This pulse causes nearly full ionization of the plasma. However, mainly due to the short duration, the heat load on the walls is low enough so that ablation of wall material is avoided. This allows this channel to be reused many times.

Our first simulations [11] have been performed on an experimentally used channel, producing results that match well with experimental results for the plasma properties that are of interest. The next step is a systematical modeling of the channel to explore the parameter space.

In [11], the influence of the current on the guiding properties has also been studied. It was found that for a wide range of currents, the plasma and guiding properties are only weakly dependent on the current.

In this contribution, we focus on an expansion of [11], namely a study in which two other parameters, the filling pressure and radius, are varied. Unlike previous treatments [12], we use a fully self-consistent two-region non-Local Thermal Equilibrium (non-LTE) simulations, in which the capillary wall is treated explicitly. Such a treatment is indispensable for accurate simulation of both the plasma and the wall for a wide range of filling densities and radii, and, consequently, a prediction of the laser guiding properties of the plasma and the stability of the wall under the heat load of the plasma. Furthermore, the results will be used to obtain a formula that predicts the guiding properties as a function of the parameters. This may guide the design of capillary discharge waveguides.

### 6.2 The physical model

The system is basically a pulsed wall-stabilized high-current hydrogen arc discharge and consists of two regions, the discharge plasma and the wall. These regions are described by separate models, coupled at the interface by boundary conditions. A detailed description can be found in [11]; A brief overview will be given here.

The basis of the plasma model is formed by a two-temperature non-LTE quasineutral fluid model, which is used extensively in the description of low-temperature plasmas and has a wide range of validity. It was found in [11] that the effect of non-LTE is quite pronounced, especially near the wall and during the initial phases of the discharge. It is expected that the wider range of parameters simulated in this work will show even larger degrees of non-equilibrium. This implies that the non-LTE approach
6.3. The parameters is suitable for the present investigation.

We describe the density of four species, namely $e$, $H^+$, $H$, and $H_2$. In principle, the capillary discharge is a two-dimensional system, exhibiting a rotational symmetry. However, since the length of the capillary is typically two orders of magnitude larger than its radius, the discharge can be described well by assuming that it only depends on the radial coordinate $r$ and the time $t$. An upper bound to the plasma efflux at the ends of the capillary can be obtained by assuming the plasma expands with the thermal speed of hydrogen for a time that is equal to the typical time until guiding becomes possible. By substituting data from [11], we obtain an upper bound of 1 mm for the distance over which the plasma expands, which is small compared to the length of the capillary.

The heat transport in the wall is modeled numerically by including the temperature-dependent thermal conductivity and heat capacity of the wall material, in this case solid polycrystalline alumina. By coupling the heat fluxes and temperatures at the interface between the models, a self-consistent model containing the wall and the plasma is created. A good treatment of the wall was found to be of great importance [11], both to be able to predict the occurrence of wall melting and ablation, but also to accurately predict both the electron temperature $T_e$ and the heavy particle temperature $T_h$ of the plasma close to the wall.

For the simulations, we use the PLASIMO code. This code is described in detail in [13–15]. It is a modeling platform that can handle LTE and non-LTE plasmas, currently in two dimensions. Furthermore, its modular structure allows for easy expansion of the code. It has been well validated by using it to simulate a wide variety of plasmas as described in [11, 16, 17].

6.3 The parameters

Using the general physical model outlined in Section 6.2, we can simulate the channel with specific values of the control parameters. The starting point of this parameter study will be an existing channel [5], which has also been the subject of previous modeling studies [11, 12]. From this basic situation, we will vary the density and the channel radius and investigate the impact of these variations on the plasma and the laser guiding properties.

6.3.1 Density

The capillary is prefilled with molecular hydrogen with a certain initial density $n_i H_2$. The hydrogen prefilling is not the only hydrogen that is present in the plasma. The hydro-
gen that is adsorbed to the capillary walls may be released when the wall is heated. This is particularly true for alumina, which is used for the capillary wall, as it is porous. For the typical surface-to-volume ratio of the systems under consideration, the bulk density of hydrogen which desorbs from the wall is estimated to be a few times \(10^{23} \text{ m}^{-3}\) [11], which is a significant but not dominant contribution for the systems under consideration. In this study, it will be assumed that all hydrogen that is present in the capillary, is present from the beginning of the discharge. The difference between the prefilled hydrogen density and the total hydrogen density makes the comparison between theory and experiment less straightforward, because the experimental parameter, the prefilling, and the theoretical parameter, the total hydrogen density, differ. The initial density is varied between \(1 \times 10^{24}\) and \(4 \times 10^{24} \text{ m}^{-3}\).

### 6.3.2 Channel radius

Studying the effect of varying the channel radius \(r_c\) is not straightforward, as it poses an apparent dilemma: Should the current \(I(t)\), the current density \(J(t)\), or another parameter be kept constant? Because the effect of temperature change is already studied in the current parameter study [11], and because the discharge should be in a certain temperature range to perform properly, we would like to keep the temperature relatively steady during the radius parameter study. The temperature in equilibrium can be estimated by the following simplified equation:

\[
- r_c \nabla \cdot \left( \lambda \nabla T \right) = S_{\text{diss}}. \tag{6.1}
\]

with \(\lambda\) the thermal conductivity, \(T\) the temperature and \(S_{\text{diss}}\) the power dissipation density, which is given by:

\[
S_{\text{diss}} = \frac{J^2}{\sigma} \approx \frac{I^2}{\pi^2 r_c^4 \sigma}, \tag{6.2}
\]

with \(\sigma\) the electrical conductivity. In first order, the left-hand side of (6.1) is proportional to \(r_c^{-2}\). Thus, in order to keep \(T\) constant, the right-hand side must have the same \(r_c\) dependence. Using (6.2), we see that \(I\) should be varied proportional to \(r_c\) to achieve this. \(r_c\) is varied between 100 and 200 \(\mu\text{m}\), while the peak current \(I_0\) is simultaneously varied between 200 and 400 A.

### 6.3.3 Current profile

For the abovementioned parameters, series of simulations will be made, in which one parameter is changed and the other is kept fixed at its default values of \(r_c = 150 \text{ \mu m}\) or
6.3. The parameters

\( n_{H_2} = 2.16 \times 10^{24} \text{ m}^{-3} \), while the current \( I(t) \) waveform is given by

\[
I(t) = \begin{cases} 
I_0 \sin \left( \frac{\pi t}{t_0} \right) & \text{0 ns} \leq t \leq t_0 \\
0 & t > t_0
\end{cases}
\]

(6.3)

where \( I_0 \) has a default value of 300 A and \( t_0 \) has a value of 200 ns. This is the same set of parameters used in our earlier study [11], based on a channel used in experiments [5]. In all cases the analysis is carried out at the peak of the current, \( t = 100 \text{ ns} \).

6.3.4 Key assumptions

Two critical assumptions are used in the model: the assumption that diffusion is negligible, and the assumption that the wall does not ablate. These assumptions were made because they do not greatly limit the applicability of the model, as both assumptions should be valid for optimal operation, as is discussed below.

In equilibrium, the electron creation process ionization balances with the destruction process recombination. This balance is disturbed by ambipolar diffusion of plasma, which transports electron-ion pairs from the centers to the wall, where recombination may occur. If the creation frequency is much larger \( \nu_{\text{crea}} \) than the diffusion frequency \( \nu_{\text{diff}} \), creation is in equilibrium with recombination, and diffusion may be neglected.

The diffusion frequency can be estimated from

\[
\nu_{\text{diff}} \approx \left( \frac{k_B T_e}{m_{H^+} \times \sigma_{HH^+} \times u_{H^+} \times n_{H}} \right) L^{-2}.
\]

(6.4)

Here, \( k_B \) is Boltzmann’s constant, \( T_e \) the electron temperature, \( m_{H^+} \) the mass of \( H^+ \), \( \sigma_{HH^+} \) the momentum exchange cross section between \( H \) and \( H^+ \), \( u_{H^+} \) the thermal speed of \( H^+ \), \( n_{H} \) the density of atomic hydrogen and \( L \) the typical diffusion length scale.

The creation frequency can be estimated from the direct and stepwise ionization rate of hydrogen [11, 18]:

\[
\nu_{\text{crea}} = n_H \left( k_{\text{ion}} T_e [\text{eV}]^{0.4} \exp \left( -\frac{E_{\text{ion}}}{k_B T_e} \right) \right) + k_{\text{exc}} T_e [\text{eV}]^{0.3} \exp \left( -\frac{E_{\text{exc}}}{k_B T_e} \right),
\]

(6.5)

with the ionization rate coefficient \( k_{\text{ion}} = 7.1 \times 10^{-15} \text{ m}^3\text{s}^{-1} \), the ionization energy \( E_{\text{ion}} = 13.6 \text{ eV} \), the excitation rate coefficient \( k_{\text{exc}} = 1.24 \times 10^{-14} \text{ m}^3\text{s}^{-1} \), and the excitation energy \( E_{\text{exc}} = 10.2 \text{ eV} \).

Substituting estimated values of \( T_e = 4 \text{ eV} \), \( \sigma_{HH^+} = 1.0 \times 10^{-18} \text{ m}^2 \) [19] and \( u_{H^+} = 1.9 \times 10^4 \text{ m s}^{-1} \) (based on a \( T_h \) of 4 eV), and equating \( \nu_{\text{diff}} \) and \( \nu_{\text{crea}} \), we find the following relation between the diffusion length and \( n_{H} \):

\[
3 \times 10^{18} L[\text{m}] = n_{H}[\text{m}^{-3}]
\]

(6.6)
Equation (6.6) makes it possible to estimate the minimal density needed to restrict diffusion in the plasma to a zone that extends a distance $L$ from the wall. This is the atomic hydrogen density near the wall, which is typically higher than the average density. Hence, the estimate in (6.6) is a conservative estimate of the minimal $n_H$. For an $L$ of 10 µm, which is a significant fraction of the radius for a typical capillary, the minimal $n_H$ is $3 \times 10^{23} \text{m}^{-3}$.

Significant diffusion not only reduces the validity of the model, but could also lead to experimental difficulties. Diffusion and wall recombination have a tendency of creating a convex $n_e$-profile, which precludes laser guiding. Eq. (6.6) makes an estimate of the minimal hydrogen density that is needed to prevent significant diffusion.

Wall ablation may occur if the channel wall is heated to temperatures above their melting point. This is not desirable from an experimental point of view, as the plasma becomes contaminated with oxygen and aluminum, which are not fully ionized at the electron temperatures in the channel. These non-fully ionized ions cause ionization-induced refractive defocusing [20, 21]. Furthermore, damage to the walls may reduce capillary lifetime. While the temperature is difficult to predict a priori, the wall model is capable of computing a temperature profile in the wall. It was found from the modeling that at $t = 100$ ns, the channel wall has not been heated to temperatures above the melting point of alumina; Hence, wall ablation does not occur, and not including it in the model is justified. Furthermore, this study becomes a predictive tool for the parameter range in which ablation does not occur.

6.4 Laser guiding theory

Pulsed capillary discharges are used to guide of laser beams. For the guiding of a Gaussian beam, a concave, parabolic profile of the electron density $n_e$ is needed [8]. Such a profile is given by

$$n_e(r) = a + \frac{1}{2} b r^2. \quad (6.7)$$

Here, $b$ is the second derivative of the electron density, and $a$ is the on-axis electron density.

The matched spot size $W$ of the laser beam is given by [12]

$$W = (0.5\pi r_e b)^{-1/4} \quad (6.8)$$

with $r_e = 2.817 \times 10^{-15} \text{m}$ the classical electron radius. The matched spot size can be obtained by fitting the central electron density with (6.7) and substituting the obtained $b$ in (6.8).
Because the simulated $n_e(r)$ is only parabolic near the center, showing steep gradients near the wall, only the data points near the center, with $r < 0.57 r_c$, are used. This is about two times larger than the typical matched spot size, which means virtually all of the laser energy indeed passes through the part of the discharge where the profile is parabolic. Sample plots of $n_e$ and the corresponding parabolic fit will be presented to demonstrate the quality of the fitting procedure.

6.5 Results and discussion

In this Section, we will discuss the influence of the control parameters on the plasma properties, and in particular $n_e$, from which the guiding properties are derived. For a thorough discussion of the plasma physics that cause the profile formation, the reader is referred to [11].

6.5.1 Density

The initial total density $n_{H_2}$ as described in Sec. 6.3. For selected values of $n_{H_2}^i$, the simulated electron densities and parabolic fits to these profiles are given in Figure 6.1. The resulting guiding parameters $a$ and $b$ are plotted in Figure 6.2.
Figure 6.2: The $a$ and $b$ parameter, which characterize the central electron density profile, as a function of the initial density. Lines have been fitted to show that $a$ and $b$ are linearly dependent on the density. The line fitted through the data points of $b$ is given in (6.9)

In Figure 6.2, we see that $a$ depends linearly on $n_{H_2}^i$. However, the slope of the line that is fitted through the data points is not unity but only 0.74, which means that the central density rises more slowly than the average density. There are two reasons why this happens.

The first reason can be explained using the Le Chatelier principle: if an equilibrium is disturbed by a change in conditions, the equilibrium shifts to counteract this change. In this case, increasing the density will decrease the ionization degree, in particular near the wall. The lower ionization degree will lead to a somewhat higher mass density near the wall, and a somewhat lower central mass (and electron) density.

The second reason is that $T_h$ and $T_e$ are lower near the wall when the amount of initial hydrogen is higher. The reason for this is that the energy consumed in ionizing the plasma, which is roughly proportional to the amount of hydrogen present, is not available to heat the wall and the plasma next to it, causing lower temperatures. A lower temperature near the wall means that the density becomes higher near the wall due to the fact that the pressure is roughly uniform over the plasma. Furthermore, a lower temperature causes a lower degree of ionization, causing the density to be even higher.

In Figure 6.1, we see that both the central $n_e$ ($a$) and $\frac{\partial^2 n_e}{\partial r^2}$ ($b$) increase with increasing initial density, as discussed above. However, we also see that the maximum of $n_e$
shifts inward for higher initial densities. This is consistent with the above discussion, as it predicts a lower degree of ionization and hence a lower $n_e$ value near the wall for increasing initial density.

Furthermore, we see that for each case, the parabolic fit of the data is excellent. The error in the fit is less than 2% for each case investigated. This suggests that good laser guiding is possible for the $n_{H_2}$-range under study.

The parameter $b$ increases linearly with increasing $n_{H_2}$, and can be approximated well by

$$b[m^{-5}] = 3.2 \times 10^7 n_{H_2}^i [m^{-3}],$$

as can be seen in Figure 6.2.

It is worth noting that channels with a higher density take a longer time to form. In order to facilitate the discussion on this behavior, we introduce the dimensionless parameter $Z$, the ratio between free and total electrons:

$$Z = \frac{n_e}{n_e + n_H + 2n_{H_2}}.$$  \hspace{1cm} (6.10)

In Figure 6.3, the axial value of $Z$ is plotted as a function of time. It indicates that the time needed for near-total ionization ($Z \approx 1$) increases with increasing filling density. However, this increase is less than linear in the density. Plasmas with a lower ionization degree and temperature have a lower conductivity and therefore a higher dissipation,
Figure 6.4: Three simulated electron density profiles at $t = 100$ ns, and parabolic fits of the central electron density. The plasma structure is roughly similar in all three cases. This means channels with the smaller radii have higher density gradients.

which largely compensates the fact that plasmas with a higher initial density need more energy to become fully ionized. The results plotted in Figure 6.3 indicate that increasing the duration of the current pulse with increasing density may be necessary to reach full ionization during the discharge, if even higher densities are studied. For lower densities, reducing the duration of the current pulse may be used to reduce the heat load on the wall, because ionization is more rapid in this case. This also has an influence on the wall temperature, which varies between $1.4 \times 10^3$ K for $n_{H_2} = 1 \times 10^{24}$ m$^{-3}$ and $1.1 \times 10^3$ K for $n_{H_2} = 4 \times 10^{24}$ m$^{-3}$. For the entire range of densities, the temperature remains below the melting point of alumina.

### 6.5.2 Channel radius

The radius of the channel $r_c$ has been varied as described in Sec. 6.3. For selected values of $r_c$, the simulated electron densities and parabolic fits to these profiles are given in Figure 6.4. The resulting guiding parameters $a$ and $b$ are plotted in Figure 6.5.

Figure 6.4 shows that a decrease in $r_c$ causes a compression of the features of the plasma in the radial direction. The parabolic fits describes the profile well, having a standard error of at most 2%.

For $a$, one would expect no significant dependence on the radius, because the av-
6.5. Results and discussion

Figure 6.5: The fitting parameters of the parabola that describes the central electron density profile. The on-axis density $a$ is not very dependent on the radius. The second derivative of the density $b$ increases slightly stronger than $r_c^{-2}$.

Average density in all plasmas is equal, and we have taken care to keep the temperature approximately constant for all the cases considered. For $b$, (6.7) suggests an approximate $b \propto r_c^{-2}$ relationship.

Figure 6.5 show that in first order, these trends are well reproduced. However, there are deviations, which are caused by differences in temperature profile between the channels of various radii. There is a slight trend toward increasing $a$ for decreasing radius. The dependence of $b$ on $r_c$ can be described by

$$b [m^{-5}] = 1.74 \times 10^{23} (r_c [m])^{-2.25},$$

(6.11)

which is a slightly stronger dependence than one would expect from (6.7). This is explained as follows.

The wall load, i.e. the power per wall area, is larger for smaller channels. This causes a higher wall temperature for channels with a smaller radius. For instance, at 100 ns, the wall temperature is $2.2 \times 10^3 K$ for $r_c = 100 \mu m$, while we find $9 \times 10^2 K$ for $r_c = 200 \mu m$. Higher wall temperatures cause a decrease of the density near the wall, increasing the central density. The decrease in density near the wall is visible in Figure 6.4 as a decrease in $n_e$. The density decrease at the wall causes an increase in $a$ and $b$ for smaller radii, as observed in Figure 6.5.

The applied current scaling is chosen such that the increased wall losses for smaller
radii are compensated by higher Ohmic dissipation densities. However, this difference in dissipation density does influence the speed at which the plasma changes from its initial state, which has a very low $Z$-value, to the laser-guiding state, which has a $Z$ of near unity in the central channel. This is illustrated in Figure 6.6, where the central $Z$ is plotted as a function of time. It is shown that the wider channels take longer to form, which is expected given the fact that their dissipation density is lower. Figure 6.6 also provides insight in the current pulse duration required for a channel to form. Optimizing the current pulse duration and amplitude could be used to regulate the wall load while maintaining a high central $Z$.

### 6.6 A formula for the guiding properties

From an application point of view, the key parameter of the channel is the matched spot size of the laser beam that can be guided through the channel. We will use the results as described in Sec. 6.5 to construct a formula for the matched spot size for as a function of various parameters.

It is known [11, 12] that after a stable channel is formed, the plasma properties remain almost independent of time for a time window that lasts several tens of nanoseconds, i.e. much longer than the laser pulse. Hence, the effect of the time at which the
guiding is performed on the guiding properties can in first order be neglected, provided that the laser is guided after the stable situation is established. Figs. 6.3 and 6.6 can be used to assess the minimum time needed for a stable channel to form.

The study on the influence of the current [11] has shown that the influence of the current is small over the range of currents that has been investigated. The current should, however, have a sufficiently high amplitude and long duration to ionize the plasma and allow for it to stabilize, but not so high that wall ablation occurs. We will not explicitly include the effect of the current in our formula of the guiding properties.

The study of the influence of \( n_{H_2}^i \) has shown that the plasma properties depend strongly on the initial density. It was found that \( b \) linearly depends on \( n_{H_2}^i \) via (6.9) over the simulated parameter range.

The study of the influence of \( r_c \) on the plasma properties while the ratio \( r_c/I_0 \) is kept constant has shown that the plasma properties strongly depend on \( r_c \). \( b \) is proportional to \( r_c^{-2.25} \), and can be approximated over the simulated parameter range by (6.11).

By combining (6.9) and (6.11), we can obtain the following relation that describes \( b \) as a function of \( r_c \) and \( n_{H_2}^i \):

\[
b[m^{-5}] = 8.0 \times 10^{-2} (r_c[m])^{-2.25} n_{H_2}^i [m^{-3}]. \tag{6.12}
\]

Substituting the value for \( b \) given by 6.12 in 6.8, we obtain a formula for \( W \):

\[
W[m] = 7.3 \times 10^3 \left( n_{H_2}^i [m^{-3}] \right)^{-0.25} (r_c[m])^{0.5625} \tag{6.13}
\]

This should be compared to the formula for \( W \) as a function of \( r_c, n_{H_2}^i \) and the ionization degree \( z \) as given by Bobrova et al. [12]. Their formula is based on a simplified 1 D model of the steady state of the plasma. Converted to use meters as input and output values, it is given by

\[
W[m] = 4.7 \times 10^3 \left( zn_{H_2}^i [m^{-3}] \right)^{-0.25} (r_c[m])^{0.5}. \tag{6.14}
\]

It is noted that this formula requires knowledge of the ionization degree, which is in principle unknown and difficult to predict; This limits the usefulness of this formula. Assuming \( z \) of order unity, for the range of investigated parameters (6.13) and (6.14) give results that differ less than 15%, which is a good agreement considering the very different methods used to obtain (6.13) and (6.14).

We will assess the validity of (6.13) over the parameter range by comparing \( W \) computed by (6.13) with \( W \) obtained by numerical experiments, consisting of making a parabolic fit of the data points of simulations for selected sets of parameters. The values of \( W \) obtained in this matter are presented in Table 6.1. Equation (6.13) predicts the value of \( W \) obtained with the simulation and parabolic fit with an accuracy better than
Table 6.1: $W_m$ as obtained by making a parabolic fit of the $n_e$ profile obtained by numerical simulation of the channel compared to the values obtained using (6.13).

<table>
<thead>
<tr>
<th>$n_H^2$ ($10^{24}$ m$^{-3}$)</th>
<th>$r_c$ (µm)</th>
<th>$I_0$ (A)</th>
<th>Fitted $W$ (µm)</th>
<th>Eq. (6.13) $W$ (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0</td>
<td>200</td>
<td>400</td>
<td>62.9</td>
<td>60.6</td>
</tr>
<tr>
<td>1.5</td>
<td>175</td>
<td>350</td>
<td>50.5</td>
<td>50.8</td>
</tr>
<tr>
<td>2.0</td>
<td>100</td>
<td>200</td>
<td>34.2</td>
<td>34.5</td>
</tr>
<tr>
<td>3.0</td>
<td>125</td>
<td>250</td>
<td>35.5</td>
<td>35.4</td>
</tr>
<tr>
<td>3.0</td>
<td>75</td>
<td>150</td>
<td>26.5</td>
<td>26.4</td>
</tr>
<tr>
<td>3.0</td>
<td>75</td>
<td>120</td>
<td>27.0</td>
<td>26.4</td>
</tr>
<tr>
<td>3.0</td>
<td>125</td>
<td>250</td>
<td>35.1</td>
<td>35.3</td>
</tr>
</tbody>
</table>

1% for the cases in which the ratio between $I_0$ and $r_c$ is kept at its default value. However, even if this is not the case, the agreement is still excellent, as the sixth experiment in Table 6.1 shows.

Furthermore, (6.13) is validated by comparing it with experimentally determined matched spot sizes for capillary discharge waveguides. In order to make this comparison as accurate as possible, we correct the initial density for hydrogen desorption from the wall, as described in [11]. This desorption is estimated to increase the initial hydrogen content by 28%. Spence et al. [5] report a spot size of 37.5 µm for a channel with $r_c=150$ µm and $p=67$ mbar, while (6.13) gives 43 µm.

Based on this evidence, the close agreement between the formula presented in [12] and the fact that the model used to derive the formula is validated with experimental results [11], we expect (6.13) to give a reasonably accurate estimate of $W$, provided the current pulse is sufficient to ionize the plasma, but not so intense that wall ablation occurs, and provided the assumptions in Sec. 6.3.4 hold.

It is worth noting that the less than linear dependence of $W$ on $r_c$ means that for small $r_c$ the matched spot size becomes bigger than the area over which $n_e$ is approximately parabolic or even bigger than $r_c$. Equation (6.13) is no longer reliable in this case, and, indeed, good guiding may not be possible.

### 6.7 Conclusion

A parameter study of the pulsed capillary discharge is performed by evaluating simulation results for varying values of two parameters: the density and the channel radius. We have verified that the assumptions used to derive the model are valid over the parameter range used, and an analysis of the dependence of the guiding properties on
these parameters is made.

Generally, the plasma structure remained the same over the range of parameters investigated. In all cases, a concave profile is formed, that can be approximated well with a parabolic form over a width significantly larger than the matched spot size. Hence, guiding of a Gaussian laser beam is possible.

By varying the initial density, it was found that $b$ is proportional to the initial density. Furthermore, the formation time of the channel increases for increasing density. This means that for higher densities, it may be necessary to use current pulses that are longer or have a higher amplitude to cause full ionization and the formation of the channel.

We have studied the influence of varying the channel radius on the plasma properties. By considering the heat transport in the steady-state plasma, it was determined that in order to keep the temperature of the plasma in a desirable range, the current should be varied proportionally with the radius. The parameter study of the radius, in which the current was scaled in this way, revealed that $b$ is proportional to $r_c^{-2.25}$.

The results of the parameter studies have been combined to find a general formula for the matched spot size of the channel, given by (6.13). Unlike previous formulae [12], no prior knowledge of the ionization degree is needed. Equation (6.13) has been verified by comparing it with additional numerical experiments, and the agreement was found to be excellent. There was also good agreement between the predicted results and measurements for experimental settings. Hence, (6.13) is expected to give a good prediction of the matched spot size, provided that no magnetization or wall ablation occurs, diffusion is limited, and the current pulse is sufficient in amplitude and duration for a well-ionised plasma to form.

**Acknowledgments**

The PLASIMO team members, in particular Bart Hartgers and Jan van Dijk, are acknowledged for their contributions to the code and the fruitful discussions. This work was performed as part of the research program of ‘Stichting voor Fundamenteel Onderzoek der Materie’ (FOM) with financial support from NWO, within FOM program 55 on Laser Wakefield Accelerators.

**Bibliography**


Study of a pulsed capillary discharge waveguide with a modulated radius

Abstract—Slow pulsed capillary discharges are under investigation for use as plasma channel waveguides in laser-wakefield acceleration and high-harmonics generation. The channel radius has a strong influence on the plasma and guiding properties. Hence, it is expected that the guiding properties can be manipulated by locally modifying the radius. This presumption has been investigated by means of a numerical simulation of a channel with a periodically modulated radius. The simulation revealed a strongly nonlinear response of the plasma and wall properties to the modulation. However, no modulation of the laser guiding properties is observed.

7.1 Introduction

The interaction between compact, high-powered femtosecond lasers [1] and plasmas gives rise to many interesting physical phenomena, such as high-harmonic generation [2, 3], X-ray lasing [4–7] and laser-wakefield acceleration [8–10]. In all these cases, the interaction length between laser and plasma is of critical importance. This interaction length can be increased to many times the Rayleigh length by means of optical laser guiding [11, 12]. This can be achieved by using a plasma waveguide with a convex, parabolic refractive index pattern.

Such waveguides can be generated in wall-ablating capillary discharges[13–17], in pinch plasmas [18–20], and in slow capillary discharges [21–24]. Our investigation focuses on the latter, which have the advantages of a long lifetime and stability.
A slow capillary discharge waveguide consists of a straight capillary that is made of a material that is resistant to short, intense thermal loads, such as alumina or quartz. This capillary is filled with a gas that is easily fully ionized, such as helium or hydrogen, with a pressure that is typically between $10^2$ and $10^4$ Pa. A current pulse of a few hundred ampere that lasts for a few hundred nanosecond is then sent through the discharge. This current pulse causes nearly full ionization of the plasma. However, mainly due to the short duration, ablation of wall material may be avoided, which allows the waveguide to be used many times.

The pulsed capillary discharge waveguide has been analyzed both experimentally [22, 23] and numerically [21, 24–26]. In this work, we focus on an expansion of this analysis by numerically modeling capillary discharge waveguides with a periodically modulated radius. This is of great interest for the following reasons:

- A real waveguide is not perfectly straight and smooth. By investigating the effect of wall irregularities, insight can be gained in the robustness of the laser guiding and plasma properties with respect to these irregularities.

- A periodic variation of the radius might impose a periodic variation in the plasma properties density. It is known that for a straight channel, the matched laser spot size and hence the intensity depends strongly on the channel radius [21, 27]. Hence, it might be expected that a periodic variation of the channel radius can be used to modulate the laser intensity along the axis of the channel. This approach might have potential applications in high-harmonic generation, where a strong enhancement of the conversion efficiency is observed when waveguides with a modulated radius are used instead of straight waveguides [28].

- Using a tapered channel [29] might lead to a variation in axial density. This can be used to keep the laser and electron bunch in phase over a longer distance in laser-wakefield acceleration. Modeling channels with a varying radius will be useful to predict the properties of tapered channels.

- By applying a longitudinal structure, the problem is no longer essentially 1D. This allows for an analysis of the relative importance of longitudinal and radial transport in the plasma.

Discharge capillaries with a well-defined periodic structure are much more difficult to manufacture than straight capillaries. Hence, numerical modeling, especially using a method that is experimentally validated [24], is particularly suitable for an investigation of the conditions required to generate modulated plasma waveguides.

In Section 7.2 this paper, we start by pointing out the requirements for laser guiding in a plasma channel. The discharge capillaries in which the waveguides are generated
will be described in detail in Section 7.3. The model used to simulate the plasma waveguides is discussed in Section 7.4. The results obtained from the numerical simulations are presented in Section 7.5, with an emphasis on the laser guiding properties of the waveguide and the heating of the capillary wall. Conclusions are presented in Section 7.6.

### 7.2 Laser guiding

For the guiding of laser beams, a convex index of refraction pattern is needed [30]. A suitable profile for the guiding of Gaussian laser beams is formed by a concave, parabolic electron density $n_e$ profile, such as given by:

$$n_e(r) = a + \frac{1}{2}br^2,$$  \hspace{1cm} (7.1)

where $a$ is the on-axis density, $b$ the second derivative of $n_e$ and $r$ is the radial coordinate. The matched spot $W_m$ for a laser in this density profile is given by [21]

$$W_m = (0.5\pi r_e b)^{-1/4},$$  \hspace{1cm} (7.2)

with $r_e$ the classical electron radius, which is given by

$$r_e = \frac{e^2}{4\pi\varepsilon_0 m_e c^2},$$  \hspace{1cm} (7.3)

where $e$ is the charge of the electron, $\varepsilon_0$ the permittivity of vacuum, $m_e$ the electron mass and $c$ the speed of light. Eq. (7.3) shows that $r_e$ has a numerical value of $2.817 \times 10^{-15}$ m. The matched spot size of a simulated plasma channel can be obtained by fitting the central electron density profile with Eq. (7.1) and substituting the obtained $b$-value in Eq. (7.2). The electron density profile needs to be concave and (approximately) parabolic only near the center of the discharge, for a radius of about 1.5 $W_m$, for the bulk of the laser power to be transmitted as a Gaussian pulse.

### 7.3 The discharge capillary

For this study, we remain as close as possible to an actually existing system, namely the capillary waveguide described by Spence and Hooker [31]. This waveguide has been the subject of earlier numerical studies [21, 24], making it a good starting point for this study.

This waveguide is generated in a cylinder-symmetric alumina capillary with a radius $r_0$ of 150 $\mu$m, which is filled with 67 mbar of hydrogen and subjected to one half-cycle of a sinusoidal current pulse with a half-time of 200 ns and an amplitude of 300 A.
The discharge capillary with a modulated inner radius. The modelled system is indicated in bold, and is assumed to be part of an infinitely long capillary. The parameter $\delta_w$ is the amplitude of the radius modulation, while the average capillary radius is 150 $\mu$m. The periodicity of the modulation is 100 $\mu$m.

The plasma thus produced will heat the wall, liberating adsorbed hydrogen. This contribution is estimated in [24] and brings the total hydrogen density to $2.16 \times 10^{24}$ m$^{-3}$.

In this study, the capillary is modified by periodically varying the radius $r_c$ with a modulation amplitude $\delta_w$:

$$r_c = r_0 + \delta_w \sin \left( \frac{2\pi z}{z_{\text{mod}}} \right),$$  \hspace{1cm} (7.4)

where $z$ is the axial coordinate. In this study, we restrict ourselves to a modulation period of $z_{\text{mod}}$ of 100 $\mu$m, which is of the same order of magnitude as the channel radius. Modulation lengths that are of the same magnitude as or longer than the capillary radius have also been simulated, but were found to preclude the formation of a stable channel, because the longitudinal bulk transport time becomes comparable to the current pulse length.

The system as it is modeled is shown in Figure 7.1. Owing to the periodicity of the system, only one half of one modulation period needs to be treated. The system will be treated for $\delta_w = 25 \mu$m, which is a significant amplitude resulting in a modulation in plasma area of almost a factor 2, and $\delta_w = 5 \mu$m, which is a much smaller amplitude that is more representative for surface roughness. For relevant cases, these results will be compared to the results for the null case $\delta_w = 0 \mu$m, which are given in [24].

### 7.4 Model of the plasma waveguide

The discharge capillary waveguide has been modeled in [24] using the PLASIMO code [32–37], with a good match between theoretical and experimental results. We use this
model as a basis for the modeling in this work. Because the model is described in detail in [24], the discussion will be restricted to a brief summary and a discussion of the new aspects of the current model that is used to treat the periodic modulation of the radius; the interested reader is referred to [24] for details.

The pulsed capillary discharge waveguide basically consists of a plasma contained in a capillary. For an accurate treatment, two different regions have to be simulated: the plasma, and the temperature evolution of the wall material. These two aspects are modeled separately, and coupled using the boundary conditions.

The basis of the plasma model is formed by a time-dependent two-dimensional non-LTE quasi-neutral fluid model, which is used extensively in the description of low-temperature plasmas and has a wide range of validity. In this approach, it is assumed that the electrons have a temperature $T_e$ and the heavy particles have a temperature $T_h$, which may differ. The Navier-Stokes equations are solved to describe the bulk flow, which reaches significant Mach numbers. The non-LTE approach allows for deviations from chemical equilibrium, and the densities of four separate species, namely $e$, $H^+$, $H$ and $H_2$, are modeled. These differences from chemical equilibrium were found to be important in the formation of the channel [24]. It is noted that the influence of molecular ions [38, 39] is negligible due to the high electron density, which leads to rapid dissociative recombination.

The difference with the earlier simulations [24–26] is the non-uniform channel radius. In order to model the longitudinal variation of the radius of the capillary we have made use of the fact that the discretization code in the PLASIMO framework has been expressed in general ortho-curvilinear (OCL) coordinates. Such coordinates allow for a boundary-fitted grid, as suggested in Figure 7.2(c). The advantages with respect to the “slicing” method in Figure 7.2(b) are twofold:

- The boundary conditions can easily be specified in terms of the single coordinate perpendicular to the boundary.
- Since all grid points are part of the region of interest, this region can be mapped to a structured (orthonormal) computational mesh.

The mapping between the computational and physical coordinate intervals are given by a metric tensor which—in view of the (local) orthogonality—contains diagonal elements only.

The implementation of the grid generation module in PLASIMO is based on the method of elliptic partial differential equations as discussed by Mobley and Stewart [40]. An advantage of this method is that the grid does not need to be isometric, as in the earlier work of Pope [41]: by specifying appropriate stretch functions the grid line density in each of the coordinate directions can be tuned. As a result one can increase
the grid line density in regions where large gradients are expected. The implementation of this method in PLASIMO is described in detail in [32, 33].

For the wall, the temperature equation is numerically solved on a two-dimensional grid that describes the innermost 5 μm of the wall. The temperature-dependent thermal conductivity [42–44] and heat capacity [42, 45] of polycrystalline alumina are used. The melting and ablation of alumina, which has a melting point of about 2300 K [42], is not included in the model. Coupling of the plasma-wall interface is achieved by matching the $T_h$ in the plasma and wall temperature $T_{\text{wall}}$ while matching the heat flux out of the plasma with the heat flux into the wall. This implies that the electron temperature $T_e$ is not coupled to the wall temperature. The coupling is underrelaxed to improve stability. Note that the very shallow penetration of the wall heating, much less than the 5 μm that is simulated, requires a very fine grid to describe correctly. Hence, solving both the wall and the plasma on the same grid will lead to unacceptably large discretization errors or
Figure 7.3: The electron density in the pulsed capillary discharge at \( t = 15 \text{ ns} \). In the narrowest part the electron density is almost a factor of 2 higher than the electron density in the broadest part.

excessive computation times.

7.5 Results and discussion

The pulsed capillary discharge waveguides with a periodically modulated radius has been modeled for two values of \( \delta_w \), namely \( \delta_w = 25 \mu\text{m} \) and \( \delta_w = 5 \mu\text{m} \). First, the formation of a stable channel is discussed. Next, the laser guiding properties at the peak of the current \(( t = 100 \text{ ns})\) are discussed based on the \( n_e \)-profiles. Finally, the heating of the wall is discussed.

Initially, the plasma has a low degree of ionization and is at rest. The current passing through the plasma heats the electrons, causing ionization. The Ohmic dissipation varies with \( r_c^{-4} \) in first order, as it is proportional to the current density \( J^2 \), while \( J \) is proportional to \( r_c^{-2} \) \([25, 26]\). Hence, the narrow parts of the channel ionize much faster than the broader parts, as can be seen in Fig. 7.3, which shows \( n_e \) as a function of \( z \) and \( r \) at \( t = 15 \text{ ns} \), for the channel with \( \delta_w = 25 \mu\text{m} \).

The pressure near the axis of the discharge increases due to heating, in particular in the narrow parts, as is shown in Figure 7.3. This pushes the plasma to the broader, colder parts of the channel, especially to the cooler walls. This creates a concave plasma bulk density profile. The plasma temperature becomes so high that the plasma is nearly fully ionized, except near the walls, resulting in a concave electron density profile. The high electron density and resulting frequent electron/heavy particle collisions make the electron temperature \( T_e \) and the heavy particle temperature \( T_h \) nearly equal over the
Figure 7.4: The electron density in the pulsed capillary discharge at t=100 ns, for various values of δ_w at the narrowest (z=0 µm) and broadest (z=50 µm) part of the channel. Note that the ordinate has a restricted range, in order to emphasize the central part of the discharge. Some data points near the wall are therefore not displayed. The central density profile is almost independent of z, but does depend on δ_w.

central part of the discharge [24]. The formation of such a stable profile takes about 70 ns. For the values of δ_w under study, the electron density profiles at the narrowest and at the broadest part of the discharge, evaluated at t = 100 ns, are presented in Figure 7.4.

Figure 7.4 clearly shows two trends. Firstly, there is almost no difference in the central electron density profile between the narrowest and the broadest part of the discharge. Because the matched spot size W_m depends solely on the central electron density profile (cf. (7.1) and (7.2)), the laser beam will not be modulated by the channel wall modulation. Because it is known that the guiding of the laser beam depends strongly on channel radius for unmodulated channels [21, 25, 26], this is an unexpected result that deserves closer inspection.

As mentioned, the central electron density profile is formed by temperature differences in the channel, with the hottest parts of the plasma having the lowest density. For δ_w = 25 µm and t = 100 ns, T_e as a function of r and z is depicted in Figure 7.5. The electron temperature in the center of the channel is high. This means that the thermal conductivity \( \lambda_e \), which scales as \( \lambda_e \propto T_e^2 \), is much higher in the center of the channel than near the walls. Hence, axial heat transport becomes much faster than radial heat transport. The fast axial transport lead to a central temperature profile that is nearly
Figure 7.5: The electron temperature in the pulsed capillary discharge for $\delta_w$ at $t=100$ ns. The temperature in the central part of the discharge is almost independent of $z$.

independent of $z$; hence, the central electron density profile is nearly independent of $z$ as well.

The second trend that is visible in Figure 7.4 is that the central density is lower for channels that have a higher $\delta_w$. When $r_c$ is modulated, the broader parts of the channel are further from the central channel, leading to lower temperatures. These low-temperature areas acts as sinks for hydrogen, particularly because the temperature becomes so low in these areas that the plasma only has a modest degree of ionization.

In Figure 7.4, fits of the central electron density to a parabola (cf. Eq. (7.4)) are presented. The resulting $a$ and $b$, and $W_m$ obtained using Eq. (7.2) are presented in Table 7.1. For all the cases, the area over which the channel is approximately parabolic is significantly larger than the matched spot size, permitting the guiding of Gaussian laser pulses.

For $\delta_w = 25 \mu \text{m}$, the matched spot size is larger than for the other cases. This is mainly due to lower plasma density in the center of the channel. The lower density reduces $b$, reducing the focusing power of the plasma and increasing the matched spot size.

Having investigated the influence that the radius modulation has on the plasma and guiding properties, we will now consider the influence it has on the capillary wall. The wall is heated by the plasma, and the highest temperatures during the discharge are reached at the plasma-wall interface. The temperature at the plasma-wall interface $T_{pw}$ as a function of $z$ and $t$ is displayed for the channel with $\delta_w = 5 \mu \text{m}$ in Figure 7.6.

Figure 7.6 shows that the temperature distribution becomes strongly peaked as the discharge progresses in time, with the highest temperatures at the narrowest part of the
Table 7.1: The $a$ and $b$ parameters and the matched spot size $W_m$ for three capillaries, at different axial positions.

<table>
<thead>
<tr>
<th>$\delta_w$ ($\mu$m)</th>
<th>$z$ ($\mu$m)</th>
<th>$a$ ($10^{24}$ m$^{-3}$)</th>
<th>$b$ ($10^3$ m$^{-5}$)</th>
<th>$W_m$ ($\mu$m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
<td>1.87</td>
<td>6.86</td>
<td>42.6</td>
</tr>
<tr>
<td>5</td>
<td>0</td>
<td>1.81</td>
<td>6.79</td>
<td>42.7</td>
</tr>
<tr>
<td>5</td>
<td>50</td>
<td>1.81</td>
<td>6.83</td>
<td>42.7</td>
</tr>
<tr>
<td>25</td>
<td>0</td>
<td>1.46</td>
<td>4.4</td>
<td>47.6</td>
</tr>
<tr>
<td>25</td>
<td>50</td>
<td>1.47</td>
<td>4.3</td>
<td>47.8</td>
</tr>
</tbody>
</table>

Figure 7.6: The temperature at the plasma/wall interface, for $\delta_w = 5 \mu$m, as a function of $z$ and $t$. The temperature starts out relatively uniform, but becomes strongly peaked near the narrow part of the channel ($z = 0$) as the discharge progresses in time. The temperature slightly exceeds the melting temperature of alumina (2300 K [42]) at this peak.

discharge, even for a small modulation of the channel radius. The dissipation is higher in the narrower parts of the channel. However, because the radius modulation is only 3%, this effect is too small to fully account for the observed peaking. There are two other effects that cause the temperature to peak.

After about 45 ns, the plasma is somewhat hotter in the narrow parts of the discharge. This leads to an increase in thermal conductivity, and increases the heat flux to the wall. The good thermal contact between this plasma area at the wall and the hotter central plasma allows for most of the heat to flow through this plasma area. The high thermal conductivity of the hot central plasma allows for fast axial heat transport. This allows the heat flux to bypass the cooler plasma areas, where the plasma cannot sustain
a large heat flux to the wall.

The transport properties of the wall respond to a change in temperature as well. In particular, the thermal conductivity of a solid is inversely proportional to its temperature [43, 44]. This causes the wall to lose its heat more slowly when it gets hotter, thus amplifying temperature differences.

The wall reaches a temperature that is above the melting point of alumina (Al$_2$O$_3$). This may lead to erosion of the narrow parts of the channel. Furthermore, the plasma might be contaminated with wall material. Because aluminum and oxygen are not fully ionized at the temperatures that are reached in the plasma, this may lead to undesired ionization induced defocusing [46]. Because the effect of ablation is not included in the model, the quantitative significance of the results is reduced from the moment ablation first occurs (for $t > 128$ ns).

For $\delta_w = 25 \mu$m, the behavior of the wall temperature is more complex, as can be seen in Figure 7.7. The wall is initially hottest at the narrowest part of the discharge, for reasons explained above. After this, the hottest part of the wall moves along the surface.

As mentioned in Section 7.4, the electrons do not transfer their heat directly to the wall, but rather transfer their heat to the heavy particles by elastic collisions, and these heavy particles transfer their heat to the wall. The elastic energy transfer rate $P_{\text{elEH}}$ between electrons and heavy particles can be approximated well by the transfer rate for a fully ionized plasma $P_{\text{fielEH}}$ for the times and regions of interest. $P_{\text{elEH}}$ is given by the product of the energy transferred per collision $E_{\text{coll}}$ and the collision frequency $\nu_{\text{el}}$ (7.5) [47]:

$$P_{\text{elEH}} \approx P_{\text{fielEH}} = E_{\text{coll}}\nu_{\text{el}}. \quad (7.5)$$

Here, $E_{\text{coll}}$ is given by

$$E_{\text{coll}} = 2\frac{m_e}{m_{H^+}} \left( k_B (T_e - T_h) \right). \quad (7.6)$$

Here, $m_{H^+}$ is the H$^+$ mass, and $k_B$ is Boltzmann’s constant. $\nu_{\text{el}}$ is given by

$$\nu_{\text{el}} = n_e n_{H^+} \frac{4\sqrt{2\pi}}{3} \left( \frac{m_e}{k_B T_e} \right)^{\frac{3}{2}} \left( \frac{e^2}{4\pi \varepsilon_0 m_e} \right) \ln \Lambda_C. \quad (7.7)$$

Here, $n_{H^+}$ is the H$^+$ density and $\ln \Lambda_C$ is the Coulomb logarithm, which is about 3 for this plasma. Eq. (7.5) predicts a maximum in the electron energy transfer rate as a function of $T_e$, corresponding to the highest local heating of the wall. As can be seen in Figure 7.5, the electron temperature near the wall is highest in the narrowest part of the channel, and lowest near the broadest part. The highest $P_{\text{elEH}}$ is reached roughly at $z = 25 \mu$m for $t = 100$ ns, as can be seen in Figure 7.8.

Comparing the electron temperature $T_e$ at the wall in Figure 7.5 and the heavy particle temperature $T_h$ at the wall in Figure 7.7 shows that $T_e$ is much higher than $T_h$ in
the narrow part of the channel. This difference can be ascribed to the decrease of $P_{\text{el EH}}$ with increasing $T_e$, allowing for a local runaway of $T_e$. This clearly underscores the importance of a non-LTE treatment.

We see that the interaction between plasma and wall is complex and highly nonlinear, with both the temperature dependence of the wall properties and the deviations from thermal equilibrium playing an important role. However, the two-region model with the underrelaxed coupling, as described in Section 7.3, was sufficiently robust to handle the nonlinearities and very different length and time scales of the transport in the plasma and the wall, in a computational time that was quite manageable (several days at most).

### 7.6 Conclusions

We have numerically modeled a pulsed capillary discharge waveguide with a sinusoidally modulated radius, using a two-region model with an underrelaxed coupling to efficiently treat the very different length- and timescales of the transport in the capillary and in the plasma. Two general trends were observed.

Firstly, while the modulation does influence the plasma near the wall, the central plasma and hence the matched spot size is not significantly modulated, due to the fast axial heat transport, for the modulation depths under study. This means that the laser beam cannot be modulated by modulating the channel radius for the modulation depths
under study, and that modulation by surface roughness has no significant influence on the laser propagation either.

Secondly, the radius modulation causes a strongly nonuniform wall temperature, and temperatures above the melting temperature of alumina are reached. For a channel with a 5 \( \mu \text{m} \) radius modulation, the wall temperature is highest at the narrowest part of the channel. Ablation there will widen the channel and hence reduce the modulation. In practice, this means surface roughness may be ablated by the discharge, smoothening the channel. For a channel with a 25 \( \mu \text{m} \) radius modulation, the hottest part of the wall moves over the surface in time. In this case, significant ablation is expected.

From this, it is concluded that surface roughness does not influence the performance of the plasma waveguide. Furthermore, wall radius modulation is not expected to be a useful technique for tailoring the laser guiding properties of the waveguide.

**Acknowledgments**

The authors would like to thank A.J. Gonsalves for the fruitful discussions. The PLASIMO team members, current and former, are acknowledged for their contributions to the code. This work was performed as part of the research program of ‘Stichting voor Fundamenteel Onderzoek der Materie’ (FOM) with financial support from NWO, within FOM program 55 on Laser Wakefield Accelerators.
Bibliography


Study of a pulsed capillary discharge waveguide with a modulated radius
Chapter 8

Modeling of a square pulsed capillary discharge waveguide for interferometry measurements

Abstract—Slow pulsed capillary discharges in round capillaries are currently under investigation for use as plasma channel laser waveguides in laser-wakefield acceleration, X-ray lasers, and higher-harmonic generation. In this study, a capillary discharge with a square cross section is presented. The electron density, which determines the laser guiding properties, can be measured by means of transverse interferometry in this device. Using a numerical model of the plasma and the capillary wall, an analysis of the discharge is made. The results predict that the square channel is capable of guiding circular laser pulses. The guiding properties are quite similar to those of a round channel with nearly the same diameter as the channel width. This suggests the results obtained by measuring the square capillary discharge are applicable for round channels as well. It was found that the wall heating was inhomogeneous, which makes the wall more susceptible to ablation, although this was not an issue for the system treated. The heating of the wall changes the transverse optical pathlength in the interferometry experiments, but not to such an extent that it significantly impacts the results.

Parts of this chapter are to be submitted for publication, in slightly altered form, by the authors B. H. P. Broks, W. van Dijk, A. J. Gonsalves, T. P. Rowlands-Rees, S. M. Hooker and J. J. A. M. van der Mullen to *Physics of Plasmas*, and by the authors A. J. Gonsalves, T. P. Rowlands-Rees, B. H. P. Broks, J. J. A. M. van der Mullen and S. M. Hooker to *Physical Review Letters*
8.1 Introduction

The interaction length between plasmas and high-powered lasers can be increased by optical guiding. Examples of the application of this interaction include high-harmonics generation [1, 2], X-ray lasers [3–8] and laser-wakefield acceleration [7, 9–12]. This can be achieved by the use of plasma waveguides.

Plasma waveguides operate by creating an index of refraction pattern which is highest in the center focusing the laser beam and counteracting diffraction. This is achieved by creating a plasma with a concave density profile. To counteract ionization-induced refractive defocusing [13], a fully ionized plasma is desirable.

A few different types of such waveguides exist, such as discharge-ablated waveguides [14–18], pinch plasmas [8, 19, 20], and slow capillary discharges [7, 21–24]. Our investigation focuses on the latter. This waveguide has the advantages of long device lifetime and of a long, stable guiding window.

Because the guiding properties depend directly on the electron density, accurate measurements of the electron density are very desirable. One method to directly measure the electron density in the plasma is via interferometry measurements. The electron density influences the index of refraction of the plasma and hence the optical path-length through the plasma, and can be measured by measuring this change in optical pathlength. This procedure has been successfully applied to round channels [21, 25] by measuring integrated along the axis. This method, however, is influenced by plasma efflux from the ends of the capillary. Furthermore, the signal is an integration of a large plasma volume, which might blur features. These problems can be avoided by performing the interferometry perpendicular to the capillary wall.

This is, however, not possible in the standard capillary plasmas, which use round alumina (polycrystalline Al₂O₃) walls. For the interferometry, optically flat sapphire (monocrystalline Al₂O₃) walls are required. The fact that the walls are flat does mean the channel is no longer round, which makes a direct comparison with round channels less straightforward.

In this work, a theoretical analysis of a square pulsed capillary discharge waveguide is made. While the model presented is valid for a wide range of external parameters, the discussion is focused on one particular configuration, which matches an experimental configuration, facilitating a comparison.

The square capillaries require a two-dimensional time-dependent non-LTE model, which is considerably more challenging computationally than the one-dimensional model used earlier [24, 26, 27]. As an additional advantage, successfully modeling the two-dimensional square capillaries can act as a proof-of-principle for modeling waveguides with other cross sections, such as elliptic or rectangular waveguides.

By modeling the plasma properties of the channel, we can provide a theoretical back-
8.2 The system

The system consists of a square, hydrogen-filled capillary, with electrodes that are connected to a high-voltage power supply at either end. By applying a current pulse, a transient hydrogen plasma is created in the capillary.

The capillary consists of two larger sapphire plates, with two smaller sapphire plates sandwiched between them in such a way a square space is left open. It is schematically drawn in Figure 8.1. Hydrogen is inserted into the capillary using gas feeds.

For the experiments [28], capillaries of various sizes are used; in this study, a capillary with a width of 465 µm is simulated. The initial hydrogen pressure and current are also variable; in this model, an initial hydrogen pressure of 4000 Pa is used, and the current \( I(t) \) is given by (8.1):

\[
I(t) = 600 \sin \left( \frac{\pi t}{230 \text{ ns}} \right) \text{ A}, \quad 0 \text{ ns} < t < 230 \text{ ns},
\]  

(8.1)

with \( t \) the time after the beginning of a current pulse. This closely matches an actual experimental setting, allowing a direct comparison between experimental and theoretical
results.

8.3 The model

The model used to simulate the square waveguide is based on the model used to simulate the round waveguide. The latter model has been discussed in great detail in [24]. It has been validated by comparison with an experiment [24], used for parameter studies [24, 26, 27], and a slightly modified version has been used to investigate the effect of modulation of the capillary radius [29]. We will restrict the discussion of the model used in this work to a brief summary, emphasizing the difference of the present model and the model used for earlier studies, and refer the interested reader to [24] for details.

The capillary discharge waveguide consists of two separate parts: a plasma and a capillary wall. These two parts are simulated with different models and coupled via boundary conditions, allowing for a self-consistent solution of the whole system.

The plasma model is a two-dimensional two-temperature non-LTE fluid model. In this model, four species are tracked: H, H₂, H⁺ and e. The powerful non-LTE approach allows for both deviations from thermal equilibrium, which are most significant during the startup of the discharge and near the walls, and of chemical equilibrium, which are most significant during the startup of the discharge. This approach, combined with a detailed description of the chemical reactions and transport coefficients, allows for an accurate description of all stages of the discharge. In particular, the regions near the wall, where a large fraction of the hydrogen is located [24, 26, 27, 29] and where significant deviations from LTE occur, can be simulated accurately using this model.

The model of the wall consists of a one dimensional description of the heat transport
into the sapphire wall for each point that is on the plasma/wall interface, allowing for a detailed description of the spatial and temporal variations of the wall temperature. Because the plasma and wall model are coupled via the boundary conditions, the wall temperature in turn affects the plasma behavior.

The sapphire wall is heated by the heavy particles in the plasma. This heating is computed by means of a one-dimensional heat transport equation of the sapphire wall for each point that is on the plasma-wall interface:

$$\frac{\partial C_{wall}T_{wall}}{\partial t} - \nabla \cdot (\lambda_{wall} \nabla T_{wall}) = 0, \quad (8.2)$$

where $C_{wall}$ is the heat capacity of the wall material per cubic meter, $T_{wall}$ is the wall temperature and $\lambda_{wall}$ is the thermal conductivity.

The transport properties of the sapphire wall of the channel currently under study are different than the transport properties of the alumina walls of the capillaries described in [24, 26, 27, 29]. There is no significant difference in $C_{wall}$, which is given for both alumina and sapphire walls by [24, 30, 31]:

$$C_{w} = 3n_{wall}k_{B} \left[ \left( \frac{\theta_{E}}{T_{wall}} \right) \frac{\exp \left( \frac{\theta_{E}}{2T_{wall}} \right)}{\exp \left( \frac{\theta_{E}}{T} \right) - 1} \right]^{2}, \quad (8.3)$$

with $n_{wall}=1.17 \times 10^{29}$ m$^{-3}$ the density of Al$_2$O$_3$ in particles per cubic meter and $\theta_{E}=690$ K the Einstein temperature. However, the heat conductivity of sapphire is slightly larger than that of alumina, and is approximated well by [32–35]:

$$\lambda_{wall} = \frac{9.9 \times 10^{3} K}{T_{wall}} \text{Wm}^{-1}\text{K}^{-1} \quad (8.4)$$

for the temperature range of interest.

Wall heating changes the optical pathlength through the system, which influences the measurements. There are two contributions to the increase in optical pathlength, namely the thermal expansion of the sapphire and the increase in refractive index of the sapphire with increasing temperature. The thermal expansion coefficient of sapphire has been obtained from [36] and is equal to $5.9 \times 10^{-6} \text{K}^{-1}$. The temperature-dependent indices of refraction $n$ of sapphire at the laser wavelengths $\lambda$ of 532 nm and 1064 nm that are used for the measurements are not available in literature to the best of our knowledge. Hence, we will estimate the index of refraction from the known index of refraction at room temperature at 532 nm and 1064 nm [31], and adding extrapolated temperature-dependent terms that are measured by Tapping et al. [37] for 633 nm and 799 nm. Because the temperature-dependent terms do not depend strongly on $\lambda$ and because the index of refraction itself is a smooth function of $\lambda$, we expect this extrapolation to be reasonably accurate. The resulting equations for $n_{532}$ and $n_{1064}$ are:
\[ n_{532} = 1.7676 + 1.02 \times 10^{-5} T[K] + 4.7 \times 10^{-9} T[K]^2 \] 
\[ n_{1064} = 1.7496 + 1.10 \times 10^{-5} T[K] + 1.6 \times 10^{-9} T[K]^2. \]

Wall ablation has the potential to damage the optically flat sapphire surfaces. Furthermore, the oxygen and aluminum that ablate contaminate the plasma. Both effects are quite undesirable. By computing the wall temperature \( T_{\text{wall}} \), it can be assessed whether it remains below the melting temperature of sapphire (2326 K) [31] and whether ablation occurs.

Unlike the alumina walls used in the previous studies, the sapphire walls in this case are smooth and not porous. This means far less hydrogen will desorb from them. Currently, we estimate that one hydrogen molecule desorbs from each surface site, which gives a contribution of \( 2.93 \times 10^{23} \text{ m}^{-3} \) at a total initial hydrogen density of \( n_{iH_2} \) of \( 1.205 \times 10^{24} \text{ m}^{-3} \).

The grid at which the simulation is carried out is shown in Figure 8.1. Although the system exhibits an eightfold symmetry, only the fourfold symmetry is exploited to arrive at a two-dimensional non-equidistant Cartesian grid that describes the plasma.

In order to compare the guiding properties of the new square channel with the common round channels, simulations of equivalent round channels are also run. These simulations use the same model that is presented [24], except for the thermal properties of the wall, for which the thermal properties of sapphire are used as described in Eq. (8.4). Two systems are simulated: one with an area that matches the area of the square channel, giving it a radius of 262 \( \mu \text{m} \), and one with a diameter matching the width of the square channel, giving it a radius of 232.5 \( \mu \text{m} \). In both cases, \( n_{iH_2} = 1.205 \times 10^{24} \text{ m}^{-3} \) and \( I(t) \) is given by Eq. (8.1).

For the simulations, we use the PLASIMO code. This code is described in detail in [38–42]. It is a modeling platform that can handle LTE and non-LTE plasmas, currently in two dimensions. Furthermore, its modular structure allows for easy expansion of the code. It has been applied to simulate a wide variety of plasma as described in Refs. [24, 26, 27, 29, 43–49].

### 8.4 Laser guiding theory

An important application of pulsed capillary discharges is the guiding of lasers. For the guiding of a Gaussian beam, a hollow, parabolic profile of the electron density \( n_e \) is needed. Such a profile is given by

\[ n_e(d) = a + \frac{1}{2} bd^2. \]
Here, \( b \) is the second derivative of the electron density, and \( a \) is the on-axis electron density.

The matched spot size \( W_m \) of the laser beam is given by [22]

\[
W_m = (0.5\pi r_e b)^{-1/4}
\]  

(8.8)

with \( r_e \) the classical electron radius which has a numerical value of \( 2.817 \times 10^{-15} \) m. The matched spot size can be obtained by fitting the central electron density with Eq. (8.7) and substituting \( b \) in Eq. (8.8).

The electron density is only approximately parabolic near the center of the discharge. This is not a significant issue provided the area over which \( n_e \) is parabolic is larger than the matched spot size, because such a spot size is sufficient for the bulk of the laser power to be transmitted as a Gaussian pulse.

### 8.5 Results and discussion

The square channel described in Section 8.2 has been simulated using the model described in Section 8.3. Selected results will be presented in this Section. In Section 8.5.1, the evolution of the plasma properties of the channel will be discussed. In Section 8.5.2, key plasma physical parameters at the maximum of the current pulse (\( t = 115 \) ns) will be presented. In Section 8.5.3 the guiding properties will be discussed based on the \( n_e \) profiles and corresponding matched spot sizes in three different chords at \( t = 115 \) ns. In Section 8.5.4, the influence of the plasma on the capillary walls will be discussed. In Section 8.5.5 a comparison with round channels of equivalent radius and area will be made.

#### 8.5.1 Evolution of the channel

The plasma channel is a strongly dynamic system, and as such, the plasma properties, including the matched spot size, vary in time. In this Section, we will discuss the evolution of the plasma using three key plasma properties: the electron density \( n_e \), the electron temperature \( T_e \), and the heavy particle temperature \( T_h \). These plasma properties are evaluated at a chord on the horizontal symmetry plane, for \( 0 \) ns \( \leq t \leq 180 \) ns, and are presented in Fig. 8.2, 8.3 and 8.4, respectively. In these graphs, \( d \) denotes the position on the chord.

Qualitatively, the behavior of the channel is similar to the behavior of the round channels presented in [24]. Hence, we will give only a brief description of the formation mechanism, and refer the interested reader to [24] for a more detailed discussion.
The electron temperature $T_e$ as a function of time and position on a chord in the horizontal symmetry plane. The isolines indicate differences of $0.5 \text{ eV}$.

The electron temperature is initially determined by the balance between Ohmic dissipation and ionization. Because the electron density $n_e$ increases much faster than the dissipation, the temperature drops in time. The plasma properties are homogeneous. The heavy particle temperature starts increasing when the plasma becomes significantly ionized, approaching local thermal equilibrium (LTE) everywhere except near the wall. The transition between these regimes is roughly around $t=50 \text{ ns}$.

The increase in electron and heavy particle temperature allows significant thermal
8.5. Results and discussion

Figure 8.4: The heavy particle temperature $T_h$ as a function of time and position on a chord in the horizontal symmetry plane. The isolines indicate differences of 0.5 eV.

gradients to form. These thermal gradients cause a pressure gradient, which pushes the plasma to the wall, forming a hollow profile in $n_e$, as is seen in Fig. 8.2, between 60 and 100 ns. The central $n_e$ profile is steepest near the peak of the current, at $t=115$ ns, meaning that the matched spot size is smallest at this time (cf. Eq. (8.8)). After the peak of the current, the plasma slowly cools as less power is dissipated.

8.5.2 Results at the current maximum

The plasma in square channels exhibits a two dimensional structure over a cross section of the discharge. We will discuss this structure in $n_e$ and $T_e$ at the peak of the current, $t=115$ ns. The electron density as a function of $x$ and $y$ is shown in Fig. 8.5, while the electron temperature $T_e$ as a function of $x$ and $y$ is shown in Fig. 8.6.

Fig. 8.5 shows that the electron density is lowest in the center and increases toward the wall. In the corner, the $n_e$ is again very low. Centrally, the density profile is close to being circular, like it would be in a round channel, while near the wall, the density is profile is nearly square, the shape of the waveguide. The consequences of this competition for the guiding properties are analyzed in Section 8.4.

The electron density profile is a result of the temperature profile, in particular $T_e$. $T_e$ is so high that the plasma is fully ionized, except near the walls. Because the plasma has a nearly uniform pressure, this results in an electron density profile that is essentially the inverse of the electron temperature. This is no longer the case near the wall, and especially in the corner, as $T_e$ is so low there that the hydrogen is no longer fully ionized.
8.5.3 Analysis of the guiding properties

In order to make a quantitative assessment of the guiding properties of the channel, the matched spot sizes at three different chords through the plasma have been determined. The fit is carried out at three chords: the Horizontal chord on the horizontal symmetry plane, the Intermediate chord that passes through the axis and through the wall at three quarters of the total width of the capillary, and the Diagonal chord on the diagonal symmetry plane, at $t=115 \text{ ns}$. The electron density and fitted parabolas at these chords are presented in Fig. 8.7.

Near the center, the electron density profile at each chord is almost identical. The matched spot size is determined by fitting a parabola like Eq. (8.7) for $d < 100 \mu m$. Fig. 8.7 shows that the fit is excellent in the region of interest. Using the resulting $b$ and Eq. (8.8), we obtain values for the matched spot of $65.0 \mu m$, $65.4 \mu m$ and $66.0 \mu m$ for the Horizontal, Intermediate and Diagonal chord, respectively. Because the matched spot size is much smaller than the fraction of the chord over which we fit, the bulk of the laser power is transmitted through this part of the channel. Hence, we fit over a sufficiently large fraction of the chord.

The difference in matched spot size between the various chords indicates that the
8.5. Results and discussion

Figure 8.6: The electron temperature $T_e$ over a cross section of one quarter of the channel at $t=115\,\text{ns}$. The isolines indicate differences of $0.5\,\text{eV}$.

Laser is transmitted as a nearly circular beam, with matched spot size shape that deviates less than $0.8\%$ from a perfect circle.

8.5.4 Analysis of the walls

Excessive wall temperatures might lead to ablation. Because the plasma heats the wall, the hottest part of the wall is the plasma-wall interface. The temperature of this interface $T_{pw}$ as a function of the position along the wall $w$ and the time has been plotted in Fig. 8.8.

Fig. 8.8 shows that the wall is initially heated almost homogeneously, but as the discharge progresses, the heating becomes ever more inhomogeneous, with the hottest part in the center of the wall. Furthermore, while the plasma temperature peaks near the current peak, the wall temperature continues to grow until about $t = 175\,\text{ns}$. The melting temperature of sapphire is not reached, hence, ablation is not expected for these circumstances.

The wall temperature is almost homogeneous initially because the plasma temperature is as well (cf. Fig. 8.3). However, the plasma heats up primarily in the center, and
Figure 8.7: The simulated $n_e$ and a parabolic fit through the center of the profile, at three different chords. The Horizontal chord lies in the horizontal symmetry plane, the Intermediate chord passes through the axis and through the wall at three quarters of the total width of the capillary and the Diagonal chord lies in the diagonal symmetry plane. The matched spot size at the three chords is also indicated.

this heat is chiefly transported via the shortest path to the wall, heating up the center of the wall. This inhomogeneous heating makes the wall more susceptible to ablation, and places design restrictions or power limitations on capillaries that have a shape different from a straight cylinder that are stricter than for straight, cylindrical capillaries. This effect is strengthened by two self-amplifying mechanisms: one in the plasma and one in the wall.

Throughout the plasma, apart from a small region near the wall, electron thermal conductivity is the dominant heat transport mechanism [24]. This transport mechanism scales with $T_e^2$. Hence, hotter parts of the plasma transport more of the heat from the center to the wall, leading to inhomogeneous heating. [27, 29].

The wall thermal conductivity scales with $T_{wall}^{-1}$, according to Eq. (8.4). This means, that when a part of the wall heats up, heat transport from this hot area is inhibited, leading to further temperature increase.

The heating of the wall induces a change in optical pathlength, both due to thermal expansion and the temperature-dependent index of refraction of the material. The increase in optical pathlength $d_{opt}$, for a wavelength of 532 nm and 1064 nm, as a function of time and position along the wall, is plotted in Figure 8.9 and Figure 8.10, respectively.

Quantitatively, Figures 8.9 and 8.10 show that the increase in optical pathlength is
8.5. Results and discussion

Figure 8.8: The simulated temperature at the plasma-wall interface. The corner is at $w=232.5 \mu m$. The isolines indicate differences of 100 K.

Figure 8.9: The increase in the optical pathlength through the wall due to wall heating, for a wavelength of 532 nm. The corner is at $w=232.5 \mu m$. The isolines indicate differences of $2.0 \times 10^{-9} m$.

small compared to the wavelength of the light used. Hence, the heating of the wall is not expected to greatly influence interferometry measurements. The difference in optical pathlength does not depend strongly on the used wavelength. The thermal expansion and the change in index of refraction make contributions to $d_{opt}$ that are roughly equal. However, unlike $T_{pw}$, $d_{opt}$ rises monotonically, which means that its influence gets more significant as time progresses.
8.5.5 Comparison with a round channel

In order to assess to what extent the plasma and guiding properties in the square capillaries resemble those in round capillaries, we will compare selected results from the square capillaries to the results of two round capillaries with the same current and initial density, and with radii of 262 µm (channel 1) and 232.5 µm (channel 2). The former has the same area as the square capillary, while the latter has diameter equal to the width of the capillary.

The electron density, which is the plasma property that is relevant for the guiding, is presented as a function of time and radial position in Figures 8.11 and 8.12, respectively.

Comparing Figures 8.2, 8.11 and 8.12 shows that the time it takes for the plasma to ionize in the square channel is nearly the same as that in channel 1. In this stage of the discharge, the primary loss mechanism for the dissipated power is ionization, which is a local process. The dissipated power depends on the current density, which is the same in the square channel and channel 1. This means the ionization rate and hence the electron density are similar in channel 1 and in the square channel during the initial stages of the discharge.

The discharge reaches a stable, hollow profile due to heat transport to the wall. The matched spot size of channel 1 and 2 has been determined by fitting (8.7) to the inner 100 µm and using Eq. (8.8). The matched spot size for channel 1 is 69.9 µm, while the matched spot size for channel 2 is 64.9 µm. The matched spot size of the square channels varies between 65.0 and 66.0 µm, depending on orientation. The matched spot size of the square channel lies between the matched spot size of channel 1 and channel 2, but
Figure 8.11: The electron density $n_e$ as a function of time for a round channel with a radius of 262 $\mu$m. The isolines indicate differences of $2.5 \times 10^{23} \, m^{-3}$.

Figure 8.12: The electron density $n_e$ as a function of time for a round channel with a radius of 232.5 $\mu$m. The isolines indicate differences of $2.5 \times 10^{23} \, m^{-3}$.
is much closer to the matched spot size of the latter. This can be explained by noting that during the guiding phase, the heat transport properties shape the channel. These are strongly dependent on the distance that has to be traveled, and because the heat transport is primarily via the path of the least thermal resistance, it is especially the shortest distance from the center to the wall that matters. This distance is identical for channel 2 and the square channel, and hence, the matched spot sizes are expected to be similar.

8.6 Comparison with the experiment

In this Section, the theoretical results will be obtained with experimental results [28]. The direct experimental results that are obtained are phases shifts, that are integrated along a chord through the discharge. To obtain the $n_e$-profile, an assumption about its shape has to be made to convert the one-dimensional integrated results to the full two-dimensional $n_e$-profile. We can avoid making this assumption by not comparing the resulting $n_e$-profiles, but by comparing the measured phase shift with the phase shift that would result from the computed $n_e$-profiles. The resulting theoretical phase shift is compared with experimental phase shifts, measured at two different laser wavelengths, at $t=127$ ns, in Figure 8.13. The figure shows an excellent agreement between the simulated and measured data, in particular in the center of the channel. The match near the walls, where the measurements are far more difficult and less accurate, is somewhat less good.

We have established in Section 8.5.5 that the results of the square channel can be approximated well with the results for a round channel. This means that we can also apply the scaling laws derived by Broks et al. [27] and Bobrova et al. [22].

The scaling laws for the matched spot size of both of these methods are compared to experimental data, obtained on a square channel with a width of 210 $\mu$m, in Figure 8.14. If the measurements are done after the channel has stabilized ($t > 80$) and if the current is sufficient for full ionization (peak current at least 250 A), then these parameters do not strongly influence the density distribution [24, 27]. The time and peak current were within this range for the experimental data obtained for this channel; hence, no large error is made by comparing data in which time and peak current vary somewhat within this range.

In Figure 8.14, it is observed that both the methods in [27] and [22] match well with the experimental data, suggesting either scaling law is suitable for the prediction of $W_m$.

The on-axis electron density $n_e(0)$ also plays a role in the applications, in particular laser-wakefield acceleration [50]. We have compared the measured $n_e(0)$ with predictions based on the work of Bobrova et al. and the scaling of Broks et al. [27], in Figure
8.7 Conclusion

The model presented in [24] has been successfully expanded to be able to describe two-dimensional capillary discharge waveguides with sapphire walls. This not only enables the study of square waveguides, but is also a proof of principle for the study of waveguides with other two-dimensional cross sections.

The square capillary discharge waveguide produces a plasma that has a matched...
Figure 8.14: Measured matched spot size $W_m$ in square plasma channels with a width of 210 $\mu$m as a function of the initial hydrogen density $n_{iH_2}$. The dotted line shows the results obtained by Bobrova et al.[22], while the solid line shows the results by Broks et al.. The dashed line shows a power-law fit to the data [27]. Graph and measurement data are courtesy of A.J. Gonsalves of Oxford University.

spot size of 65.0 $\mu$m on a chord that lies on the horizontal symmetry axis, 65.4 $\mu$m on a chord that goes through the axis of the capillary and through the wall at three quarters of the total height, and 66.0 $\mu$m on a chord that lies on the diagonal symmetry axis. This means a Gaussian laser pulse with a circular shape can be guided with only very little deformation of the circular shape.

Both the evolution and the plasma parameters of the square channel match those of equivalent round channels closely. This means that the square channel is a good model system to measure plasma properties that otherwise cannot be measured in this way in a round channel.

The heating in the capillary wall is strongly inhomogeneous, with the highest temperatures in the center of the capillary wall. The increase in local temperature makes the wall more susceptible to ablation, which damages the wall and contaminates the plasma with species that are not fully ionized. For the setting we have studied, the temperature remained well below the ablation threshold.

The heating of the capillary wall induces a change in optical pathlength, due to the temperature dependence of the index of refraction and due to thermal expansion of the
wall material. For the setting that has been studied, the change in optical pathlength is less than a tenth of a wavelength, and is hence not expected to have a significant influence on interferometry measurements.

Because the plasma and guiding properties of the square capillary discharges closely match those of the round capillary discharges, and because the temperature-induced index of refraction changes in the wall are small, the square capillary discharge is expected to be a suitable system to perform interferometry measurements on to gain insight in the electron density profile and guiding properties of capillary discharge waveguides. Furthermore, owing to the similar plasma and guiding properties, the square capillary discharge waveguide is a suitable for the same applications as a round capillary discharge waveguide. The measured phase shifts have been compared to the phase shifts that would have been generated by a plasma with the simulated electron density profile. This comparison reveals an excellent agreement between simulation and experiments, in particular in the central part of the channel.

The measured match spot size and central densities are compared to empirical scaling laws derived by Broks et al. [27] and Bobrova et al.[22]. It was found that the matched
spot size matches either theory well, but that only the theory of Broks et al. [27] gives a good prediction of the central electron density.

Acknowledgments

The authors would like to thank Walter van Dijk for the fruitful discussions. The PLASIMO team members, current and former, are acknowledged for their contributions to the code. This work was performed as part of the research program of ‘Stichting voor Fundamenteel Onderzoek der Materie’ (FOM) with financial support from NWO, within FOM program 55 on Laser Wakefield Accelerators.

Bibliography


Chapter 9

Theory of a photoconductor switched high-voltage spark gap

Abstract—In this Chapter, a photoconductor switched high-voltage spark gap, with an emphasis on the switching behavior is modeled. It is known experimentally that not all of the voltage that is present at the input of the spark gap is switched, but rather, a fraction of it drops across the spark gap. This voltage drop depends on the voltage that is present at the input of the spark gap with higher voltages resulting in a smaller drop. We have investigated two possible causes of this: the cathode fall and the resistance of the plasma arc. Using an analytical model of the cathode fall, we have established that the cathode fall can be excluded as the cause of the observed voltage drop. A one-dimensional, time-dependent non-local thermal equilibrium fluid model of the arc plasma has been made. Using this model, the plasma properties have been analyzed for various values of the switched current, with emphasis on the conductivity. A good qualitative match between the observed and simulated dissipation in the gap was found. This indicates the finite arc resistance is the cause of the observed voltage drop.

9.1 Introduction

Spark gaps are used for the rapid switching of high voltages and high currents in a wide range of applications. Our interest in spark gaps lies mainly in the construction of compact pulsed DC electron accelerators [1]. With these accelerators, electric fields can be achieved that are an order of magnitude higher than the electric fields that can

This Chapter, in slightly altered form, has been accepted for publication in Journal of Applied Physics by the authors B.H.P. Broks, J. Hendriks, W.J.M. Brok, G.J.H. Brussaard, and J.J.A.M. van der Mullen

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be achieved with conventional radio frequency accelerators. For these compact DC accelerators, high-voltage pulses with a short rise time, short duration and favorably no time jitter are required.

Spark gaps have been used to switch large voltages for over a century. A conventional spark gap consists of two electrodes with a gas-filled gap between them. When a voltage higher than the breakdown voltage is applied across the spark gap, breakdown occurs, and the gas between the electrodes ionizes. An arc plasma with a high degree of ionization and good conductivity forms, closing the spark gap.

The breakdown process occurs by Townsend avalanches and streamers, and requires that the ambient electron density of for instance air, which is typically $10^9 \text{ m}^{-3}$ increases by several orders of magnitude to achieve good conduction. This is an inherently stochastic process, lengthening the rise time and producing time jitter.

By axially focusing a laser into the spark gap, the initial ionization degree of air can be raised by several orders of magnitude, creating a well-defined pre-ionized channel between the electrodes [2]. The higher initial electron density smoothes statistical fluctuations, and the electron density has to rise less to achieve good conductivity. Because of these two effects, the rise time and jitter are shorter, as low as sub-nanosecond, while allowing pulses in the order of 1-100 kV to be transmitted [3].

The use of Ti:sapphire lasers, that can produce terawatt pulses with a duration in the femtosecond range, allows the concept of the laser-triggered spark gap to be taken one step further. By focusing the laser between the electrodes, a plasma can be created with an ionization degree that is so high that no further ionization is necessary to achieve good conduction: in other word, the laser not so much triggers the spark gap, but rather switches it. Recently, we have reported on experiments [4, 5], in which a terawatt laser was used to photoconductively switch voltages of up to 4.5 kV with a jitter better than the resolution of the measuring equipment, 12 ps. Furthermore, this switch is capable of switching voltages as low as 10 % of the self break down voltage.

However, in these experiments it was observed that, in particular for lower voltages, a significant part of the voltage drops across the spark gap due to plasma processes. This voltage drop can be observed in Fig. 9.1, where the input voltage $V_{\text{in}}$ and the output voltage $V_{\text{out}}$ are plotted.

In this Chapter, we will investigate in detail two probable causes of the observed voltage drop, namely the cathode fall and the plasma arc resistance, to determine the cause of the voltage drop over the discharge, using theoretical models. First, in Section 9.2, a description of the setup will be given. In Section 9.3 a simple, analytical model of the cathode fall formation in the laser-produced plasma has been made, and based on this, the relevance of the cathode fall to the switching behavior will be discussed. In Section 9.4, a time-dependent numerical model of the arc plasma in the spark gap is described. Using this model, an analysis of the plasma and the switching behavior is
The setup

The experimental setup has been described in detail in [4, 5]. In this Section, the discussion will be limited to the most essential parts.

The spark gap is schematically drawn in Fig. 9.2. The spark gap is basically a 1 mm interruption in a coaxial transmission line structure, that has a characteristic impedance of 55 Ω. One side of this structure is charged to a high voltage $V_{in}$, in our case 4.5 kV, limited by the self breakdown voltage of the spark gap. The gap is filled with either nitrogen or air at atmospheric pressure. Because there is little experimental difference between the switching behavior of the gases [5], the discussion will be restricted to the case of nitrogen.

The gap is switched using a Ti:Sapphire laser that has a pulse energy of 35 mJ and a pulse duration of 200 fs. The laser is focused into a filament that connects the two electrodes two cylindrical lenses. The intensity over the entire gap is above the intensity threshold for tunnel ionization. The focus has a full width half maximum (FWHM)
Figure 9.2: A schematic drawing of the spark gap switch [4, 5]. The gap is an interruption in a coaxial transmission line structure, and is filled with either nitrogen or air. By focusing a terawatt laser in a line focus in the spark gap, a laser-produced plasma bridges the gap between the electrodes, switching the voltage that is over the gap.

height of $20 \pm 3 \, \mu m$, a length of $949 \pm 6 \, \mu m$ [5] and a width of approximately $100 \pm 3 \, \mu m$. The radius of curvature of the electrodes is 3 mm. This means, that the plasma is a thin filament that connects both electrodes, while it fills only a fraction of the gap between the electrode. The switched pulse has a length of 2 ns and a rise time of less than 100 ps.

As mentioned in Section 9.1, we will model two different aspects of the plasma: the initial cathode fall formation, and the evolution of the arc plasma between the electrodes. The initial plasma composition and size has not been measured, and has been estimated from the size of the laser focus and the power density in the focus. A schematic representation of both model areas is given in Fig. 9.3.

For the modeling of the cathode fall plasma, an analytical model is used, which requires the size of the plasma as an input. The plasma is approximated by a homogeneous slab, with a size determined by the FWHM of the laser focus: $l = 1 \, mm$, $w=100 \, \mu m$ and $h=20 \, \mu$. We will see in Section 9.3 that this model is of a sufficient accuracy to discuss the importance of the cathode fall.

For the modeling of the arc plasma, a one-dimensional time-dependent model is used. The plasma is considered to be homogeneous in the $x$ and $y$-directions, with $l = 1 \, mm$ and $w=100 \, \mu m$ while the $z$-direction, in which the largest gradients appear, is resolved on a numerical grid.
9.3 The cathode fall

To conduct current, a spark gap plasma requires a mechanism that supplies electrons from the cathode to the plasma. In a conventional arc, the electrodes are hot, and thermionic emission is the main source of electrons from the cathode. In this case, however, the cathode is initially cold, and does not heat up significantly in the short discharge time ($\sim 2\,\text{ns}$). Hence, thermionic emission can not produce a significant amount of electrons; instead, field emission and secondary emission must be the dominant processes supplying electrons to the discharge. Both these processes are enhanced by a cathode fall, a region of positive space charge in front of the cathode. This creates an electric field that may liberate electrons directly due to field emission, and accelerates ions toward the cathode, causing secondary emission.

In this Section, we will present a simple, analytical model of the plasma, which we use to describe the formation of the cathode fall. We are particularly interested in an upper estimate of the voltage drop over the cathode fall $V_{\text{cath}}$, which acts as a parasitic loss in the switch, and in an upper estimate of the time needed to form a cathode fall $t_{\text{cath}}$, as this may limit the switching time.

In this analytical model, we will approximate the laser-produced plasma by a uniform slab, with dimensions as shown in Fig. 9.3 and an $n_e$ of $5 \times 10^{23} \,\text{m}^{-3}$. The density estimate is based on an analysis of the laser intensity, and is detailed in Section 9.4.2, and is equivalent to an ionization degree of about 5%. The mechanism of the cathode fall formation in this system will be explained using the graphs in Fig. 9.4.
Figure 9.4: A schematic explanation of the cathode fall formation. The cathode is at $\xi = 0$, and the anode is at $\xi \gg 1$. (a) The femtosecond laser pulse produces a plasma on a time scale that is essentially instantaneous for the plasma we are interested in. (b) The electrons move because of the applied electric field, creating a cathode fall in which the field is compressed. This field facilitates electron emission by the cathode (bottom graph). The heavier ions are essentially motionless on this time scale.

The initial plasma is produced by the femtosecond laser pulse at a time scale that is much shorter than the plasma processes under consideration. The top graph of Fig. 9.4 schematically shows the laser-produced plasma density and the applied voltage. In the initial laser-produced plasma, the electric field applied across the spark gap will move electrons to the anode. The ion movement to the cathode is orders of magnitudes slower and negligible at this timescale. The movement of the electrons leaves a region near the cathode depleted of electrons, with a positive space charge that is very high due to the high ion density. The very high positive charge density causes an electric field to form near the cathode that is much higher than the average field, which facilitates the electron emission. This process is shown in the bottom graph of Fig. 9.4.

9.3.1 The cathode fall voltage

We will now estimate an upper bound of the cathode fall voltage. For this, the electron emission current from the cathode as a function of the electric field at the cathode $E_{\text{cath}}$ has been calculated using the Fowler-Nordheim equation [6, 7]

$$I_{\text{field}} = K_4 E_{\text{cath}}^2 A \exp \left( \frac{-K_5}{E_{\text{cath}}} \right),$$  \hspace{1cm} (9.1)

in which $K_4$ and $K_5$ are emission coefficients and $A$ is the cathode area. As noted in [6], (9.1) typically overestimates the field required for significant field emission by at
9.3. The cathode fall

least one order of magnitude. This is due to surface roughness, which may greatly enhance the local field and hence the emission. A typical value for this field enhancement parameter $\beta$ is 50, and we will use this value in the calculation.

Incorporating $\beta$ in (9.1) and substituting values for $K_4$ and $K_5$ we obtain [8]:

$$I_{\text{field}} = \frac{1.54 \times 10^{-6} \times 10^{4.52\phi^{-0.5}} (\beta E_{\text{cath}})^2 A}{\phi} \exp \left( \frac{-6.53 \times 10^9 \phi^{1.5}}{\beta E_{\text{cath}}} \right),$$

(9.2)

with $E_{\text{cath}}$ in V m$^{-1}$, the work function $\phi$ in eV, $A$ in m$^2$ and $I_{\text{field}}$ in A.

The cathode tips consist of a tungsten-copper alloy, which has an unknown work function. The work function of tungsten $\phi_W$ is 4.54 eV [9] and the work function of copper $\phi_{Cu}$ is 4.65 eV [10, 11]. The value of the work function of an alloy is typically between the value of the work function of the constituents [12].

For an electrode area of $2 \times 10^{-9}$ m$^2$ (cf. Fig. 9.3), $I_{\text{field}}$ as a function of $E_{\text{cath}}$ has been plotted in Fig. 9.5 for both tungsten and copper cathodes. Fig. 9.5 shows that $I_{\text{field}}$ is negligible for values of $E_{\text{cath}}$ lower than $9 \times 10^7$ V m$^{-1}$, and increases very rapidly with increasing $E_{\text{cath}}$ for higher values of $E_{\text{cath}}$. The maximum switched current of 50 A can be supplied by field emission by a field of $1.2 \times 10^8$ V m$^{-1}$. Noting that contributions from secondary emission might reduce the required $E_{\text{cath}}$ even further, we will use this value as an upper bound for the actual $E_{\text{cath}}$.

The cathode fall is governed by the Poisson equation

$$\nabla \cdot \vec{E}_{\text{cath}} = -\nabla^2 V_{\text{cath}} = \frac{\rho}{\epsilon},$$

(9.3)

with $\rho$ the charge density and $\epsilon$ the permittivity of the plasma.

The charge density $\rho$ is determined by the initial electron density and is in our case equal to $8.0 \times 10^4$ C m$^{-3}$. For $\epsilon$ we can take $\epsilon_0$, the permittivity of vacuum.

By approximating the $\nabla$-operators with $L^{-1}$, $L$ being the typical length scale of the cathode fall we find that $L \approx 10^{-8}$ m and $V \approx 2$ V. This cathode fall is high enough for field emission, but negligible compared to the typical voltages that are switched (a few kV).

9.3.2 The cathode fall formation time

The formation time of the cathode fall can be estimated from the time that an electron needs to travel over a distance equal to the size of the cathode fall. This time can be calculated using

$$q_e E_{\text{gap}} = m_e a \Rightarrow a = qV_{\text{gap}} \frac{m_e l}{m_e l}$$

(9.4)
Figure 9.5: The field emission current as a function of the electric field at the cathode surface, for values of the work function corresponding to copper and tungsten cathode material. The field emission current of the actual cathode material lies between the two curves in this graph.

and

\[ L = \frac{1}{2} at^2 \Rightarrow t = \sqrt{\frac{2L}{a}}, \quad (9.5) \]

where \( q_e \) is the charge of an electron, \( E_{\text{gap}} = \frac{V_{\text{gap}}}{l} \) is the field over the whole spark gap of length \( l \), \( a \) is the acceleration, \( m_e \) is the electron mass and \( t \) is the time. Using Eqs. (9.4) and 9.5, the previously obtained value of \( L \), and a value of 1 kV for \( V_{\text{gap}} \), we find that \( t_{\text{cath}} \) is about \( 3 \times 10^{-13} \) s. Note that this formation time is of the same order of magnitude as the inverse of the plasma frequency \( f_p \), which is equal to 6.4 THz for the electron density under consideration (\( 5 \times 10^{23} \) m\(^{-3} \)). The measurements of the switching time are limited to the measuring apparatus and were found to be shorter than 100 ps [4, 5], which is much longer than the estimated cathode fall formation time. The cathode fall formation time is also much shorter than rise time of the electric field in the gap due to the geometry, which is a few tens of picoseconds [13]. Hence, the cathode fall formation time has no influence on the rise time of the switched pulse.

9.3.3 Conclusions

Using an analytical model, the cathode fall formation dynamics have been investigated. The cathode fall mechanism can supply sufficient current for switching to occur. This
requires a voltage which is negligible compared to the switched voltage. The cathode fall voltage rise time is much shorter than the observed rise time. Hence, the cathode fall cannot explain the observed voltage losses in the switching, and does not limit the switching time. The main reason for this is the large value of $n_e$; for plasma with a lower densities, the cathode fall may be quite significant.

9.4 The arc plasma

In this Section, a numerical model of the arc that connects the electrodes will be presented. Firstly, the numerical model used will be discussed, and key assumptions are justified. Then, the model parameters, such as the current and the laser-produced plasma density which serves as a starting condition for the computation will be presented. Based on the results from the numerical model, the general behavior of the plasma will be discussed. Finally, the impact that the finite plasma conductivity has on the switching behavior will be investigated by comparing experimental results with numerical results for various switched currents.

9.4.1 The model

The model used for the description is a two-temperature time-dependent quasineutral non-LTE model. It is based on the model of a hydrogen-filled pulsed capillary discharge that is presented in [14].

For the numerical solution of the model we use the PLASIMO code. This code is described in detail in [15–19]. It is a modeling platform that can handle LTE and non-LTE plasmas, currently in two dimensions. Furthermore, its modular structure allows for easy expansion of the code. It has been applied to simulate a wide variety of plasma as described in [14, 20–26].

The system is described by a set of partial differential equations, consisting of the Navier-Stokes equations that describe the bulk flow, two heat transport equations that describe the electron and heavy particle heat transport, respectively, and chemistry equations that describe the densities of individual species. For details of the equations, the reader is referred to [14].

These equations are discretized on a control volume grid, that is oriented as shown in Fig. 9.3. The plasma is considered to be infinitely long and homogeneous in the $x$ and $y$ direction, while the computation is carried out in the $z$-direction, where the largest gradients are to be expected. Because is symmetric, only the top half of the plasma is modeled.

The chemistry model assumes that the plasma is operated in pure nitrogen. The
chemistry model is restricted to 5 species, namely N\(_2\), N, N\(_2^+\), N\(^+\) and e. The expected very high energy density in the system means that more complex molecular species such as N\(_4^+\) are very rapidly destroyed and play no significant role. Vibrational excitation of N\(_2\) is not included in the model, as the resulting electron temperatures are so high that direct ionization is easily possible. Heavy particle reactions are not very important due to the low heavy particle temperature and corresponding slow reaction rate.

Because of the high electron density \(n_e\) (\(\sim 10^{23}–10^{24} \text{ m}^{-3}\) and temperature \(T_e\) (1.5–8 eV) and relatively low heavy particle temperature \(T_h\) (0.03–1.0 eV), electron kinetics dominate the reactions in the plasma. These \textit{a priori} assumptions are justified by the results of the model as presented in sec. 9.4.4).

The following reactions are included in the model:

- Electron impact ionization of N\(_2\), described by
  \[ N_2 + e \rightarrow N_2^+ + 2e. \] (9.6)

  The rate coefficient for this equation is obtained by integrating the energy-dependent cross section with a Maxwell-Boltzmann Electron Energy Distribution Function (EEDF) for various temperatures and fitting an Arrhenius-like formula through the results. The energy-dependent cross section of this reaction is well-known, and we use the value obtained by Deutsch \textit{et al.} [27] from a theoretical computation, which matches earlier experimental [28–32] and theoretical [33, 34] results well.

  The rate coefficient \(k_{MI}\) of this reaction as a function of \(T_e\) is described by the Arrhenius rate
  \[ k_{MI} = 3.0 \times 10^{-18} T_e [K]^2 \exp \left( \frac{-E_{MI}}{k_B T_e} \right) \text{ m}^3 \text{ s}^{-1}, \] (9.7)
  with \(E_{MI}\) the ionization energy of 15.6 eV and \(k_B\) Boltzmann’s constant. This rate describes the ionization process well (fitting error less than 6%) between 1.5 and 10 eV.

  In the model, the reverse reaction is also included; its rate is computed from (9.7) by detailed balancing.

- Dissociative recombination of N\(_2^+\), described by
  \[ N_2^+ + e \rightarrow 2N. \] (9.8)

  The rate coefficient \(k_{DR}\) for this rapid process had been obtained by Cunningham and Hobson [35] and is given by:
  \[ k_{DR} = 1.47 \times 10^{-12} T_e [K]^{-0.37} \text{ m}^3 \text{ s}^{-1}. \] (9.9)
9.4. The arc plasma

- Electron impact dissociation of $N_2$, described by

$$N_2 + e \rightarrow 2N + e.$$  \hspace{1cm} (9.10)

A recommended cross section for this process has been obtained from Cosby [36], who uses measurements and previous results by [37]. A good fit by an Arrhenius-like rate could only be obtained by increasing the energy threshold $E_{\text{Diss}}$ from the binding energy of 9.8 eV to 11 eV. Using this fit, the following expression for the rate coefficient $k_{\text{Diss}}$ is obtained:

$$k_{\text{Diss}} = 2.0 \times 10^{-18} T_e \left[ K^{0.8} \exp \left( \frac{-E_{\text{Diss}}}{k_B T_e} \right) \right] \text{m}^3 \text{s}^{-1}. \hspace{1cm} (9.11)$$

The fit of (9.11) is accurate to within 16% between 1.5 and 20 eV.

In the model, the reverse reaction is also included; its rate is computed from (9.11) by detailed balancing.

- Electron impact ionization of atomic N, described by

$$N + e \rightarrow N^+ + 2e.$$  \hspace{1cm} (9.12)

Voronov [38] gives an Arrhenius-like rate coefficient, which, with appropriate parameters, can be used to find rate coefficients for many single atoms and ions, including the rate coefficient $k_{\text{AI}}$ for electron impact ionization of atomic N. $k_{\text{AI}}$ is given by

$$k_{\text{AI}} = 4.82 \times 10^{-14} \frac{1}{U + 0.0652} U^{0.42} \exp(-U) \text{m}^3 \text{s}^{-1}, \hspace{1cm} (9.13)$$

in which the reduced temperature inverse $U$ is given by

$$U = \frac{E_{\text{AI}}}{k_B T_e}. \hspace{1cm} (9.14)$$

According to [38], this fit is accurate to within 10% between 1 eV and 20 keV.

In the model, the reverse reaction is also included; its rate is computed from (9.14) computed by detailed balancing.

It is worth noting that in this plasma, the typical electron temperature is so high that direct ionization in (9.12) is dominant over stepwise ionization. Hence, the latter process is neglected.

Because of its crucial influence on the voltage drop over the arc, and hence on the difference between $V_{\text{in}}$ and $V_{\text{out}}$, care has been taken to accurately describe the conductivity $\sigma$ of the plasma. The description is based on the Frost mixture rule [39]. In the
limit of a fully singly-ionized plasma, this mixture rule converges to the Spitzer-Härm value $\sigma_{SH}$ of [40–42]

$$\sigma_{SH} = 1.53 \times 10^{-2} \frac{T_e^3}{\ln \Lambda_C} \text{Sm m}^{-1} \tag{9.15}$$

with the electron temperature $T_e$ in K and $\ln \Lambda_C$ the Coulomb logarithm. For a non-fully singly ionized plasma, the collisions between electrons and neutrals, viz. $N$ and $N_2$, cause the conductivity to be lower. In order to obtain a good value for $\sigma$, it is necessary to have good values for the energy-dependent elastic momentum transfer collision cross section for both $e$ and $N$, which will be denoted as $\sigma_{Ne}$ and for $e$ and $N_2$, which will be denoted as $\sigma_{N_2e}$.

A value for $\sigma_{Ne}$ that is valid between 0 and 1000 eV has been obtained by combining values from [43] that give $\sigma_{Ne}$ for electron energies between 0 and 12 eV, and values from [44] that give $\sigma_{Ne}$ for electron energies between 20 and 1000 eV. The gap in the energy range has been bridged by an interpolation.

A value for $\sigma_{N_2e}$ that is valid for electron energies between $10^{-2}$ and $10^3$ eV has been obtained from [45, 46].

For the typical $T_e$ in the system, these cross sections are valid for the majority of electrons. Together with the Coulomb cross section, which describes the interaction of the electron with the charged particles in the discharge, these cross sections are sufficient as input for the Frost mixture rules that describe $\sigma$.

The transport processes by the heavy particles are relatively unimportant due to the relatively high mass of the heavy particles and relatively low $T_h$. Their behavior will be dealt with using the default transport models in PLASIMO.

With the abovementioned model, we can simulate the transient behavior of the discharge plasma. However, the starting conditions are determined by the initial laser-produced plasma, the properties of which we will describe in the next Section.

### 9.4.2 The initial condition

The spark gap is switched by a pulse of a femtosecond-terawatt laser, which produces a plasma in a time that is much faster than all non-photoinduced plasma processes. The composition of the initial plasma is currently not know. Hence, we will provide an estimate of the composition based on theoretical arguments.

The intense laser field of around $10^{18} - 10^{19}$ W m$^{-2}$ [4] is sufficient for photoionization to be the main ionization mechanism [47–49]. This mechanism mainly produces $N_2^+$, dissociation of laser-produced $N_2^+$ also produces $N^+$. The higher ion states of nitrogen are neglected, as their density is lower by orders of magnitude.

Based on the measurements of the switching behavior as a function of laser intensity [4], we are lead to believe that for the laser energies used, the ionization degree is far
9.4. The arc plasma

less than unity.

The focal height of the laser has been measured [5], and based on this measurement, we will approximate the $n_e$-profile in this direction with a Gaussian that has a full width half maximum (FWHM) of 20 $\mu m$. The laser-produced plasma is schematically shown in Fig. 9.3.

Using these estimates, we have approximated $n_e$ by

$$n_e = 5 \times 10^{23} \exp \left( \frac{-z^2}{2(20 \mu m)^2} \right) \text{m}^{-3}. \quad (9.16)$$

We further assume that of the corresponding ions, 90% is $N^+_2$ and 10% is assumed to be $N^+$. $n_N$ is low, as there is no efficient laser-induced production mechanism of this species, and assumed to be $4.2 \times 10^{21} \text{m}^{-3}$. $n_{N_2}$ has a density that is determined such that the local mass density always matches the mass density of nitrogen at atmospheric density and room temperature.

9.4.3 The current

In Sections 9.4.1 and 9.4.2, a model for the initial conditions has been presented. The plasma evolution now depends on the current $I$ that passes through the plasma. This current is supplied by the transmission line, the amplitude depending on the charging voltage of the line, and the duration on the length of the transmission line.

When the gap is closed, the voltage is divided over the two sides of the line. This leads to a halving of the amplitude but a doubling of the duration of the current pulse the gap. The length of the transmission line is 0.3 m, resulting in a pulse duration of 2 ns [5].

We have investigated the switching behavior for various values of the arc current. The maximum voltage across the gap that has been switched is 5 kV [4, 5]. For a geometrical impedance of approximately 55 $\Omega$ [4, 5], this results in a block-shaped current pulse that lasts 2 ns and has a maximal amplitude $I_{\text{max}}$ of 50 A in the ideal case, because the voltage over the gap is half the switched voltage. The spark gap plasma is simulated for various values of $I_{\text{max}}$ between 0 and 50 A in increments of 5 A.

Although the gap is closed at a femtosecond time scale by the laser pulse, the electric field that is induced by the charged transmission line needs a much longer time to adapt to this situation. In [13], it has been determined that it takes several tens of ps for the electric field in the spark gap to stabilize. In the model, we approximate this by using
The plasma evolution is most for pronounced for the highest power input of $I_{\text{max}}=50$ A. Hence, a detailed analysis of the plasma physics will be made for this case. For the other values of $I_{\text{max}}$, the discussion will be restricted to the key quantity, the resistance.

### 9.4.4 Modeling results for $I_{\text{max}} = 50$ A

The plasma that is present in the spark gap with an $I_{\text{max}}$ of 50 A will be discussed based on five properties: $n_e$, $T_e$, $T_h$, $\sigma$, and the central plasma composition.

The electron density $n_e$ as a function of $t$ and $z$ is presented in Fig. 9.6, in which two observations are made. The central $n_e$ rises by more than one order of magnitude during the discharge. Furthermore, the discharge does not expand much: Outside of the laser-ionized channel, there are few seed electrons, so ionization outside of the central channel is slow, and the discharge time is too short for a significant convective expansion. Between 200 ps and 2 ns, the central $n_e$ increases almost linearly in time. The electrons, which are heated by Ohmic dissipation, lose their heat primarily through electron impact ionization reactions. As the dissipation is roughly constant over the time range considered, the production is constant, causing the linear increase. It is worth noting that in the 2 ns of the discharge, no equilibrium between the Ohmic dissipation and the energy loss by thermal conduction is established, meaning there is a net ionization.

After the current pulse, the plasma decays. This decay is much slower than the plasma creation during the pulse.

The electron temperature $T_e$ as a function of $t$ and $z$ is presented in Fig. 9.7. During the discharge, the plasma is coldest in the center, where the dissipation is highest. The electron density peaks in the center, causing the dissipated power per electron to be lowest there. The increase of $n_e$ is also causing a decrease the dissipated power per electron and hence a decrease in $T_e$ in time. After the discharge, $T_e$ rapidly collapses, because the electrons are no longer heated by Ohmic dissipation. $T_e$ is chiefly determined by the energy gained and lost due to recombination and ionization, respectively.

The heavy particle temperature $T_h$ as a function of $t$ and $z$ is presented in Fig. 9.8. In the beginning, the background gas heats up rapidly. This is due to the energy that is liberated in the dissociative recombination of $\text{N}_2^+$ via reaction (9.8). With the depletion
9.4. The arc plasma

Figure 9.6: The electron density $n_e$ in the spark gap as a function of $t$ and $z$, for $I_{\text{max}}=50$ A

Figure 9.7: The electron temperature in the spark gap as a function of $t$ and $z$, for $I_{\text{max}}=50$ A. The electron temperature is less relevant outside of the central ionized channel, because $n_e$ is very low there (cf. Fig. 9.6).

of $N_2^+$, this process becomes less important, and the heating slows down. $T_h$ remains lower than $T_e$ for the entire duration of the simulation.

$\sigma$ as a function of $t$ and $z$ is presented in Fig. 9.9. In first few picoseconds, the electron temperature reaches a value that is determined by the loss from ionization and the gain from Ohmic dissipation. Because $\sigma$ depends on $T_e$, it also reaches a fairly stable value at this time scale. When the electron density starts increasing (cf. Fig. 9.6), $\sigma$ increases as well. This effect is stronger than the decrease in $\sigma$ due to the decrease in $T_e$. The
Spitzer-Härm value $\sigma_{SH}$ of Eq. (9.15) of the conductivity is not reached in this plasma. Because of the very large momentum transfer cross section between electrons and nitrogen atoms and molecules, a very high $n_e$ is needed to make the Coulomb collisions dominant, which is the case in the Spitzer-Härm limit.

When the current pulse ends, $T_e$ drops sharply, leading to a strong drop in $\sigma$. This indicates that a high $T_e$, and associated with this, Ohmic dissipation in the plasma, is necessary for switching.

Because the Spitzer-Härm limit has not been reached, further ionization can increase the conductivity. Because $n_e$ increases with increasing dissipation, as seen in Fig. 9.6, conductivity will be better for higher currents and hence for higher switched voltages. In other words, a smaller fraction of the voltage will be lost over the gap for higher switched voltages.

The evolution of the on-axis composition of the plasma as a function of $t$ is presented in Fig. 9.10. The predominant ion species in the laser-produced plasma is $N_2^+$.

The dissociative recombination increases for increasing $n_e$ and decreasing $T_e$ (cf. Eq. (9.9)). Hence, $n_{N_2^+}$ drops during the evolution of the discharge. Another sharp drop is observed after the current pulse ends. At this time, $T_e$ drops, causing the rate of dissociative recombination to be even higher, and $n_{N_2^+}$ to drop further.

There is a rapid production of N, chiefly by dissociation of N$_2$, although dissociative recombination of N$_2^+$ also gives a modest contribution. This N is subsequently ionized to N$^+$, which becomes the dominant ion species. Except for $n_{N_2^+}$, the plasma composition does not significantly change after the pulse ends. The change $n_{N_2^+}$ is caused by an increase in the dissociative recombination speed, which is inversely proportional to $T_e$. 

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**Figure 9.8:** The heavy temperature $T_h$ in the spark gap as a function of $t$ and $z$, for $I_{max}=50$ A.
Figure 9.9: The electrical conductivity $\sigma$ in the spark gap as a function of $t$ and $z$, for $I_{\text{max}}=50$ A.

Figure 9.10: The densities of various plasma constituents as a function of $t$.

(cf. (9.9)).
Figure 9.11: Left axis: the total resistance $R$ of the spark gap plasma at $t = 2\text{ ns}$, as a function of $I_{\text{max}}$. The resistance increases dramatically for decreasing currents. Right axis: the simulated and measured [5] percentage of the applied voltage that is switched by the spark gap. The match is good, in particular for lower currents, although for higher currents, the simulated gap resistance is higher than the observed resistance.

9.4.5 Conductivity for various currents

One of the aims of this study is investigating the cause of the voltage that is lost in the switching. It is observed that the relative loss is larger for decreasing voltages. As mentioned in Sec. 9.4.4, the plasma conductivity is lower for lower dissipated powers. By running the simulation for various values of $I_{\text{max}}$, this has been verified. A typical result, the total resistance of the plasma arc $R$ at $t = 2\text{ ns}$, is presented in Fig. 9.11.

Fig. 9.11 shows a qualitative trend that matches the experimental observations well. The gap resistance becomes very high when little current is switched, while it is much lower, down to about 20 $\Omega$, for the higher switched currents, which is much smaller than the system impedance of 55 $\Omega$. For this ratio of the resistances, over 73% of the voltage on the gap is switched and 27% is dissipated. In this case, the voltage drop across the spark gap in the model is higher than in the experiment. In the low-current limit, the highest impedance of about 800 $\Omega$ would mean that about 94% of the voltage is across the gap and only 6% reaches the output, meaning no switching occurs. Furthermore, the response of the resistance on the current is strongly nonlinear, with a very sharp increase in voltage drop for lower currents, as is observed in the measurements.

Quantitatively, the agreements between the simulations and the experiments are less
The simulated resistances are a factor of 2 to 3 higher than the resistances that are measured. This can be explained by the poorly known starting conditions, which make a more accurate simulation of the plasma impossible. We have quantitatively analyzed the influence of the initial density. In particular the width of the distribution was important, as the resistance of the channel is determined by the width. The exact value of the electron density is of less importance, as only a modest fraction of the electrons originates from the original plasma. Furthermore, a lower electron density leads to more dissipation and hence more electron production.

We have neglected the fact that the laser power and hence the initial plasma density is also a function of $x$, dropping off near the electrodes. This could lead to a configuration in which two zones with lower plasma density near the electrodes determines the switching behavior, while a more strongly ionized plasma in the center acts as a good conductor between these zones. Without knowledge of the initial plasma density, this hypothesis cannot be verified.

9.5 Conclusion

The photoconductively switched spark gap is experimentally known to be capable of switching several kV of voltage with jitter that is typically in the 10s of ps [4, 5]. Voltages down to 10% of the self-break down voltage can be switched. One presently unresolved question is the cause of the voltage drop over the gap, which is most pronounced for lower voltages. We have investigated two plausible causes of this behavior: the cathode fall and the finite resistance of the arc plasma in the spark gap.

Based on a simple, analytical model of the cathode fall, the formation time is estimated to be less than 300 fs, while the voltage drop over the cathode fall is less than 2 V. The formation time is orders of magnitude shorter than the geometric rise time of the switched pulse, and is therefore not limiting in the switching behavior. The cathode voltage drop is typically one tenth of a percent of the switched voltage, and is much smaller than the experimentally observed voltage drops across the spark gap. From this, we conclude that the cathode fall behavior cannot be the cause of the observed voltage drop.

A non-LTE model has been used to compute the plasma properties of the arc plasma, including the electrical resistance of the spark gap plasma that bridges the electrodes. Due to the poorly known initial conditions, the model cannot be expected to give a full quantitative explanation of the switching behavior of the spark gap. Qualitatively, the arc plasma becomes a good electrical conductor for higher switched voltages, with a maximum switched voltage of 70% for the current range investigated, and a poor conductor for lower switched voltages, switching 6% in the limit of no voltage. The strong
nonlinearity of the switching behavior, i.e. a strong decrease in the switching efficiency for lower voltages, as is experimentally observed is also present in the modeled results. Based on this, we conclude that the voltage drop across the plasma is likely caused by the finite resistance of the arc plasma.

Quantitatively, the match is very good for the lower current, but somewhat less so for the higher currents, as experimentally, virtually all the voltage is switched for higher voltages, while in the model, a maximum of 73% of the voltage is switched.

The successful modeling of the plasma proves the feasibility of modeling the plasma in a photoconductively switched spark gap. The PLASIMO code is capable of simulating the complex, strongly dynamic behavior of the plasma on these short time scales.

Acknowledgments

The PLASIMO team members, current and former, are acknowledged for their contributions to the code. This work was funded by the Technology Foundation STW, applied science division of NWO and the technology program of the Ministry of Economic Affairs, the Royal Netherlands Academy of Arts and Sciences, and the Foundation for Fundamental Research on Matter (FOM).

Bibliography


Chapter 10

Numerical investigation of the discharge characteristics of the pulsed discharge nozzle

Abstract—The characteristics of the plasma generated by a pulsed discharge slit nozzle (PDN) are investigated. The PDN source is designed to produce and cool molecular ions creating an astrophysically relevant environment in the laboratory. A discharge model is applied to this system to provide a qualitative as well as a quantitative picture of the plasma. We find that the properties and behavior of the plasma are characteristic of those of a glow discharge. We model the electron density and energy, as well as the argon ion and metastable atom number density. The results reveal a high abundance of metastable argon atoms in the expansion region, which is more than one order of magnitude higher than the abundance of electrons and ions. These findings confirm experimental observations, which concluded that large molecular ions are dominantly formed through Penning ionization of the neutral molecular precursors seeded in the supersonic expansion of argon gas. The simulations presented here will help optimize the yield of formation of molecular ions and radicals in the PDN source; they will also provide key physical insight into the characteristics of interstellar molecules and ions analogs in laboratory experiments.

10.1 Introduction

Carbon containing species such as polycyclic aromatic hydrocarbons (PAHs) are important building blocks of interstellar dust and are detected in meteorite samples and inter-

This Chapter was published, in slightly altered form, as B. H. P. Broks, W. J. M. Brok, J. Remy, J. J. A. M. van der Mullen, A. Benidar, L. Biennier, F. Salama, Physical Review E, 71, 036409 (2005)
planetary dust particles. Observational, laboratory, and theoretical studies have shown that PAHs, in their neutral and ionized forms, are indeed an important and ubiquitous component of the interstellar medium [1]. PAHs might account for two unexplained phenomena, which have been challenging the astrophysical community for almost a century: (a) the unidentified infrared bands observed in the infrared emission spectrum of the interstellar medium (ISM), and (b) the diffuse interstellar bands seen in the visible and the near infrared absorption spectrum of diffuse interstellar clouds [2].

In order to assess the PAH proposal, which was first introduced by Leger and Puget [3] and by Allamandola et al. [4], we are constrained to compare astronomical observations with laboratory data, obtained under conditions that come close to those that reign in the ISM. This implies measuring the spectra of neutral and ionized PAHs isolated in the gas phase at low (cryogenic) temperature. Until recently, the best compromise was achieved by trapping PAH molecules in a rare gas solid matrix using matrix isolation spectroscopy. PAHs are large and nonvolatile molecules and the study of cold and ionized PAHs analogs in the gas phase remained an unexplored area until 1999 [5, 6].

Supersonic plasma sources have opened new possibilities for the generation of astrochemical species. These sources have been intensively used to produce isolated gas-phase carbon chains, which are cooled down and stabilized in the supersonic expansion, thus facilitating mass or optical detection [7]. Recently, supersonic plasma sources have been used to generate cold PAH cations in the gas phase in number densities high enough to enable the measurement of their optical absorption spectra by cavity ring down spectroscopy [8–10].

Such sources offer a powerful tool to produce and investigate radicals and ions, which are translationally and ro-vibrationally cold and vibrationally excited [11]. Supersonic expansion sources with long-slit orifices have more particularly proven to be efficient in generating high column densities of molecular ions and radicals, because of the increase in the absorption pathlength [12].

In spite of the potential of supersonic plasma expansions for producing astrochemical species, few studies have been carried out on the characteristics of the plasma generated. The research described in the present paper investigates a pulsed planar expansion generated by a pulsed discharge nozzle (PDN), that has been developed at NASA Ames to generate PAH cations in the gas phase [8].

The PDN source is based on a slit jet mounted in a vacuum chamber (see Fig. 10.1). The shutter runs at 10 Hz and typically opens for a duration of 1.2 ms, generating intense short gas pulses with flow rates of 8 cm³ per pulse. Two negatively biased jaws forming the cathode are mounted on each side of the 200 µm wide and 10 cm long slit and are 400 µm apart. They are insulated from the PDN assembly (anode) by a 1.5 mm thick insulator plate (Macor® plate).

On the left hand side, neutral gas enters with a pressure of approximately 10⁵ Pa.
The right hand side opening originates in a vacuum chamber, which is pumped to a backing pressure of 20 Pa. Because of this steep pressure gradient, the gas flows through the discharge region and expands supersonically in the vacuum chamber.

A voltage ranging from $-400$ to $-600$ V (through 1 k$\Omega$ ballast resistors) is applied for 500 $\mu$s during the gas pulse, ensuring that electrons flow against the supersonic stream and that the discharge remains confined as well as uniform along the slit. The plasma is thus generated in the expanding gas. The geometry of the source leads to a residence time of a few microseconds for the molecules in the active region of the discharge.

The plasma generated in the PDN source has been experimentally characterized as a glow discharge in the abnormal regime. A detailed qualitative analysis can be found in Refs. [13, 14]. Estimates of the electron temperature and electron density in the plasma have been obtained in these studies but do not provide a full and proper comprehensive description of the plasma properties.

In this Chapter, we will continue the research outlined above. We use a two-dimensional fluid model [15] to gain insight into the physicochemical processes occurring in the PDN source. It will be shown that our model is capable of generating a picture that is qualitatively and quantitatively consistent with the experimental observations from Refs. [13, 14].

In Sec. 10.2, we describe the model on which the simulations are based. This includes a brief description of the fluid equations, the data on species and reactions, the representation of the geometry and how the flow is calculated and taken into account in
the discharge model. In Sec. 10.3, we present simulation results on the spatial profile of relevant quantities associated with the plasma. We will also discuss and interpret these results from both a physical and a chemical point of view.

10.2 Model

In this Section we will describe the model in its various aspects: the equations that are solved, the data on species and reactions, the numerical approximation of the geometry of the device, and finally the background density field and flow field that we obtained from separate calculations.

10.2.1 Model description

In order to describe the PDN numerically, we made use of an existing discharge model and modified it slightly to take the flow of the background gas and the variations in the background gas density into account. This model has previously been used for describing plasma display panel pixels [16] and the ignition of compact fluorescent lamps [15]. It is described fully in Ref. [15] and references therein. Briefly, this is a time-dependent, two-dimensional fluid model, in which several balance equations are solved in conjunction with the Poisson equation.

The balance equations are solved in the drift diffusion approach for a number of species, such as electronically excited species, ions and electrons, all specified in Sec. 10.2.2,

\[
\frac{\partial n_p}{\partial t} + \nabla \cdot \Gamma_p = S_p
\]  

(10.1)

with

\[
\Gamma_p = \pm \mu_p E n_p - D_p \nabla n_p + v_b n_p,
\]  

(10.2)

where \(n_p\) is the density of species \(p\), \(S_p\) the source of the species due to reactions, \(\mu_p\) and \(D_p\) the species’ mobility and diffusion coefficient respectively, \(E\) the electric field, and \(v_b\) the flow velocity of the background gas. The last term in Eq. (10.2) has been added, as compared to the model described in Refs. [15, 16]

In addition to the balance equations for the various species, a balance equation is included for the electron energy, with a source term representing the energy gained in the electric field and the energy lost in collisions. The inclusion of this equation allows us to specify the various transport and reaction rate coefficients as functions of the mean electron energy. This approach differs from the commonly used local field approximation, in which these quantities are specified as functions of the local electric field strength and the assumption is made that the electron energy is solely determined by local conditions.
The electron transport and reaction rate coefficients were obtained from an external Boltzmann solver [17]. This solver calculates the electron energy distribution function in the pulsed Townsend approximation. From the resulting electron energy distribution functions at different reduced electric field strengths, the various coefficients are calculated as a function of the mean electron energy. These data are used as input to the model via lookup tables. The transport coefficient for the other species and the reaction rate coefficients for heavy particle reactions are taken from the literature and presented to the model as functions of the reduced electric field. These reaction and transport coefficients and the literature from which they originate are listed in Sec. 10.2.2.

As mentioned earlier, a distinct feature of the PDN is a gas flow through the device in the direction of the cathode. Since the power dissipated in the discharge is low enough to not cause appreciable gas heating, the flow of the background gas can be described independently from the discharge. In the discharge model we therefore made use of pre-calculated velocity and density fields (see Sec. 10.2.4 for these flow calculations) and used them as fixed input values. The nonuniform density field influences itself by its influence on the reduced electric field and thus on the source term in Eq. (10.1) and the various transport coefficients in the drift diffusion Eq. (10.2). The velocity field enters as an extra term in Eq. (10.2). Especially for the neutral species, the flow term forms an important contribution to the fluxes of these species.

As mentioned earlier, refer to Ref. [15] for the full set of equations, including boundary conditions. These equations are discretized on a rectangular uniform grid and solved via a control volume method.

10.2.2 Species

In this first paper we assume that the presence of PAHs, which are highly diluted in the argon carrier gas (typically about 0.1 % [8]), does not affect the discharge behavior appreciably.

The set of species and reactions that we incorporated in the model is equivalent to the one used in Ref. [15]. Briefly, the set of species consists of the electrons $e$, three effective excited states of argon, and the argon ion $\text{Ar}^+$. The first effective excited state, denoted $\text{Ar}^*$, represents the four levels in the 4s manifold of argon. This species is assumed to be metastable because the electron impact cross section for creating a metastable 4s level is significantly larger than the cross section for creating a resonant 4s level. The second excited state, denoted $\text{Ar}^{**}$, represents the 4p manifold and higher levels. In order to take into account collisional quenching of the two metastable states to the two resonant states in the 4s manifold, a third excited species $\text{Ar}_r^*$ is specified, which is destroyed by de-excitation to the ground level. The transport coefficients for these species are obtained from Refs. [17–20]. The value of the secondary emission coefficient for $\text{Ar}^+$
### Table 10.1: The reaction rate coefficients used by the model. Reactions marked ¹ are obtained from the forward reaction using microscopic reversibility.

<table>
<thead>
<tr>
<th>Nr.</th>
<th>Reaction</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>$\text{Ar} + e \rightarrow \text{Ar} + e$</td>
<td>[23]</td>
</tr>
<tr>
<td>1</td>
<td>$\text{Ar} + e \rightarrow \text{Ar}^* + e$</td>
<td>[24]</td>
</tr>
<tr>
<td>2</td>
<td>$\text{Ar} + e \rightarrow \text{Ar}^{**} + e$</td>
<td>[24]</td>
</tr>
<tr>
<td>3</td>
<td>$\text{Ar} + e \rightarrow \text{Ar}^+ + 2e$</td>
<td>[23]</td>
</tr>
<tr>
<td>4</td>
<td>$\text{Ar}^* + e \rightarrow \text{Ar} + e$</td>
<td>[24]¹</td>
</tr>
<tr>
<td>5</td>
<td>$\text{Ar}^* + e \rightarrow \text{Ar}^{**} + e$</td>
<td>[25]</td>
</tr>
<tr>
<td>6</td>
<td>$\text{Ar}^{*} + e \rightarrow \text{Ar}^+ + 2e$</td>
<td>[26]</td>
</tr>
<tr>
<td>7</td>
<td>$\text{Ar}^{**} + e \rightarrow \text{Ar} + e$</td>
<td>[24]¹</td>
</tr>
<tr>
<td>8</td>
<td>$\text{Ar}^{**} + e \rightarrow \text{Ar}^+ + e$</td>
<td>[25]¹</td>
</tr>
<tr>
<td>9</td>
<td>$\text{Ar}^{**} + e \rightarrow \text{Ar}^* + 2e$</td>
<td>[27]</td>
</tr>
<tr>
<td>10</td>
<td>$\text{Ar}^* + \text{Ar}^* \rightarrow \text{Ar}^+ + \text{Ar} + e$</td>
<td>[28]</td>
</tr>
<tr>
<td>11</td>
<td>$\text{Ar}^* + \text{Ar}^{**} \rightarrow \text{Ar}^+ + \text{Ar} + e$</td>
<td>[28]</td>
</tr>
<tr>
<td>12</td>
<td>$\text{Ar}^{<strong>} + \text{Ar}^{</strong>} \rightarrow \text{Ar}^+ + \text{Ar} + e$</td>
<td>[28]</td>
</tr>
<tr>
<td>13</td>
<td>$\text{Ar}^* + e \rightarrow \text{Ar}^*_r + e$</td>
<td>[29, 30]</td>
</tr>
<tr>
<td>14</td>
<td>$\text{Ar}^*_r \rightarrow \text{Ar} + h\nu$</td>
<td>[15]</td>
</tr>
<tr>
<td>15</td>
<td>$\text{Ar}^{**} \rightarrow \text{Ar}^* + h\nu$</td>
<td>[15]</td>
</tr>
</tbody>
</table>

on the electrode materials is strongly dependent on the quality of the surface, which is unknown. In this Chapter we use an estimated value of 0.01 electrons per incident ion.

The various reactions that are included in the model are listed in Table 10.1, including the original references to the literature from which the cross sections or rate coefficients were obtained. Note that the reaction rate coefficient for reactions in which electrons are involved were calculated using the above-mentioned Boltzmann solver. It is noted that heavy particle reactions are not taken into account. In contrast to low-pressure glow discharges [21, 22], this is justified, as the high density leads to frequent collisions between ions and background atoms, effectively preventing the ions from gaining a high speed in the electric fields that are present.

We do not take electron-electron collisions into account. Since the ionization degree is low and the electron density fairly high, inelastic collisions with the background gas are more common than electron-electron collisions. This is true in particular for the sheaths, where most of the reaction kinetics takes place.
10.2.3 Geometry

As schematically shown in Fig. 10.1 and described in more detail in the introductory Section, the pulsed discharge nozzle is composed of two electrode plates, separated by an insulator plate. A large aspect-ratio slit in this structure forms the discharge region, of which a cross section is shown in Fig. 10.2.

Experimental measurements show that the discharge has the positive current-voltage characteristic that is typically associated with an abnormal glow. This means that the discharge covers the whole electrode along the length of the slit, as opposed to the partial coverage that one would find in the normal glow regime. Given the nature of the plasma, and given the large aspect ratio of the slit, the device can be represented by a two-dimensional model. The Cartesian coordinates $x$ and $y$ thus describe the direction from anode to cathode and the direction perpendicular to this respectively (see Fig. 10.2). The omitted $z$ direction points along the length of the slit.

Note that the device is mirror symmetric with respect to the center plane of the slit. We therefore only need to describe one side of it. Also, the current is expected to run between the sections of the anode and the cathode that are the closest to each other. As a result, the calculation domain of the discharge will be confined to a small region along the $x$ axis in the channel. Our simulation will thus focus on the region highlighted in the figure. The validity of the latter assumption will be discussed in a later Section.

Finally, since the time scale needed by the discharge to reach steady state was found to be small compared to the typical discharge pulse length of 500 $\mu$s, we consider a DC operated discharge only. The ballast resistor in series with the discharge was set to $10 \, k\Omega \, cm^{-1}$, corresponding to $1 \, k\Omega$ for the slit length of 10 cm.

10.2.4 Bulk flow

The PDN flow is modeled by A. Benidar using a separate computer program MB–CNS that can calculate transient compressible flows in two-dimensional geometries. It is based on a finite volume formulation of the Navier-Stokes equations [31, 32]. For the reasons stated above, the problem is considered in two dimensions.

The program takes the following quantities as input: the geometry of the source, the molecular mass, viscosity, thermal conductivity, and heat capacity of the gas, and finally the initial conditions, consisting of the reservoir temperature and pressure. From these parameters, the program calculates a grid and subsequently the temperature, pressure, mass density, velocity, and Mach number of the flow profile.

The results of the two-dimensional flow simulation are described in detail in Ref. [33]. Only the main relevant features are briefly summarized below.

The physical conditions are calculated for long hydrodynamic times, close to the
Figure 10.2: A schematic drawing of the geometry of the pulsed discharge nozzle. The size of the central plasma channel is exaggerated for clarity. The dashed square is the part of the device that is simulated. The gas flows from a high-pressure source on the left to a vacuum chamber on the right.

stationary regime (>1 ms). The profiles of the key thermodynamic parameters along the flow axis center line are shown in Fig. 10.3.

Three zones can be distinguished in the flow pattern: (1) the gas injection channel, (2) the discharge region and (3) the postdischarge region. The flow velocity steadily increases in the injection channel but remains subsonic. The expansion becomes supersonic (Mach number $M > 1$) at the beginning of the second zone. The sudden growth of the Mach number in the first $\sim 200 \mu m$ is followed by a drop in the next $\sim 200 \mu m$, caused by the compression of the expansion by the walls of the interelectrode region. Then the Mach number gradually rises in the remaining 1.1 mm to achieve Mach 2 at the tip of the cathode. Finally, the postdischarge zone is characterized by the typical behavior of planar expansions with a temperature that is inversely proportional to the distance from the exit.

10.3 Results and discussion

10.3.1 Introduction

Using the model presented in Sec. 10.2, we simulated the pulsed discharge nozzle for a source voltage of $-500$ V. By using the time-dependent equations described in Sec. 10.2 on a suitable starting condition, a steady-state solution (differing less than 1% from the
10.3. Results and discussion

Figure 10.3: Key thermodynamic parameter profiles along the flow axis center line, obtained for an Ar backing pressure of $10^5$ Pa and a reservoir temperature of 339 K, with: (a) the temperature, (b) the velocity, (c) the pressure and (d) the density.

solution after a much longer time of 50 $\mu$s) is obtained in less than 15 $\mu$s. This is much shorter than the typical discharge time of 0.5 ms [8], meaning that stable conditions are present during most of the discharge. The discussion is restricted to this stable situation.

The plasma is analyzed, mainly using contour maps of the five most important physical quantities:

- The electron density $n_e$, in Fig. 10.4.
- The argon ion density $n_{\text{Ar}^+}$, in Fig. 10.6.
- The voltage $V$, in Fig. 10.7.
- The average electron energy $\varepsilon$, in Fig. 10.8.
- The argon metastable density $n_{\text{Ar}^*}$, in Fig. 10.9.
Each of these figures will be discussed. Finally, we will present some results of operation at different source voltages.

### 10.3.2 The electron density

The electron number density is shown in Figs. 10.4 and 10.5. It is, as expected, very small (< $1 \times 10^{17}$ m$^{-3}$) both in the interelectrode sheath, which lies under the insulating plate, and in the cathode fall near the surface of the cathode.

In the case of the interelectrode sheath, this is to be expected, as the ambipolar flux of the plasma to the wall and the recombination at the wall will cause an electron density which is lower than in the plasma bulk. The interelectrode sheath thickness ranges from about 25 $\mu$m near the anode to 100 $\mu$m near the cathode. This should be equal to several times the Debye length $\lambda_D$, which is given by Ref. [34]:

$$\lambda_D = \sqrt{\frac{\varepsilon_0 k_B T_e}{n_e e^2}}. \quad (10.3)$$

Substituting a value for $n_e$ of $3 \times 10^{18}$ m$^{-3}$ (from Fig. 10.5) and a value for $T_e$ of 3 eV (from Fig. 10.8), which are both typical for the central area between the insulating plates, in Eq. (10.3), we obtain a value of 8 $\mu$m for $\lambda_D$. The interelectrode sheath is several times $\lambda_D$ near the cathode, as expected, but near the anode, it is thinner because the plasma is pushed to the wall by the internal expansion.
In the cathode fall region, the electron density is also low, causing a positive space charge, characteristic for this region. The central electron density of the device reaches values of up to $3.4 \times 10^{18} \text{ m}^{-3}$. Along the cathode, the discharge becomes less intense further away from the tip. This is caused by the variation in the background gas density, which is the highest near the electrode tip and drops when moving further away from it. This lowering of the background gas density means that collisions between electrons and atoms of the background gas become less frequent, hence the ionization degree decreases. This furthermore implies that only a small part of the electrode is in fact responsible for the bulk of the plasma production. This justifies, \textit{a posteriori}, not simulating the full cathode, but only the areas that are the closest to the outflow opening.

10.3.3 The Ar$^+$ density

The Ar$^+$ density in the discharge is depicted in Figs. 10.5 and 10.6. There is a zone with a low ($< 5 \times 10^{17} \text{ m}^{-3}$) Ar$^+$ density in the center of the channel, near the anode. There is a distinctive ridge near the cathode surface, where the Ar$^+$ density is high, reaching a maximum near the tip and dropping off along the cathode surface.

Between the insulating plates, the Ar$^+$ density is nearly equal to the electron density of about $3 \times 10^{18} \text{ m}^{-3}$, indicating a quasi-neutral plasma.

\textbf{Figure 10.5:} The steady-state density of Ar$^+$ and electrons of the pulsed discharge nozzle for a source voltage of -500 V, at the symmetry plane at $y=0 \text{ mm}$ and near the wall at $y=0.1875 \text{ mm}$. 

[Graph showing density distribution]
Near the cathode, however, the Ar$^+$ density is far higher than the electron density, reaching values of up to $4.5 \times 10^{18} \text{ m}^{-3}$. This creates a band of positive space charge close to the cathode, with a maximum space charge of $0.72 \text{ C m}^{-3}$. This band causes the cathode fall region of the plasma. It is noted that the difference between the electron density and the Ar$^+$ density is the highest near the tip of the cathode. This also means
10.3. Results and discussion

**Figure 10.8:** The steady-state average electron energy $\epsilon$ of the pulsed discharge nozzle for a source voltage of -500 V. Notice that the colour map is logarithmic, and ranges from 3.16 eV to 74.8 eV. The bold lines indicate the different parts of the device.

**Figure 10.9:** The steady-state Ar$^+$ density of the pulsed nozzle discharge for a source voltage of -500 V. The colorbar and isolines range from 0 to $1 \times 10^{20}$ m$^{-3}$ in steps of $1 \times 10^{19}$ m$^{-3}$. The bold lines indicate the different parts of the device.

that the net space charge is the highest there. This is consistent with the cathode being more efficient in the region displaying the highest background gas density, as explained in Sec. 10.3.2.

Furthermore, there is an area of low Ar$^+$ density near the symmetry plane of the plasma on the anode side. This is caused by the interelectrode expansion, which pushes
ions toward the insulating plate. Because of the high densities of charged particles and modest fields between the plates, significant deviations from quasineutrality cannot occur in this region. Hence, the electron density also becomes low in this area (Fig. 10.4). As can be seen in Fig. 10.5, the electron density at the symmetry plane is higher than the ion density between the isolating plates, indicating that the lighter and faster electrons can diffuse somewhat further upstream than the heavier ions before the resulting space charge fields contain them.

### 10.3.4 The voltage

The voltage $V$ in the discharge is depicted in Fig. 10.7. The cathode, which is at $-476 \text{ V}$, is clearly visible. The difference between the source voltage and the cathode voltage is due to the voltage drop over the electrode ballast resistor.

A sharp drop in voltage over the cathode fall is also visible. Because of this, the voltage in the rest of the discharge is rather uniform, and consequently the electric field is small. This is commonly the case in glow discharges [35].

### 10.3.5 The electron energy $\varepsilon$

Fig. 10.8 depicts the average electron energy $\varepsilon$ in the discharge on a logarithmic scale. In the cathode fall, $\varepsilon$ is in the order of 60 eV, far higher than the ionization energy of argon. Hence, a strong ionization of argon is expected there. This is consistent with the results in Figs. 10.4 and 10.6. In the interelectrode region, $\varepsilon$ is about 4 eV. Near the anode, $\varepsilon$ is significantly higher, 6 eV. This is due to the compression of the plasma by the internal expansion, which increases the local dissipation density. In the expansion zone, however, the electric field is small, which results in little heating of the electrons, and a drop in their temperature down to 5 eV is seen in the simulation domain. This indicates that electron kinetics are mostly important near the cathode.

The dissociation energy ($\sim 3-4 \text{ eV}$) of PAHs is significantly lower than the ionization energy ($\sim 7 \text{ eV}$) [36]. This means that processes involving bulk electrons, which have mean electron energies of about 5 eV, are far more likely to dissociate rather than ionize the PAH, as the density of electrons with an energy above the dissociation threshold is far higher than the density of electrons with an energy above the ionization threshold. In this plasma, both average electron energy and density are fairly low, especially in the downstream area, so electron excitation of PAH is of minor importance compared to other ionization mechanisms.

The high value of $\varepsilon$ in the cathode fall means that the bulk of the electrons have sufficient energy to ionize argon on impact. The ionization rate thus mainly depends on the background gas density.
10.3.6 The Ar$^*$ density

The Ar$^*$ density in the discharge is depicted in Fig. 10.9. It can be seen that metastable argon can reach high densities, with a maximum density that is 20 and 30 times higher than the maximum Ar$^+$ and $e$ densities, respectively. There are two production centers of Ar$^*$ in the simulation domain: one near the anode and one near the cathode tip. The chief mechanism of transport of Ar$^*$ is bulk flow, as the flow is fast in this plasma compared to the diffusion, and neutral Ar$^*$ is not subject to drift induced by electric fields. This is clearly visible in Fig 10.9, as expanding plumes of Ar$^*$ follow the bulk flow coming from the production zones.

High number densities of Ar$^*$ are generated in the plasma due to the significant fraction of electrons with an energy higher than 11.5 eV and the large Ar excitation cross section. The results show that Ar$^*$ is more abundant in the plasma by more than one order of magnitude than electrons with an energy above the PAH ionization potential. Therefore, the production of PAH cations by Penning ionization should dominate over electron impact ionization assuming cross sections of the same magnitude based on the scarce data available [$4.2 \times 10^{-20}$ m$^2$ for Penning ionization of C$_{60}$ with Ar$^*(4p^3D_3)$ [37] and $< 10^{-20}$ m$^2$ for electron impact ionization (12 eV) of C$_{60}$ [38]). These results are confirmed by previous experimental observations [8].

10.3.7 Other source voltages

Based on the five plasma parameters discussed earlier, $n_e$, $n_{Ar^+}$, $V$, $\varepsilon$, and $n_{Ar^*}$, and further calculations not detailed here, such as numerical experiments in which the background gas density and flow velocity are varied, we will now discuss the mechanisms that are responsible for the operation of the pulsed discharge nozzle as a chemical source of cold PAH ions.

Because of the unusual geometry of the source, the variations in background gas density, and the presence of a significant bulk flow, it is not possible to solely look at classical gas discharge theory to adequately explain the operation of the pulsed discharge nozzle.

In a normal glow, the current can vary with only minimal variations in voltage. This is because the cathode is only partially covered with an electron-emitting cathode fall. Increasing or decreasing the area covered changes the amount of electrons emitted and consequently the current but does not change the voltage, provided the cathode surface and gas composition near the cathode are homogeneous.

In this device, the variations in the background gas density near the cathode mean that parts of the cathode fall will produce electrons more easily than others. This device will thus not exhibit the current-voltage behavior typical of a normal glow, as increasing
Figure 10.10: The simulated (solid line) and measured (crosses) current of the pulsed discharge nozzle as a function of source voltage.

the active area of the cathode would require using parts of the cathode which are less prone to electron emission due to the lower background density, which would require the voltage to rise when the current rises.

In addition to the results presented above, we have simulated the pulsed discharge nozzle for a variety of input voltages. A graph of the current density in the device is given in Fig. 10.10. Below $-380$ V, no stable solution is found. The current-voltage relation is positive, which is consistent with the above explanation.

Experimental data in Ref. [13] also show a positive current-voltage characteristic. Generally, the quantitative match between the measured and simulated voltage is excellent, the measured currents being 30% lower than the simulated data at most, while the threshold voltage matches within 2%. A better match cannot be expected given that many input parameters, such as reaction cross sections and the secondary emission coefficient, are not known to such a precision.

The discrepancy between the simulated and experimental current-voltage characteristics gives an indication of the error of the computed values of $n_e$ and $n_{Ar^+}$, which are expected to be of the same magnitude, as the conductivity is dependent on these parameters. This puts the expected error in $n_e$ and $n_{Ar^+}$ at around 50%. The error in $n_{Ar^*}$ is expected to be of a similar magnitude.
10.4 Conclusions

We have developed a model of the pulsed discharge nozzle and used it to obtain values for key plasma parameters and to gain insight into the physical processes that determine its behavior.

The plasma in the pulsed discharge nozzle is clearly a glow discharge, with a current-voltage characteristic typical of the abnormal regime. The plasma has a cathode fall near the cathode, a plasma sheath that separates it from the insulating wall, and a quasineutral bulk. For the typical operating voltages, the electron density is in the order of \(3 \times 10^{18} \text{ m}^{-3}\), which is at the high end of the value range that is typical for glow discharges.

The shape of the cathode and the variations in the density of the background gas cause the cathode to be most active near its tip. This also means that the current-voltage characteristics may be quite different from those of a standard parallel-plate glow discharge.

The argon metastables that are formed are not affected by the electric field, and therefore can be transported downstream easily. This reduces the chance of quenching and stepwise ionization, boosting the ratio of metastables to ions and strongly enhancing the behavior of the PDN as a source of cold metastable noble gas atoms. A numerical model in which flow was excluded has confirmed this behavior.

The low electron temperature in the expansion zone causes PAH ionization by electron impact to be of less importance. The flux of cold, metastable argon, with a well-defined excitation energy, makes it possible to ionize PAHs with limited fragmentation. By choosing a suitable noble carrier gas, the reaction energy can be adjusted as necessary. These traits make this PDN source particularly suitable for generation of astrochemically relevant species.

A comparison between the simulated and measured current-voltage characteristics shows an excellent match over the complete range of simulated voltages. This indicates \(n_e\) is computed accurately, and suggests the densities of the other plasma species are accurately computed as well.

Using this model, a controlled parameter study can now be performed by varying the background gas mixture and/or the geometry of the source to optimize the yield of metastable carrier gas atoms and, hence, the yield of interstellar analogs in the laboratory.
Acknowledgments

This project is supported under the NASA Science Mission Directorate APRA program and NASA Ames DDF project. B. H. P. Broks, W. J. M. Brok and J. Remy respectively acknowledge support from the Stichting voor Fundamenteel Onderzoek der Materie (FOM), from Philips Research and from the Center for Plasma Physics and Radiation Technology (CPS).

Bibliography


Numerical investigation of the pulsed discharge nozzle
Modeling the influence of anode-cathode spacing in a pulsed discharge nozzle

Abstract—The pulsed discharge nozzle (PDN) is a spectrochemical source that is designed to produce and cool molecular ions in an astrophysically relevant environment in the laboratory with limited fragmentation. In order to gain a better understanding of the PDN and to optimize the yield of molecular ions and radicals in the PDN source, a parameter study of the influence of the interelectrode distance on the plasma properties is carried out by means of a discharge model, providing a qualitative as well as a quantitative picture of the plasma. We model the electron density and energy, as well as the argon ion and metastable atom number density for various of the interelectrode distances. The results reveal that increasing the interelectrode distance does not significantly influence the plasma at the cathode and at the anode. However, a positive column forms between the electrodes, which increases in length as the interelectrode distance increases. This is additional evidence the PDN is a glow discharge. This positive column does not contribute significantly to the formation of metastable argon atoms. Because metastable argon is thought to be the primary agent in the formation of molecular ions through Penning ionization of the neutral molecular precursor there is no benefit to be expected from an increase of the interelectrode distance. In fact, electron impact dissociation of the molecules in the column might even make the source less efficient for longer column lengths. The simulations presented here provide physical insight into the characteristics of interstellar species analogs in laboratory experiments.

This Chapter was published, in slightly altered form, as B. H. P. Broks, W. J. M. Brok, J. Remy, J. J. A. M. van der Mullen, A. Benidar, L. Biennier, F. Salama, Spectrochimica Acta B 60, 1442-1449 (2005)
11.1 Introduction

Interstellar dust is known to contain complex carbon molecules, such as polycyclic aromatic hydrocarbons (PAHs)\cite{1}. Observational, laboratory and theoretical studies have shown that PAHs, in their neutral and ionized form, are an important component of the interstellar medium. PAHs might be responsible for two phenomena that have challenged the astrophysical community for over a century: the unidentified infrared bands observed in the emission spectrum of the interstellar media, and the diffuse interstellar bands seen in the visible and the near infrared absorption spectrum of diffuse interstellar clouds\cite{2}. In order to assess the PAH proposal, which was first introduced by Leger and Puget\cite{3} and by Allamandola et al.\cite{4}, we are constrained to compare astronomical observations with laboratory data, obtained under conditions that come close to those that reign in the interstellar medium. This places certain demands on the spectrochemical source used to ionize PAHs: the background gas must be at cryogenic temperature, and PAHs must be ionized in their ground state. Supersonic plasma sources meet these demands\cite{5}, because they produce metastables which are capable of soft Penning ionization of PAHs\cite{6}.

These sources operate by evaporating and seeding PAHs, which are large and non-volatile molecules, in a noble carrier gas. The PAHs are cooled down and stabilized in the supersonic expansion. Recently, supersonic plasma sources have been used to generate cold PAH cations in the gas phase and combined to cavity ring down spectroscopy to measure their optical absorption spectra\cite{7–9}.

Such sources offer a powerful tool to produce and investigate radicals and ions, which are translationally and rotationally cold\cite{10}. Supersonic expansion sources with long-slit orifices have particularly proven to be efficient in generating high column densities of molecular ions and radicals, because of their geometry\cite{11}.

The Pulsed Discharge Nozzle (PDN) source, which has been developed at NASA Ames\cite{7}, is such a supersonic source that can produce PAH metastables. The geometry of the PDN is schematically illustrated in Fig. 10.1. It is based on a slit mounted in a vacuum vessel. The gas shutter runs at 10 Hz and typically opens for a duration of 1.2 ms, generating intense short gas pulses with flow rates of 8 cm$^3$ per pulse. Two negatively biased jaws forming the cathode are mounted on each side of the 200 $\mu$m wide by 10 cm long slit and are 400 $\mu$m apart. They are insulated from the anode by a Macor$^\text{©}$ spacer plate which is 1.5 mm wide.

On the left hand side, neutral gas, in our case argon, enters with a pressure of approximately 10$^5$ Pa. The right hand side opening originates in a vacuum chamber, which is pumped to a backing pressure of 20 Pa. Because of this steep pressure gradient, the gas flows through the discharge region and expands supersonically in the vacuum chamber.

A negative voltage of several hundreds of volts (through 1 k$\Omega$ ballast resistors) is
applied for 500 $\mu$s during the gas pulse. The plasma is thus generated in the expanding gas. The geometry of the source leads to a residence time of a few microseconds for the molecules in the active region of the discharge.

In order to interpret the results of the measurements on this source, and compare them to the measurements done on the astrophysical medium, it is desirable to understand the operation of the source so one knows in what form and what environment the species that is to be analyzed is produced. Furthermore, understanding a source facilitates optimizing it.

The plasma generated in the PDN source has been qualitatively analyzed in Refs. [12, 13]. Estimates of the electron temperature and electron density in the plasma were obtained in these studies but do not provide a full and comprehensive description of the plasma properties by themselves.

In order to obtain a quantitative insight in the discharge properties, a detailed numerical study of the device was made in Ref. [6]. In this study, a precalculated flow field was used as an input for a glow discharge model. The results matched the available experimental results very well. An important conclusion was that the PDN acts as a source of cold metastables. These metastables, rather than the electrons, are the spectrochemical agents that ionize the PAHs, doing so via Penning ionization. Furthermore, it was concluded form the current-voltage characteristic that the PDN is a glow discharge in the abnormal regime.

In Ref. [6], we have speculated that changing the geometry could improve the yield of ionized PAH in the source. In particular, it was thought that by increasing the spacer plate width and hence the interelectrode distance, the residence time of PAHs in the plasma is increased, which should result in a higher yield of PAH ions. We investigate this proposal by a numerical study of three PDN assemblies with an interelectrode distance of 1.5 mm, 4 mm and 10 mm, respectively. The study will be carried out using the model outlined in Ref.[6]. By comparing the results for the various interelectrode distances, conclusions can be drawn about the impact of the interelectrode distance on the source efficiency in the production of cold molecular ions. Furthermore, additional insight into the physics of the source may be gained and can lead to improvements of the source, such as a reduced dissociation of molecules.

Of equal importance is the additional physical insight in the plasma that produces the spectrochemical species. It was concluded in Refs. [12, 13] that the plasma behaves as a glow discharge. This means that lengthening the device could lead to the formation of plasma regions, e.g. a positive column, that are not present in a shorter device [14, 15].

We will start by giving a short outline of the modeling procedure. Particular emphasis is put on the geometry of the PDN source used in [7–9]. Then, a brief discussion of the general results will be given, where we compare several simulation results which are obtained when the interelectrode distance is set at 1.5 mm (PDN 1), 4 mm (PDN 2)
and 10 mm (PDN 3) long, respectively. Conclusions will be drawn about the operation of the source.

11.2 The model

In this Section, we will describe the model that is used to compute the plasma in the PDN. The fundamental assumption in the model is that the plasma does not influence the flow. This is justified by the low degree of ionization that is prevalent in the PDN. It allows us to treat the flow separately, and use the results as input for the plasma calculation. We use a two-dimensional model in which it is assumed that the slit is infinitely long in the z-direction. The length of the slit in the z-direction is indeed much longer than in the other dimensions, justifying this assumption.

11.2.1 The plasma model

The model used to describe the PDN plasma in this study is basically the same as in [6]. It has also previously been used successfully to describe plasma display panel pixels [16], the ignition of compact fluorescent lamps [17] and the plasma needle [18]. A full description of the model can be found in Refs. [6, 17] and references therein. We will give a very short overview of the basic model, and refer the reader to Ref. [6, 17] for further details.

The plasma model is based on a two-dimensional time-dependent fluid approach. The conservation equations are solved in the drift-diffusion approach for six species, the electron, $e^{-}$, and four argon species, $\text{Ar}^{+}$, $\text{Ar}^{*}$, $\text{Ar}^{**}$ and $\text{Ar}^{*}_{r}$. The electrons $e^{-}$ and argon ions $\text{Ar}^{+}$ are treated as species. The four 4s levels of Ar are lumped in the $\text{Ar}^{*}$ species, which is considered metastable, because the electron impact cross section for creating a metastable 4s level is significantly larger than the cross section for creating a resonant 4s level. The second excited state, denoted $\text{Ar}^{*}_{r}$, represents the 4p manifold and higher levels. In order to account for collisional quenching of the two metastable states to the two resonant states in the 4s manifold, a third excited species $\text{Ar}^{*}_{r}$ is specified, which is destroyed by de-excitation to the ground level. The gas flow is incorporated in the conservation equations as a source term. The electric fields are obtained by solving the Poisson equation.

For the electron energy, a conservation equation that includes source terms for the energy gained in the electric field and lost in collisions is solved. The electron transport and reaction rate coefficients were obtained from an external Boltzmann solver [19], using input data from [17, 20–27].
11.2. The model

The flow influences the plasma in two ways. First, the flow is a momentum source for the species. This momentum source enters the drift-diffusion equations. This effect is particularly important for the neutral species, as there are no electric forces to modify their speed. Second, the background gas density varies significantly in the device, affecting the reaction time and the diffusion of ionic species.

The PDN flow is modeled by a separate computer program MB–CNS that can calculate transient compressible flows in two-dimensional geometries. It is based on a finite volume formulation of the Navier-Stokes equations [28, 29]. Details can be found in [30]. The bulk flow velocity in the $x$-direction at the symmetry plane of the PDN is plotted in Fig. 11.1. The Ar background density $n_{Ar}$ in the $x$-direction at the symmetry plane of the PDN is plotted in Fig. 11.2. The PDN widens at two points: at the anode and at the cathode. At both points, the density drops and the velocity increases. In the interelectrode region, the flow remains supersonic.

There is a significant difference between the argon number density $n_{Ar}$ near the cathode in PDN 1 and PDN 2 and 3, in particular off the symmetry plane.

**Figure 11.1:** The bulk flow velocity in the $x$-direction at the symmetry plane of the PDN, for a 1.5 mm interelectrode distance (PDN 1), a 4 mm interelectrode distance (PDN 2) and a 10 mm interelectrode distance (PDN 3). In all cases, the anode is positioned at $x = 0$ mm. The position of the cathode is indicated by arrows (Adapted from Ref. [30]).
Figure 11.2: The Ar background density in the x-direction at the symmetry plane of the PDN, for a 1.5 mm interelectrode distance (PDN 1), a 4 mm interelectrode distance (PDN 2) and a 10 mm interelectrode distance (PDN 3). In all cases, the anode ends at x=0 mm. The position of the cathode is indicated by arrows (Adapted from Ref. [30]).

11.2.3 The geometry

The computation is carried out on a two-dimensional Cartesian grid. In our original study with an interelectrode distance of 1.5 mm [6] (PDN 1), a grid was used that consisted of 224 by 49 grid cells that were $12.5 \times 12.5 \mu m^2$. Increasing the interelectrode distance increases the length of the device in the x-direction, and therefore also the length of the grid needed. Increasing the grid length by simply adding grid cells would result in a model that is computationally too slow to be feasible (with the present code implementation). Hence, we increase the size of the cells in the x-direction. We use 200 by 50 grid cells with a $25 \times 12.5 \mu m^2$ cell size for PDN 2 (4 mm), and 230 by 50 grid cells with a $50 \times 12.5 \mu m^2$ cell size for PDN 3 (10 mm interelectrode distance). The position of the grid in the device is schematically represented in Fig. 10.2, with the difference that the width of the spacer plate is not fixed at 1.5 mm.

11.3 Results and discussion

Using the model presented in Sec. 11.2, we simulated the pulsed discharge nozzle. By using the time-dependent equations described in Sec. 11.2 on a suitable starting condi-
11.3. Results and discussion

Figure 11.3: A schematic drawing of the geometry of the pulsed discharge nozzle. The size of the central plasma channel is exaggerated for clarity. The dashed rectangle is the part of the device that is simulated. The gas flows from a high-pressure source on the left to a vacuum chamber on the right. Note that we consider 3 situations, that differ from each other with respect to the length of the spacer plate. In PDN 1, this plate is 1.5 mm, in PDN 2, this plate is 4 mm, and in PDN 3, this plate is 10 mm (Adapted from [6]).

The plasma has a finite resistance. This means, that the voltage drop for a certain current in principle depends on the interelectrode distance. To be able to make a proper comparison between the various models, we will use a source voltage that results in the same average current density for all three cases, namely 1 mA mm$^{-2}$. At this current density, a current of 40 mA flows through the PDN.

11.3.1 General operation discussed using the results of PDN 1

The general operation of the discharge is treated in detail in [6]. We will summarize only the salient details here.

The results discussed in [6] have been obtained for a source voltage of -500 V. In order to have an average current density of 1 mA mm$^{-2}$, we will rather consider the results for -465 V, which present no essential difference with the results at -500 V.
Figure 11.4: The steady-state electron density of PDN 1 for a source voltage of -465 V. The color bar and isolines range from 0 to $4 \times 10^{18}$ m$^{-3}$ in steps of $5 \times 10^{17}$ m$^{-3}$. The bold lines indicate the different parts of the device.

The study of PDN 1, with a -465 V source voltage, revealed that the discharge acts mainly as a source of Ar$^*$, electrons $e_\text{r}$, and ions Ar$^+$. Both metastables and electron-ion pairs are mainly produced in two rather small areas of the discharge, namely near the anode and near the cathode tip, as shown in Figs. 11.4, 11.5 and 11.6. There is a difference between the electron and Ar$^+$ distribution, especially near the cathode, where there is a cathode fall. The cathode fall is characterized by a peak in the $n_{\text{Ar}^+}$, and a low $n_e$.

It is worth mentioning that the dominant formation mechanisms are different for both zones. Near the anode, where the average electron energy $\epsilon$ is about 7 eV, the dominant formation mechanism of Ar$^*$ is excitation of Ar to Ar$^*$. Near the cathode, where the average electron energy is about 40 eV the dominant mechanism of Ar$^*$ is excitation of Ar to Ar$^{**}$ and subsequent decay to Ar$^*$. Comparing Fig. 11.4 and Fig. 11.6 shows that $n_{\text{Ar}^*}$ in the discharge is far larger than $n_e$. This observation, combined with typical reaction cross sections [31, 32] for the processes under consideration and the typical electron energy calculated [6], leads to the conclusion that Ar$^*$ is the main agent in the formation of ionized PAHs.

The area immediately downstream from the cathode has a very low electron density, as is shown in Fig. 11.4. This is due to the presence of a cathode fall. The voltage drop over the cathode fall is large, accounting for the bulk of the voltage drop over the glow discharge plasma. The cathode fall is also clearly visible in Figure 11.5 as an area with a high Ar$^+$ density. The Ar$^+$ is highest near the tip of the cathode. This is because the background gas density is higher there than further upstream from the cathode.
Figure 11.5: The steady-state \( \text{Ar}^+ \) density of PDN 1 for a source voltage of -465 V. The color bar and isolines range from 0 to \( 4 \times 10^{18} \text{ m}^{-3} \) in steps of \( 5 \times 10^{17} \text{ m}^{-3} \). The bold lines indicate the different parts of the device.

Figure 11.6: The steady-state \( \text{Ar}^* \) density of PDN 1 for a source voltage of -465 V. The colorbar and isolines range from 0 to \( 9 \times 10^{19} \text{ m}^{-3} \) in steps of \( 1 \times 10^{19} \text{ m}^{-3} \). The bold lines indicate the different parts of the device.
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Figure 11.7: The steady-state electron density of the pulsed discharge nozzle with an interelectrode distance of 4 mm (PDN 2) for a source voltage of -470 V. The color bar and isolines range from 0 to $5 \times 10^{18} \text{m}^{-3}$ in steps of $5 \times 10^{17} \text{m}^{-3}$. The bold lines indicate the different parts of the device.

11.3.2 Results and discussion for the PDN 2 configuration

A source voltage of -470 V is used in the model to obtain an average current density of 1 mA mm$^{-2}$.

Although one might expect that the voltage drop over the interelectrode distance would require PDN 2 to have a significantly higher source voltage than PDN 1 to produce the same average current density, this is not the case. The Ar density near the cathode is higher for PDN 2 than for PDN 1. Because higher Ar density causes the ionization rate at a given electron temperature and electron density to be higher, the cathode operation will be more efficient in this case. This means that the voltage drop over the cathode is lower for PDN 2 than for PDN 1, and this effect largely compensates the difference in interelectrode distance.

The electron density for PDN 2 is given in Fig. 11.7. The two active areas, one near the cathode and the other near the anode, are clearly visible, and are more distinct than for PDN 1. There is a clear dent in the electron density profile near the cathode. This is caused by the cathode fall, which is the most active near the tip of the cathode.

Comparing the $n_e$ profile of PDN 2 in Fig. 11.7 with the $n_e$ profile of PDN 1 in Fig. 11.4, we see that the $n_e$ profiles in the anode region are similar.

Near the cathode, the situation is different. PDN 2 has a higher electron density near the cathode than PDN 1. This effect will be treated in more detail in the discussion of the PDN 3.
A clear difference between the $n_e$-profiles of PDN 1 and PDN 2 is that the electron density peaks in the center of the discharge in most of the interelectrode distance of PDN 2 while it peaks off the center in PDN 1. This is caused by the internal expansion of the plasma after the anode, which pushes the plasma to the wall. This process competes with the increased loss of electron-ion pairs by ambipolar diffusion near the walls. In PDN 1, the internal expansion covers a large part of the interelectrode space, while this part is much smaller in PDN 2. In this zone, the plasma density indeed peaks off the symmetry plane in both cases. In the interelectrode space of PDN 2 that lies after the expansion, the electron density indeed peaks at the symmetry plane.

The Ar$^+$ density for PDN 2 is given in Fig. 11.8. Comparing Figs. 11.7 and 11.8 shows that the plasma is close to quasineutral, except for the cathode fall. The cathode fall of PDN 2 is more strongly concentrated at the tip of the cathode than the cathode fall of PDN 1.

The Ar$^*$ density for PDN 2 is given in Fig. 11.9. Ar$^*$ is mainly produced near the anode and near the cathode and is subsequently transported downstream. While the density of Ar$^*$ is highest near the anode, the production is in fact highest near the cathode. This is caused by the fact that the bulk velocity is low near the anode, while it is much higher near the cathode. Hence, the metastables generated near the cathode are more effectively carried away, resulting in a lower density there. For spectrochemical applications, the flux plays a more important role than the actual number density.

The plasma column produces less electron-ion pairs per Ar$^*$ produced than the pro-

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**Figure 11.8:** The steady-state Ar$^+$ density of the pulsed discharge nozzle with an interelectrode distance of 4 mm (PDN 2) for a source voltage of -470 V. The color bar and isolines range from 0 to $5 \times 10^{18} \text{ m}^{-3}$ in steps of $5 \times 10^{17} \text{ m}^{-3}$. The bold lines indicate the different parts of the device.
Figure 11.9: The steady-state Ar$^*$ density of the pulsed discharge nozzle with an interelectrode distance of 4 mm (PDN 2) for a source voltage of -470 V. The colorbar and isolines range from 0 to $90 \times 10^{18} \text{ m}^{-3}$ in steps of $1 \times 10^{19} \text{ m}^{-3}$. The bold lines indicate the different parts of the device.

Production regions near the anode and cathode. Because the column is larger in a plasma with a larger interelectrode distance, fewer metastables are produced per electron-ion pair in at the larger distances.

Comparing the $n_{\text{Ar}^*}$ profile of PDN 2 in Fig. 11.9 with the $n_{\text{Ar}^*}$ profile of PDN 1 in Fig. 11.6, we see that the anode regions are similar in the two cases. The bulk of the metastables generated near the anode decay before reaching the cathode in PDN 2, causing a clear distinction between the two production zones. The Ar$^*$ density is generally somewhat lower in PDN 2 than in PDN 1. The power dissipated in PDN 1 and PDN 2 is nearly equal. Because PDN 2 produces more electrons, mainly in the plasma column, it is not surprising that there is less power available for the production of Ar$^*$, as observed. Furthermore, the average electron energy near the cathode is higher for PDN 1 (about 40 eV) than for PDN 2 (about 30 eV), because the cathode fall is larger. The reaction rates predict that the ratio between produced metastables and electrons is higher at 40 eV than at 30 eV. This means that PDN 2 should produce less metastables per electron in the cathode region, as is observed.

11.3.3 Results and discussion for the PDN 3 configuration

A model of PDN 3 has been made. A source voltage of 610 V is now used in the model to obtain an average current density of 1 mA mm$^{-2}$.

The electron density for PDN 3 is given in Fig. 11.10. The plasma structure is quite
Figure 11.10: The steady-state electron density of the pulsed discharge nozzle with an interelectrode distance of 10 mm (PDN 3) for a source voltage of -610 V. The color bar and isolines range from 0 to $4.5 \times 10^{18} \text{ m}^{-3}$ in steps of $5 \times 10^{17} \text{ m}^{-3}$. The bold lines indicate the different parts of the device.

similar to the plasma structure in PDN 2 shown in Fig. 11.7. There is an area with a high electron density at the anode, separated from the positive column. Again, the internal expansion of the gas pushes the plasma to the wall near the anode. The positive column fills the largest part of the discharge. A distinctive dent in the electron density profile near the cathode tip is visible, similar to the dent observed in PDN 1 (Fig. 11.4) and PDN 2 (Fig. 11.7). This dent is caused by the cathode fall.

The $\text{Ar}^+$ density for PDN 3 is given in Fig. 11.11. Comparing Figs. 11.10 and 11.11 shows that the plasma is close to quasineutral, except for the cathode fall. The cathode fall of PDN 3 is more strongly concentrated at the tip of the cathode than the cathode fall of PDN 1.

The $\text{Ar}^*$ density for PDN 3 is given in Fig. 11.12. The distribution of $\text{Ar}^*$ is quite similar to the distribution of $\text{Ar}^*$ in PDN 1 and PDN 2. There are two clearly separated zones with a high $\text{Ar}^*$ density, one near the anode and one near the cathode. In the positive column, $n_{\text{Ar}^*}$ is low and at both electrodes, $n_{\text{Ar}^*}$, is lower in PDN 3 than in PDN 2.

11.3.4 General discussion

The spectrochemical characteristics of the source have been examined as a function of the interelectrode distance. We have investigated the trends induced by the variation in this parameter by considering the production of the two most important species for ion
Figure 11.11: The steady-state $\text{Ar}^+$ density of the pulsed discharge nozzle with an interelectrode distance of 10 mm (PDN 3) for a source voltage of -610 V. The color bar and isolines range from 0 to $4.5 \times 10^{18}$ m$^{-3}$ in steps of $5 \times 10^{17}$ m$^{-3}$. The bold lines indicate the different parts of the device.

Figure 11.12: The steady-state $\text{Ar}^*$ density of the pulsed nozzle discharge with an interelectrode distance of 10 mm (PDN 3) for a source voltage of 610 V. The colorbar and isolines range from 0 to $90 \times 10^{18}$ m$^{-3}$ in steps of $1 \times 10^{19}$ m$^{-3}$. The bold lines indicate the different parts of the device.
yield, namely $n_e$ and $n_{Ar^+}$, at a constant average current density of 1 mA mm$^{-2}$.

The investigation of the electron and Ar$^*$ density shown in Figs. 11.4, 11.5, 11.7, 11.8, 11.10 and 11.11 reveals that for longer interelectrode distances, a quasineutral positive column forms, which is not present in PDN 1. The electron and ion density near the anode do not vary significantly with varying interelectrode distance for the range of distances investigated in this work. This is consistent with the classical glow discharge theory [14, 15]: when a glow discharge contains a positive column, the column length increases with the length of the discharge, while having little impact on the other features. This reinforces the idea suggested in [6, 12, 13] that the source operates as a glow discharge.

The presence of the positive column can also be shown in a different way. A positive column typically has a uniform electric field with a value that is small compared to the electric fields observed in the cathode and anode falls. The potential at $y = 193.75 \mu m$, which is close to the edge of the plasma at $y = 200 \mu m$, is given in Fig. 11.13 for the three different PDN configurations.

The voltage graphs in Fig. 11.13 all have two very distinct features: a cathode fall, visible as a sharp spike in the potential under the cathode tip, and an anode fall, which is visible as a rather steep voltage gradient near the anode. The spike is caused by
the voltage drop over the cathode fall, which is necessary to extract electrons from the cathode. These features do not depend strongly on the length of the device, as is to be expected from classical glow discharge theory.

Between the cathode and the anode fall, an area with a lower and constant electric field of $3 \times 10^4 \text{ V m}^{-1}$ for PDN 2 and PDN 3 is visible. Because the background density is very similar for PDN 2 and PDN 3, it is expected that the field in the positive column is the same as well. In PDN 1, this area is very short, and the field strength is somewhat lower than for PDN 2 and PDN 3. This suggests that no real positive column is formed in PDN 1, a conclusion supported by the electron density map of Fig. 11.4 and experimental evidence [12]. A positive column field of $2.6 \times 10^4 \text{ V m}^{-1}$ is large. In this particular case, the small height of the plasma causes the diffusive loss of metastables and electron-ion pairs to the wall to be very large [33, 34]. The positive column field must therefore be large to compensate these losses in order to sustain the plasma.

Increasing the interelectrode distance apparently results in the formation and lengthening of a positive column. It is expected that a further increase of the electrode distance will result in a further lengthening of the positive column.

11.4 Conclusions

We have simulated the properties of the PDN for three different interelectrode distances, namely 1.5, 4 and 10 mm, with a source voltage that generates an average current density of 1 mA mm$^{-2}$.

The flow field changes significantly when going from the experimental PDN, which has an interelectrode distance of 1.5 mm, to PDN configurations with larger interelectrode distances. In the former case, there is however no clear distinction between the expansions at the anode and at the cathode, while in the latter cases, a clear distinction is observed. The Ar density near the cathode in PDN 1 is significantly lower than the Ar density near the cathode of PDN 2 and PDN 3, which causes the cathode fall of PDN 1 to be higher than the cathode fall of PDN 2 and PDN 3.

In the PDN with a 1.5 mm interelectrode distance, the production of the species that are the most relevant for ionization of the PAH, namely Ar$^*$ and e, is highest in two distinct zones: at the cathode and at the anode.

Increasing the interelectrode distance results in a larger separation of the two production zones. Furthermore, a positive column forms, stretching from under the cathode tip to about 1 mm from the anode. This positive column has a relatively high $n_e$, but a low $n_{Ar^*}$. The formation of a positive column for longer device lengths reinforces earlier evidence that the source operates as a glow discharge.

Increasing the interelectrode distance length hence results in an increase of the area
in which the PAHs are exposed to electrons. There is no increase however in the area in which the PAHs are exposed to Ar$^*$. In fact, the Ar$^*$ density decreases slightly for increased interelectrode distance.

PAH molecules, which are the primary focus of spectrochemical research for the source under consideration have an ionization energy ($\sim 7$ eV) [35] which is higher than the typical bond dissociation energy ($\sim 3-4$ eV). Soft penning ionization by Ar$^*$ can produce PAH ions with limited fragmentation [6, 7]. However, the average electron energy is so low ($\sim 4$ eV) [6] in the positive column that far more electrons are capable of bond dissociation than of ionization. Hence, a high electron density is expected to be detrimental to the production of ionized PAHs.

A third possible reaction channel is the formation of PAH ions is by charge transfer reactions with Ar$^+$. This process, however, is likely to have a rather low reaction rate, because the thermal velocity of Ar$^+$ is low, about three orders of magnitude less than the electron thermal velocity. Furthermore, the density of Ar$^+$ is much lower than that of Ar$^*$. Quantifying this contribution requires knowledge of the charge transfer cross section between the PAH and the Ar$^+$, which to the best of our knowledge is not known for singly-ionized argon in low-energy collisions.

A quantitative analysis of the effect of varying $n_{Ar^*}$ and $n_e$ on the formation of excited PAHs would require a simulation that specifically includes PAHs as species in the model. However, the lack of data for the PAH reactions makes this task difficult for the time being.

Acknowledgments

This project is supported under the NASA Science Mission Directorate APRA program and NASA Ames DDF project. B. H. P. Broks, W. J. M. Brok and J. Remy respectively acknowledge support from the Stichting voor Fundamenteel Onderzoek der Materie (FOM), from Philips Research and from the Center for Plasma Physics and Radiation Technology (CPS).

Bibliography


Abstract—In this Chapter, we study a cascaded arc in argon that is used as a broadband light source for spectroscopic applications. In this arc, the arc channel is geometrically constricted. A numerical model is used to investigate the plasma parameters and light output of the arc. It is found that constricting leads to a higher electron density in the constricted area, which strongly enhances the local broadband emission of the plasma. A parameter study, in which the current is varied, is performed. The simulated arc voltages are compared with measured arc voltages, and an excellent agreement is found. Furthermore, it is found that the emissivity increases strongly for increasing current, making the current a suitable control parameter to control the light output of the arc.

12.1 Introduction

The cascaded arc, which has been invented by Maecker [1] in 1956, is used extensively as a source of light [2, 3] and ions [4–6]. This work is focused on the use of the cascaded arc as a light source.

A cascaded arc, filled with a noble gas such as argon or xenon, can be used to generate a dense plasma in the arc channel. Such a dense plasma produces a strong broadband emission by Bremsstrahlung and free-bound radiation, ranging from the VUV [7] to the infrared [8]. The solid angle over which this light is emitted is small due to the geometry of the arc, leading to an intense light bundle with a small entendue. This is...
favorable for imaging in many spectroscopic applications [3]. Furthermore, the high stability of the cascaded arc allows it to be used as a radiation standard in the VUV [9, 10].

The geometry of the arc has a strong influence on the light emission. A smaller arc channel at the same arc current will lead to a higher dissipation and electron density, which will result in higher wall heat loads and more emission of radiation. Burm et al. proposed an arc with a varying channel diameter to locally enhance the electron density to enhance the radical yield of a flowing arc [11–13]. In the present work, we will make a numerical analysis of the behavior of a stationary arc, in which a constricted channel is used to enhance the local electron density and light emission, as proposed in [14]. In order to increase the accuracy of the modeling, accurate and recent cross sections for the various elastic and inelastic processes are used. Furthermore, corrections for the weak plasma nonideality are made to improve the accuracy of the transport properties. The influence of multiply-ionized Ar species is also included in the model.

In this work, we will present the design of the cascaded arc that is studied in Section 12.2. In Section 12.3, the model used to describe the arc is treated in detail. In Section 12.4, selected plasma physical parameters and the light emission for one particular set of control parameters is treated in detail. In Section 12.5, results of a parameter study in which the arc current is varied are discussed, including a comparison with experimental results. In Section 12.6, general conclusions are presented.

12.2 The pinched cascaded arc

The system modeled in this work is based on the experimental setup described in [14]. In this work, the plasma in the arc channel is simulated. This part of the system is presented in Figure 12.1.

The arc consists of a stack of 7 copper plates with a width of 5 mm which are electrically insulated by PVC spacer plates with a width of 1 mm. The bore of the plates becomes smaller towards the center of the channel, leading to a constricted arc channel.

The cathode and anode are connected to a power supply which can deliver up to 30 ampere of current. The arc channel is filled with a noble gas, in this case Ar. This gas is flushed slowly (~0.5 sccm) to remove impurities from the arc channel. This slow flow has no noticeable effect on the arc behavior. The arc can operate over a range of pressures; in this work, we restrict ourselves to a working pressure of 3.3 bar.
12.3 The model

The arc model is based on the well-known two-temperature non-Local Thermal Equilibrium approach, which has been successfully used to model a flowing cascaded arc [15]. In this approach, it is assumed that all heavy particle species have the same temperature $T_h$, which may differ from the electron temperature $T_e$. The heating of the wall by the heavy particles is included in the model as well. Furthermore, deviations from chemical equilibrium are allowed, meaning that the balance between ionization and recombination can be out of equilibrium. One important difference between the model used in this work and the model in [15] is that the computation of flows is not necessary for this work and is not carried out.

For the simulations, we used the PLASIMO code. This code is described in detail in [16–18]. It is a modeling platform that can treat LTE and non-LTE plasmas, currently in two dimensions. Furthermore, its modular structure allows for easy expansion of the code. It has been applied to simulate a wide variety of plasma as described in Refs. [19–21].

In our model, the densities of four different species are simulated: Ar, Ar$^+$, Ar$^{2+}$ and e. The local densities of these species are determined by diffusion and reactions. The reaction rates are obtained using the data given in Section 12.3.1. We use the Frost mixture rules [22] to determine the transport properties of the electrons, which is the method of choice for dense plasmas with a low $T_e$, such as the plasma in the cascaded arc [23]. The elastic collision cross sections that are required as input for the computation of the transport properties are discussed in Section 12.3.2. Cool plasmas with a high electron density $n_e$ may give rise to plasma nonideality. The treatment of plasma
nonideality used in this work is presented in Section 12.3.3. The relation between the plasma parameters and the emitted continuum radiation is presented in Section 12.3.4. The grid that is used for the simulations is discussed in Section 12.3.5.

### 12.3.1 Inelastic processes

For the simulation of this plasma, three reactions are dealt with: direct ionization of Ar, stepwise ionization of Ar, and direct ionization of Ar$^+$. These will be described in this Section.

The balance between direct ionization of Ar and the backward 2-electron recombination is given by

$$\text{Ar} + e \rightleftharpoons \text{Ar}^+ + e + e. \quad (12.1)$$

The forward reaction rate $k^f_{\text{dir}}$ of this process can be obtained by convoluting the ionization cross section for this reaction with the electron energy distribution function (EEDF) of the plasma. This EEDF can be approximated well with a Maxwell-Boltzmann distribution due to the high plasma density and resulting frequent electron-electron collisions. The ionization cross sections are obtained from [24, 25], resulting in a $k^f_{\text{dir}}$ that is given by:

$$k^f_{\text{dir}} = 2.61 \times 10^{-17} T_e [K]^{0.685} \exp \left( -\frac{E_{\text{Ar}^+}}{k_B T_e} \right) \text{m}^3 \text{s}^{-1}, \quad (12.2)$$

with $E_{\text{Ar}^+}$ the ionization energy of argon of 15.8 eV and $k_B$ Boltzmann’s constant. The backward process is obtained by using detailed balancing.

A second contribution to the production of Ar$^+$ is the stepwise ionization of Ar via the first excited state Ar$^*$. Together with the corresponding reverse process they can be represented by:

$$\text{Ar} + e \rightleftharpoons \text{Ar}^* + e \rightleftharpoons \text{Ar}^+ + e + e \quad (12.3)$$

The energy gaps at the top of the atomic system are of the same magnitude as, or smaller than, the typical electron energy in the arc, causing fast reactions between the excited states and the ion state [26]. Hence, the excitation of Ar is the rate-determining step, with a rate $k^f_{\text{step}}$ that is given by [26]

$$k^f_{\text{step}} = 6.79 \times 10^{-17} T_e [K]^{0.5} \exp \left( -\frac{E_{\text{Ar}^*}}{k_B T_e} \right) \text{m}^3 \text{s}^{-1}, \quad (12.4)$$

where $E_{\text{Ar}^*}$ is the excitation energy of 12.1 eV. The rate of the reverse process is obtained using detailed balancing.

The creation and destruction of Ar$^{2+}$ is incorporated in the model by the direct electron impact ionization of Ar$^+$ and the corresponding backward process:
Table 12.1: The cross section models used to compute the transport parameters in the plasma.

<table>
<thead>
<tr>
<th>Interaction</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ar + Ar</td>
<td>VHS-theory [28]</td>
</tr>
<tr>
<td>Ar + Ar^+</td>
<td>Elastic + Charge transfer [29]</td>
</tr>
<tr>
<td>Ar + Ar^{2+}</td>
<td>Elastic + Charge transfer [29]</td>
</tr>
<tr>
<td>Ar + e</td>
<td>[30, 31]</td>
</tr>
<tr>
<td>Ar^+ + Ar^+</td>
<td>Coulomb (see Sec. 12.3.3)</td>
</tr>
<tr>
<td>Ar^+ + Ar^{2+}</td>
<td>Coulomb (see Sec. 12.3.3)</td>
</tr>
<tr>
<td>Ar^+ + e</td>
<td>Coulomb (see Sec. 12.3.3)</td>
</tr>
<tr>
<td>Ar^{2+} + Ar^{2+}</td>
<td>Coulomb (see Sec. 12.3.3)</td>
</tr>
<tr>
<td>Ar^{2+} + e</td>
<td>Coulomb (see Sec. 12.3.3)</td>
</tr>
<tr>
<td>e + e</td>
<td>Coulomb (see Sec. 12.3.3)</td>
</tr>
</tbody>
</table>

\[
\text{Ar}^+ + e \rightleftharpoons \text{Ar}^{2+} + e + e. \tag{12.5}
\]

The rate \( k_{\text{double}} \) for this process is obtained using cross section data from [27]:

\[
k_{\text{double}}^f = 2.82 \times 10^{-17} T_e [K]^{0.671} \exp \left( \frac{-E_{\text{double}}}{k_B T_e} \right) \text{ m}^3 \text{ s}^{-1}. \tag{12.6}
\]

Here, \( E_{\text{double}} \) is the energy difference between \( \text{Ar}^+ \) and \( \text{Ar}^{2+} \), which amounts to 27.6 eV.

12.3.2 Elastic collisions

The four species in the plasma allow for ten different binary interactions. An overview of these reactions is given in Table 12.1 Six of these are interactions between charged particles, which can be treated with standard Coulomb theory, modified with corrections for plasma nonideality. These corrections will be discussed in Section 12.3.3. For the other four interactions, a detailed description is given in this Section.

The interaction of \( \text{Ar} \) with \( \text{Ar} \) is modeled with the variable hard sphere (VHS) model of Bird [28]. This model is an extension of the standard hard-sphere cross section model that includes an energy dependence of the cross section. The \( \text{Ar-Ar} \) collision cross section \( \sigma(\text{Ar, Ar}) \) is given by [28]

\[
\sigma(\text{Ar, Ar}) = 3.43 \times 10^{-18} E[K]^{-0.31} \text{ m}^2. \tag{12.7}
\]

Two possible interactions between \( \text{Ar} \) and \( \text{Ar}^+ \) that are included in the model are elastic interactions and charge exchange, which can be thought of as the transfer of an
electron from Ar to Ar\(^{+}\), effectively transferring momentum from the neutral species to the ions and vice-versa. Both processes are included in the model; the effective total Ar-Ar\(^{+}\) cross section \(\sigma(\text{Ar, Ar}^{+})\) is given by [29]:

\[
\sigma(\text{Ar, Ar}^{+}) = 2.93 \times 10^{-18} E[K]^{-0.1} \left(1 + \frac{174}{E[K]}\right)^{0.6} m^2. \tag{12.8}
\]

For Ar and Ar\(^{2+}\), elastic interactions and charge exchange are possible as well. For this process, the combined rate \(\sigma(\text{Ar, Ar}^{2+})\) is given by [29] and is equal to:

\[
\sigma(\text{Ar, Ar}^{2+}) = 6.03 \times 10^{-17} E[K]^{-0.1} \left(1 + \frac{E[K]}{1.16 \times 10^5}\right)^{0.28} m^2. \tag{12.9}
\]

The elastic cross section of Ar and \(e\), \(\sigma(\text{Ar, e})\), is of particular importance, because electrons are the dominant particles for many transport phenomena, such as heat transport and electrical conductivity. Due to the Ramsauer-Townsend effect [32], \(\sigma(\text{Ar, e})\) varies strongly as a function of the interaction energy. Cross section data for \(\sigma(\text{Ar, e})\) is obtained from [30] for the energy range between 0.02-19 eV and from [31] for the energy range between 19-54 eV.

### 12.3.3 Plasma nonideality

A cornerstone of the theory of transport in plasmas is the assumption that the plasma is ideal, i.e. the number of electrons in a Debye sphere \(N_{\Lambda D}\) is much greater than 1 [33]:

\[
N_{\Lambda D} = \frac{1}{9} \Lambda C \gg 1. \tag{12.10}
\]

where the parameter \(\Lambda C\) is given by

\[
\Lambda C = \frac{12\pi (\epsilon_0 k_B T_e)^{\frac{3}{2}}}{3e^3 n_e^{\frac{1}{2}}}. \tag{12.11}
\]

The condition in (12.10) condition is not well satisfied for relatively cool, dense plasmas, such as the cascaded arc that is simulated in this work, as can be seen in (12.11). It is, however, possible, to modify the treatment of Coulomb collisions so that is also valid for weakly nonideal plasmas, which are plasmas for which the relation

\[
N_{\Lambda D} = \frac{1}{9} \Lambda C \gtrsim 1. \tag{12.12}
\]

is fulfilled. The validity of (12.12) will be verified in sec. 12.4.

The effects of weak plasma nonideality can be incorporated using a modified Coulomb logarithm. Based on an analytical curve fit of numerical solutions of the Fokker-Planck
12.3. The model

Ordonez and Molina [34] propose the following modified Coulomb logarithm $\lambda_C$:

$$\lambda_C = \ln(0.6\Lambda_C) = \ln \Lambda_C - 0.51.$$  \hspace{1cm} (12.13)

The curve fit differs by less than 4% of the numerical solution for $\lambda_C \gtrsim 2$, offering a very simple yet highly accurate method to treat weak plasma nonideality.

12.3.4 Continuum radiation

The plasma is used as a source of continuum radiation, with emissivity $\varepsilon$. This continuum radiation is composed of free-free radiation, with emissivity $\varepsilon_{ff}$, and free-bound radiation, with emissivity $\varepsilon_{fb}$ [33, 35, 36]:

$$\varepsilon = \varepsilon_{fb} + \varepsilon_{ff}.$$  \hspace{1cm} (12.14)

The plasma can be considered optically thin for continuum radiation, and as the amount of energy lost in radiation is typically only a few percent of the total input power, it is not necessary to use full radiation transport treatment, or to include the radiation losses in the energy balance equations. Instead, the radiation output is computed as a post-processing step from the primary plasma properties, by evaluating (12.14).

The free-free and free-bound contribution to $\varepsilon$ are given by [33, 35, 36]:

$$\varepsilon_{ff}^i = \frac{C_1 (n_{Ar^+} + 4n_{Ar^{2+}}) n_e}{\lambda^2 \sqrt{T_e}} \exp \left( - \frac{hc}{\lambda k_B T_e} \right) \tilde{\xi}_{ff}(\lambda, T_e)$$  \hspace{1cm} (12.15)

$$\varepsilon_{fb} = \frac{C_1 (n_{Ar^+} + 4n_{Ar^{2+}}) n_e}{\lambda^2 \sqrt{T_e}} \left[ 1 - \exp \left( - \frac{hc}{\lambda k_B T_e} \right) \right] \tilde{\xi}_{fb}(\lambda, T_e)$$  \hspace{1cm} (12.16)

in which $C_1 = 1.5 \times 10^{-45}$ Wm$^4$eV$^{1/2}$sr$^{-1}$, $n_e$ the electron density, $\lambda$ the wavelength, $k_B$ Boltzmann’s constant, $h$ Planck’s constant, and $c$ the speed of light. The free-free Biberman factor $\tilde{\xi}_{ff}$ and the free-bound Biberman factor $\tilde{\xi}_{fb}$ are both of order unity. The form of (12.15) and (12.16) allows for a considerable simplification:

$$\varepsilon = \frac{C_1 (n_{Ar^+} + 4n_{Ar^{2+}}) n_e}{\lambda^2 \sqrt{T_e}} \tilde{\xi}(\lambda, T_e)$$  \hspace{1cm} (12.17)

with $\tilde{\xi}(\lambda, T_e)$ the Biberman factor which is given by:

$$\tilde{\xi} = \tilde{\xi}_{total} = \tilde{\xi}_{ff} \exp \left[ - \frac{hc}{\lambda k_B T_e} \right] + \tilde{\xi}_{fb} \left[ 1 - \exp \left( - \frac{hc}{\lambda k_B T_e} \right) \right].$$  \hspace{1cm} (12.18)

Equation (12.17) clearly shows the trend of the plasma emission as a function of the plasma properties $T_e$, $n_e$, $Ar^+$ and $Ar^{2+}$. To obtain quantitative results, the average value $\tilde{\xi}_{ff} = 1.23$ from [3] is substituted, and data for $\tilde{\xi}_{fb}$ is obtained from [37].
The cascaded arc consists of a stack of plates with bores that have a varying diameter, leaving an arc channel that has a radius that varies with discrete steps (viz. Fig. 12.1). Although it is possible to simulate such a stepped geometry, we used a different approach, in which the stepped diameter of the arc is approximated with a smoothly varying function for the arc channel radius for two reasons:

- A smooth grid considerably simplifies the computation. In particular, resolving the sharp edges and small structures in the wall would require a very fine grid, leading to a much longer computational time.
- Erosion of the arc channel is likely to smooth the corners of the plates, which causes the actual arc channel to resemble the smoothed channel more closely.

The computational domain and the actual shape of the arc channel are presented in Figure 12.2. An ortho-curvilinear grid [38, 39] is used to discretize the equations on the computational domain. The implementation of this method in PLASIMO is described in more detail in [16, 40, 41].

12.4 Results and detailed discussion for \( I = 30 \, \text{A} \)

The model of the cascaded arc, as presented in Section 12.3, is run for \( I = 30 \, \text{A} \). Selected results, such as the electron density \( n_e \) profile, the electron temperature \( T_e \) profile and the heavy particle temperature \( T_h \) profile, are discussed to analyze the behavior of the cascaded arc. The Coulomb logarithm is discussed in view of the weak plasma nonideality. The influence of \( \text{Ar}^{2+} \)-ions is discussed as well.

The electron density is presented in Figure 12.3. It is clear that \( n_e \) is strongly peaked in the pinched area of the arc. This can be readily explained by the much higher dissipation density in the pinched area. The dissipation density \( P \) is given by

\[
P = \frac{J^2}{\sigma} \tag{12.19}
\]

with \( J \) the current density and \( \sigma \) the conductivity. As the total current through the arc is constant, \( J \) is much higher in the pinch, leading to a much higher local dissipation density and hence to a much higher electron density.

The electron temperature is presented in Figure 12.4. \( T_e \) is also peaked in the pinched area, however, much less so than \( n_e \). This can be explained by noting that the plasma is close to Saha equilibrium. For an argon plasma, the Saha relation for \( n_e \) is given by

\[
n_e = \sqrt{12n_{\text{Ar}}} \left( \frac{\hbar^2}{2\pi m_e k_B T_e} \right)^{-\frac{3}{2}} \exp \left( \frac{-E_{\text{Ar}^+}}{k_B T_e} \right) \tag{12.20}
\]
12.4. Results and detailed discussion for $I = 30\, \text{A}$

Figure 12.2: The actual geometry and the computational domain. The volume of the computational domain differs by less than 0.5% from the volume of the actual arc channel. Note that this picture is not to scale: the length of the arc is larger by more than an order of magnitude than its radius. Note that the exact geometry differs from the geometry used in [14].

with $n_{\text{Ar}}$ the Ar density, $h$ Planck’s constant, $k_B$ Boltzmann’s constant, and $m_e$ the electron mass. Equation (12.20) shows that $n_e$ depends exponentially on $T_e$, which means that $T_e$ should increase with increasing $n_e$, but much less strongly, as is observed.

Fig. 12.4 also shows that $T_e$ remains constant near the walls. This can be explained by noting that the electrons do not get cooled directly by the wall. Rather, the electrons transfer their heat by elastic collisions to the heavy particles, and these get cooled at the wall. The ionization degree near the wall is low, which means that Coulomb collisions with a large cross section become less frequent, while electron-neutral collisions, which have a smaller cross section, become more frequent. This causes a lower collision frequency between electrons and heavy particles, and hence small elastic heat losses. This, combined with a high electron thermal conductivity and electron heat production by three-particle recombination cause the electron temperature profile to be rather flat at
The electron density $n_e$ in the pinched cascaded arc with $I = 30\, A$ and $p = 3.3\, \text{bar}$. The electron density is strongly peaked in the pinched area of the arc.

The heavy particle temperature is presented in Figure 12.5. Except near the walls, $T_h$ is nearly equal to $T_e$. The high ionization degree causes very frequent collisions between electrons and heavy particles, and hence an efficient elastic energy transfer between them. This allows for only small differences between $T_e$ and $T_h$. However, near the wall, the ionization degree is lower (cf. Fig. 12.3), and consequently, the elastic energy transfer is much smaller, allowing for larger differences in temperature, as the heavy particles are cooled efficiently at the wall.

The combination of Fig. 12.3, 12.4 and 12.5 indicate that the cascaded arc is a dense plasma, that is close to local thermal equilibrium in the center, but shows some deviations from local thermal equilibrium near the wall, illustrating the usefulness of a non-LTE treatment. Similar deviations from local thermal equilibrium are also found by Janssen [15] for a flowing straight cascaded arc with comparable parameters.

As discussed in Section 12.3, we correct for plasma nonideality under the assumption that the plasma is only weakly nonideal, i.e. (12.12) should be valid. A graph of $N_{\Lambda D}$ is presented in Figure 12.6. The lowest $N_{\Lambda D}$ with a value of 2.7 is found in the pinch, where $n_e$ is highest. Hence, our treatment of nonideality (cf. Eq. (12.13)) is applicable. The resulting $\ln \Lambda_C$ has a value of about 4, which means that the results of this treatment of nonideality differs by about 15% from the results of the standard treatment.

The density of $\text{Ar}^{2+}$ in the plasma is given in Figure 12.7. $n_{\text{Ar}^{2+}}$ is more than three orders of magnitudes lower than $n_e$ (cf. Fig. 12.3) and $n_{\text{Ar}^+}$, and is hence expected to have a negligible influence on the plasma behavior. $n_{\text{Ar}^{2+}}$ is highest in the pinch, where $T_e$ and $\text{Ar}^+$ are highest, but even there, the $T_e$ of 1.6 eV is too low to significantly ionize...
12.4. Results and detailed discussion for $I = 30\, \text{A}$

**Figure 12.4:** The electron temperature $T_e$ in the pinched cascaded arc with $I = 30\, \text{A}$ and $p = 3.3\, \text{bar}$. The electron temperature is peaked in the pinched area of the arc, and remains constant near the walls.

**Figure 12.5:** The heavy particle temperature $T_h$ in the pinched cascaded arc with $I = 30\, \text{A}$ and $p = 3.3\, \text{bar}$. The heavy particle temperature is peaked in the pinched area of the arc, and drops off sharply near the walls.
Figure 12.6: The number of particles in the Debye sphere $N_{\Lambda_D}$ in the pinched cascaded arc with $I = 30 \text{ A}$ and $p = 3.3 \text{ bar}$. $N_{\Lambda_D}$ is lowest in the pinch, and increases strongly towards the walls.

Figure 12.7: The $\text{Ar}^{2+}$ density in the cascaded arc. $n_{\text{Ar}^{2+}}$ is much lower than $n_e$. Furthermore, the density is strongly peaked in the pinch.
12.5 A current parameter study

Figure 12.8: The emissivity at 300 nm. The emissivity is highest near the axis of the channel, and strongly peaked in the pinch.

Ar$^+$ to Ar$^{2+}$, which requires an ionization energy of 27.6 eV.

For the application, the light output, particularly the UV output in the pinched area, is of great importance. The emissivity at 300 nm $\varepsilon_{300}$ is presented in Figure 12.8, which shows that $\varepsilon_{300}$ is highest in center of the channel, in particular in the pinch. The pinching increases $\varepsilon_{300}$ with more than a factor of 2. According to (12.17), $\varepsilon_{300}$ increases with the square of $n_e$, as the influence of Ar$^{2+}$ is negligible. As $n_e$ is highest in the pinch (cf. Fig. 12.3), $\varepsilon_{300}$ is highest here as well.

12.5 A current parameter study

In order to explore the parameter range over which the pinched cascaded arc can be operated, a parameter study in which the current is varied from 10 to 30 A is presented. In order to verify the correctness and accuracy of the model, the resulting operating voltage is compared to experimental arc voltages. Furthermore, we will present the peak emissivities of the channel at selected operating currents, in order to characterize the light output.

The voltage-current characteristic of the arc is given in Figure 12.9. The agreement between experiment and simulations is excellent. Small discrepancies can be caused by the cathode and anode falls of the plasma, which are not included in the model. As the plasma voltage depends on an amalgam of all key plasma properties—temperature, electron, ion and neutral density—an excellent prediction of the voltage over such a wide parameter range is strong evidence for the validity of the model.
The spectra for selected arc currents, at the area of the highest emissivity, are presented in Figure 12.10. It is observed that the emissivity increases with increasing current. The increase in current leads to an increase in power dissipation (cf. Fig. 12.9), and hence an increase in electron density, which leads to an increase in emissivity (cf. (12.17)). Furthermore, the wavelength at which the emissivity is highest becomes shorter for higher currents, due to the higher electron temperature in the plasma [37]. Figure 12.10 shows that the current is an effective parameter to control the intensity of the plasma emission.

Figure 12.10 shows that for $I = 30$ A, the emissivity peaks in the ultra-violet at 300 nm. For a black-body radiator, the wavelength with the highest emissivity $\lambda_{\text{max}}$ is predicted by Wien’s law [42],

$$\lambda_{\text{max}} T = k_w$$  \hspace{1cm} (12.21)

with Wien’s constant $k_w = 2.8978 \times 10^{-3}$ m K. Substituting the peak temperature in the cascaded arc for $I = 30$ A of $T = 1.6$ eV in (12.21), we find that $\lambda_{\text{max}} = 163$ nm. However, the observed maximum emission power lies at a wavelength of 300 nm, which implies the pinched cascaded arc is not a black body radiator.

### 12.6 Conclusions

A two-dimensional non-LTE model is used to simulate a pinched cascaded arc light source. The accuracy of this modeling is verified by comparing the voltage-current characteristic resulting from these simulations with one that is obtained experimentally. Excellent agreement was found, suggesting that the model is accurate.

The electron density is peaked in the pinched area of the plasma, leading to a peak in the emissivity there. This demonstrates the usefulness of pinching the arc to locally enhance the emissivity.

The plasma is hottest in the pinched area. While the electron and heavy particle temperatures are nearly identical in the center of the discharge, there are significant differences in temperature near the walls, underscoring the need for a non-LTE treatment.

A parameter study of the current revealed that the emissivity increases strongly with increasing current. Furthermore, the peak emissivity shifts slightly to the ultraviolet for increasing current. The current is an effective parameter to control the light emission of the plasma.
12.6. Conclusions

Figure 12.9: The measured and simulated voltage-current characteristics of the pinched cascaded arc for an operating pressure of 3.3 bar. The match between theory and experiment is excellent. The measurement data are courtesy of R.A.B. Zijlmans.

Figure 12.10: The simulated spectra of the pinched arc for selected currents, for an operating pressure of 3.3 bar. The emissivity increases for increasing current, in particular for shorter wavelengths. Furthermore, wavelength at which the emissivity is highest becomes shorter for higher currents.
Acknowledgments

This Chapter is based on a joint effort by the author of this thesis and his trainee student J.G. Keizer. R.A.B. Zijlmans is gratefully acknowledged for the measurement data provided, and for fruitful discussions. D.C. Schram is acknowledged for the fruitful discussions.

Bibliography


Chapter 13

General Conclusions

13.1 Introduction

As the title of this work indicates, the goal of this work is twofold: to test and extend a general modeling platform for the simulation of transient plasmas, and to apply this framework to plasmas that are used in the generation and guiding of light.

The modeling framework used in this thesis is based on two different codes: PLASIMO (see Section 1.3.1) and MD2D (see Section 1.3.2). The former code is applicable to dense, flowing, quasineutral plasmas, while the latter code is more suitable to less dense plasmas that do not flow, but do exhibit strong deviations from quasineutrality. In both codes, a multi-fluid approach, in which chemical nonequilibrium is assumed, plays a key role.

The applications of this framework include the simulation of various plasma sources. The results of these simulations have been verified by comparing them to experimental results. Using such a wide variety of different test cases is an excellent way of construction and testing a plasma simulation platform that is desired to have a wide range of applicability.

In this Chapter, we will first present conclusions about the modeling framework in Section 13.2. Then, the conclusions from the different numerical case studies will be presented in Section 13.2, with an emphasis on the applications. With this well-validated platform, it becomes possible to go beyond merely reproducing experimental results and gaining physical understanding. Rather, it becomes possible to make well-supported predictions with regards to systems that are not yet experimentally investigated. This Chapter is closed with suggestions for further research, which will be presented in Section 13.4.
13.2 The modeling platform

The codes that were available at the start of this project were the result of years of concentrated effort by various programmers. However, the simulation of plasmas that have durations in the nanosecond range required significant improvements and additions to the codes that were used. A further challenge was presented by the fact that some of the discharges had a very high power density, yet still did not approach local thermal equilibrium at the timescales involved.

The multi-fluid approach was verified using experimental data for the wide variety of plasmas, such as a terawatt laser waveguide, a photoconductively switched spark gap, a spectrochemical plasma source, and a cascaded arc light source. In particular, it revealed a general trend in intense, pulsed plasmas: During the ionization of these plasmas, the electron temperature initially drops as the ionization degree rises. This behavior is opposite to the behavior that would be predicted by a single-fluid model. This underscores the necessity of a multi-fluid approach.

The extendibility and modularity of the framework proved to be a critical feature for this work, allowing for several improvements to be implemented. The key improvements are:

- The implementation of chemistry source terms and transport coefficients for three different gases, using a novel, reusable cross section implementation.
- An accurate treatment for weak plasma nonideality, improving the accuracy of the computed transport coefficients by up to 30%.
- A multiregion method for the treatment of highly localized and transient wall heating, allowing for efficient and accurate simulation of the heat transport in plasma and wall.
- A correct treatment of the energy source terms for dissociative recombination and similar processes.
- The ability to parse discontinuous, time-dependent input currents or powers.
- More fine-grained control over the numerical time steps.

The numerical studies are augmented by theoretical work. This includes a derivation of the conservation equations that are the heart of the numerical method used, from the fundamental Boltzmann Transport Equation, in Chapter 2. A derivation and interpretation of the resulting equations is provided. These results are then used to construct a multi-fluid description. Two salient numerical challenges in this work, namely the different time time scales at which various plasma processes occur and the different length
scales that exist in the plasma wall system, are treated and more detail, and the strategy to overcome these challenges is discussed.

The models made with the PLASIMO and MD2D models that are used in this work can be seen as examples of *grand models*: Large, complex, extendible codes, that yield accurate results, but require expertise to use while the computational costs are substantial. Such models can be complemented by simple, easy-to-use models, that yield accurate estimates of the key plasma parameters. In this work, we present such a model in Chapter 4. This model is based on the method of disturbed Bilateral Relations (dBR) and can be used to make fairly accurate predictions of plasma properties using a minimum of computational time, and a minimum of assumptions, for various degrees of nonequilibrium. Because the dBR method allows various degrees of departure from equilibrium, it has a wide range of applicability, wider than that of most global models [1]. It was found that, for simple test cases, the results of this model match well with that of the grand model PLASIMO well, making it a suitable code to perform a rapid analysis of a large variety of plasmas.

### 13.3 The case studies

The modeling platform has been used to simulate slow pulsed capillary discharges. These discharges have a convex index of refraction profile, which is caused by a concave electron density profile, making them suitable for the guiding of terawatt laser beams [2]. The first investigation, which is presented in Chapter 5, explores the physics of the capillary and verifies the correctness and accuracy of the model, by comparing simulation results with measurements from an experimental device [2]. It was found that the model is suitable for the simulation of pulsed capillary discharge waveguides, especially when the liberation of hydrogen from the capillary wall is taken into account. Furthermore, it was found that there exists an optimal range for the discharge current: too low currents may lead to incomplete ionization of the central plasma channel, while too high currents may lead to wall ablation. Both are undesirable for the application.

This model is then used in Chapter 6 to perform a parameter study on the capillary discharge waveguide, in which both radius \( r_c \) and filling density were varied. It was found that the channel radius \( r_c \) has a significant influence on the guiding properties, with \( W_M \propto r_c^{-0.56} \). Furthermore, it was found that the current should be adjusted with the radius to avoid incomplete ionization and ablation. The filling density \( n_i \) was found to have a modest influence on \( W_M \), with \( W_M \propto n_i^{0.25} \). Because the acceleration properties depend on the on-axis density and the laser power [3], the acceleration properties can be tailored well using the channel parameters. There is, however, a minimum density at a certain radius to prevent diffusion from destroying the needed concave electron
density profile. The results of the parameter studies in Chapters 5 and 6 can be cast into a single, empirical equation for the guiding properties as a function of external parameters, which can be used to predict the laser guiding properties of capillaries with given control parameters. This is very useful in the design of practical slow pulsed capillary discharge waveguides.

The fact that the channel radius strongly influences the guiding properties suggests that modulating the channel radius can be used to locally tailor the guiding properties. Based on this suggestion, a model of a channel with a modulated radius is made in Chapter 7 to predict the guiding behavior of the channel. The model is based on the well-validated models used in Chapter 5 and 6. We can also investigate the robustness of the plasma and guiding properties with respect to surface roughness using the same method. It was found that modulating the channel radius did not modulate the guiding properties significantly, but does promote wall ablation. Hence, we predict that periodic radius modulation is a poor technique to locally enhance the laser intensity.

As an alternative to axial electron density measurements, transverse interferometry measurements have been made [4] on pulsed capillary discharge waveguides to determine the electron density profile and hence the guiding properties. In order to translate the experimental results from the square channels to a prediction of the behavior of the more common round channel, a the results of numerical model of this square channel have been compared to the results for a round channel in Chapter 8. An excellent agreement between the experimental and theoretical results has been obtained, far better than that of earlier work by Bobrova et al. [4, 5]. It was found that the plasma and guiding properties in a square waveguide resemble those in a round waveguide with a radius slightly larger than the width of the capillary. Furthermore, the heating of the wall was found not have a significant influence on the interferometry measurements. The results from the simulations can be used to extrapolate a probable 2D density profile from the 1D measured profile.

A novel, fast method of switching large voltages is the photoconductively switched spark gap. By using a terawatt laser to create a plasma in the gap, this device can switch voltages in the kV range with sub-100 ps jitter. It was experimentally found that voltages significantly below the self-breakdown threshold were not switched fully, instead losing a large fraction over the gap. In Chapter 9 we have investigated the cause of this by analytical and numerical modeling. It was found that the finite plasma resistance is the most probable cause of the observed voltage drop. It is thought that in particular the regions near the electrodes, where the laser intensity is lower, may contribute significantly to the observed finite resistance. Furthermore, it was found that even at the short timescale (2 ns), the plasma undergoes a significant chemical conversion from molecular to atomic species, underscoring the importance of plasma processes for the operation of the spark gap.
At NASA Ames, a new spectrochemical device for the analysis of organic molecules in the interstellar medium has been created. This has helped solving an 80-year old mystery in astronomy, namely the cause of the so-called Diffuse Interstellar Bands [6] that are present in the interstellar spectrum. It was revealed that Polycyclic Aromatic Hydrocarbons (PAHs) and their cations are an important cause of these bands [7].

In Chapter 10, a model of this device is presented. It was found that the discharge acts like an expanding glow discharge. No plasma column is present, and the background gas is not heated significantly. The source acts as a source of metastables, which can create excited, but ro-vibrationally cold PAH, in a state that resembles their state in interstellar space. A comparison of the simulated voltage of the device and the measured voltage revealed an excellent match for a wide range of operating currents. Because the voltage depends on all plasma properties, this inspires confidence in the simulation results.

Using this basic model, a proposal was investigated whether lengthening the source increased the yield of spectrochemically relevant species in Chapter 11. Based on this study, we predict that this is not the case. Lengthening did cause the formation of a positive column, where the high electron density may lead to fragmentation of the PAH, which will likely even reduce the yield of the desired analyte, namely cold, excited PAHs.

Cascaded arcs are used among others as sources of intense, broadband light. In Chapter 12, we have modeled a geometrically pinched cascaded argon arc that is used as a light source. It was found that the pinching leads to a strong local enhancement of the emissivity, which is desirable for the applications. Furthermore, a parameter study of the arc current was performed. It was found that the emissivity increases strongly with increasing current, making the current a good control parameter of the plasma emissivity.

The parameter study of the current also yielded arc voltages, which are compared with experimental arc voltages. This revealed an excellent agreement, which is evidence for the correctness of the model. As essentially the same model is used for the pulsed capillary discharge waveguide and the spark gap, where the experimental data is more scarce and less reliable, this finding supports the conclusions drawn and the predictions made for these applications as well.

## 13.4 Outlook

The modeling platform, based on a multi-fluid approach, that is presented in this work, is thoroughly tested and found suitable for the simulation of the wide variety of transient plasmas. There are several other interesting cases that are related to the research
presented in this thesis that can be treated with the same modeling platform. These include:

- The pulsed capillary discharge waveguide is doped with other gases such as xenon for high-harmonics generation. Simulation of this device should be possible with the present modeling platform, provided xenon chemistry is added. The work of Keizer [8] on xenon cross sections can be used as a basis for this.

- Magnetization can be used to influence the transport properties in the pulsed capillary discharge waveguide, in particular for low-density discharges [9]. A theoretical prediction of these guiding properties would be useful to complement experimental studies.

- The plasma efflux from the ends of the pulsed capillary discharge waveguide has the potential to disturb the laser guiding properties. This effect could be modeled using the two-dimensional model of the waveguide.

- Tapered waveguides are used to achieve better phase matching in the waveguide. Using the model presented in this work, the plasma in such a waveguide may be analyzed and the guiding and phase matching properties could be optimized.

- A detailed analysis of the photoconductively switched spark gap was not possible due to the poorly known initial conditions. If more accurate initial conditions become available, more accurate modeling should be possible. This may require two- or even three-dimensional modeling. Quantitative significance could also be improved by taking into account vibrational excitation of nitrogen. This can be done with limited computational cost by lumping together several vibrational levels.

- The Pulsed Discharge Nozzle has been simulated without including the particles that are analyzed, the PAHs. While the present studies have provided useful insight on the behavior of this device, its operation as a source of cold, excited PAHs is best assessed when these are included in the model as well.

- The Pulsed Discharge Nozzle has been simulated using a Boltzmann solver for the determination of the rate coefficients in the plasma. A more accurate model may be obtained by hybrid modeling.

- The cascaded arc that is simulated is stationary. It is possible to reach higher power densities and hence a higher output by a pulsed operation of the source using a higher current density. The numerical model is suitable to predict the light output of the arc in this operation regime, without risk of damaging the experimental apparatus.
• The cascaded arc may also be operated using xenon as an operating gas. This will increase the light output. Initial studies of this problem are made by Keizer [8].

Bibliography


Summary

In this work, a multi-fluid plasma modeling framework is used to model several transient plasmas with uses in the generation and guiding of light. By using a single framework for all these applications, we can make optimal use of the commonalities that exist between these plasmas, which increases the reliability and accuracy of the model while greatly reducing the development time. The plasmas that are modeled using this treatment are a waveguide for terawatt lasers, an ultrafast spark gap switch, a pulsed nozzle discharge that can emulate the conditions that exist in outer space, and an intense cascaded arc light source.

One unifying characteristic of all the plasmas in this work is that they are fluids. This allows us to use integrated moments of the general particle transport equation, the Boltzmann Transport Equation, to describe the plasma. Within this common theme, there are many variations, such as plasma density, duration, chemical composition and power density. Only a flexible, extendible model that allows for large deviations of equilibrium can treat such a diverse set of plasmas.

Terawatt laser waveguides are used to increase the interaction length between the laser beam and matter. These interactions lead to spectacular phenomena, such as particle acceleration with a high acceleration gradient and emission of photons in the extreme ultra-violet or soft X-ray regime. We have used the modeling framework to describe one type of waveguide, the pulsed capillary discharge waveguide, in which a dense hydrogen plasma guides the laser. The first modeling performed was on an experimental capillary discharge, to explain the physical phenomena observed and to verify the quality of the model from the match between theory and experiment. The next step was a parameter study of this waveguide, in which an empirical formula describing the laser guiding behavior was derived as a function of the control parameters. The proposal to use a waveguide with a modulated radius was investigated, which was thought to locally enhance the laser intensity. It was found that this was ineffective, and that a channel with a modulated radius is quite susceptible to wall ablation. Finally, a novel, experimental capillary with a square instead of a round cross section was modeled. These capillaries are used in accurate plasma density measurements, excellent agreement between the modeling and these measurements was found. These
simulations are of great use for the interpretation of the experimental data.

A terawatt laser pulse can also be used to close an electric spark gap, leading to ultra-fast switching. It is experimentally found, however, that not all the voltage over this gap is switched, in particular not for lower voltages. We have modeled the spark gap analytically and numerically to explain this voltage drop, and it was found that the finite conductivity of the plasma arc that connects the electrodes is the cause. Furthermore, insight was gained in the behavior of this strongly transient plasma.

Diffuse interstellar bands are absorption lines in the interstellar spectrum that are somewhat more diffuse and broad than normal absorption lines. These features have defied explanation for almost a century. Recently, a novel experimental apparatus was constructed at NASA Ames to investigate a possible cause of these bands, namely Polycyclic Aromatic Hydrocarbons (PAHs) and their cations. This apparatus, called the Pulsed Discharge Nozzle (PDN) is capable of generating excited and ionized PAHs in an environment that resembles the interstellar medium. These PAHs may subsequently be studied by spectroscopy. We have modeled this PDN, and verified that it produces a high flux of cold, metastable atoms of the noble carrier gas, which may then create the desired species by soft Penning ionization. Furthermore, very good agreement between the model and experimental results was found, and the discharge was characterized as a glow discharge. The proposal to increase the interelectrode distance in the device to enhance the yield of PAH in the desired state was investigated. It was found that this did not significantly increase the yield, and may even be counter-productive. It did, however, provide additional strong evidence that the discharge is a glow discharge.

The final application of the modeling framework treated in this work is the modeling of a cascaded arc light source. This light source is an intense source of short-ranged radiation, with peak emission in the blue and near ultra-violet part of the spectrum. The novel feature of this arc is a geometric pinching of the arc channel, that leads to a strong local enhancement of the dissipation and the light output. A model of this plasma was made to investigate the operation of the arc, in terms of light output and plasma properties. The model was found to match the experimental trend extremely well. As essentially the same model is used for the pulsed capillary discharge waveguide and the spark gap, this finding supports the results obtained for these applications as well.
Samenvatting

In dit werk wordt een raamwerk voor de multfluïde-modellering van plasma’s gebruikt om een aantal transiënte plasma’s te modelleren die gebruikt worden in het opwekken en geleiden van licht. Door het gebruik van een enkel raamwerk voor al deze applicaties kan optimaal gebruik worden gemaakt van de overeenkomsten die tussen deze plasma’s bestaan, hetgeen de betrouwbaarheid en nauwkeurigheid van het model sterk vergroot, terwijl het de ontwikkeltijd verlaagt. Met deze methode zijn een golfgeleider voor terawatt-lasers, een ultrasnelle vonkbrug, een spuitstuk met daarin een gepulste ontlading en een cascadeboog die als lichtbron gebruikt wordt gemodelleerd.

Wat de plasma’s in dit werk unificeert is dat het alle vloeistoffen zijn. Dit staat ons toe een benadering te gebruiken op basis van de geïntegreerde momenten van de algemene deeltjestransportvergelijking, de Boltzmann Transport Vergelijking. Binnen dit kader zijn er echter sterke verschillen in plasmadichtheid, tijdsduur, gassamenstelling en vermogensdichtheid. Dit vereist een model dat flexibel en uitbreidbaar is en waarin grote afwijkingen van evenwicht beschreven kunnen worden.

Golfgeleiders worden gebruikt om de interactielengte tussen een terawatt-laserstraal en materie te vergroten. Deze interacties leiden tot spectaculaire verschijnselen, zoals deeltjesversnelling met een hoge accleratiegradiënt en de emissie van fotonen in het extreem ultra-violet of zachte röntgenstralen. Wij hebben het modelleringstraamwerk gebruikt om een type golfgeleider, namelijk de golfgeleider in een gepulste capillaire ontlading te modelleren. In dit capillair geleidt een dicht waterstofplasma de laser. De eerste modellering is gedaan aan een capillaire ontlading die gebruikt wordt in experimenten, met als doel inzicht in de fysische processen die waargenomen worden te verklenen en om de overeenkomst tussen theorie en de kwaliteit van het model te toetsen door vergelijking met experimentele waarnemingen. De volgende stap was het doen van een parameterstudie aan deze ontlading, waarbij een empirische formule die de lasergeleiding relateert aan de controleparameters is opgesteld. Verder is er onderzoek gedaan naar het idee om een capillaire met een gemoduleerde straal te gebruiken, om de laserintensiteit lokaal te verhogen. Uit de modellering bleek dat deze techniek niet effectief is, en dat een capillaire met een gemoduleerde straal zeer gevoelig is voor ablatie van de wand. Tenslotte is een nieuw capillaire met een vierkante in plaats van een
ronde doorsnede gemodelleerd. Dit capillair wordt gebruikt voor nauwkeurige metingen van de dichtheid. De resultaten van de metingen komen uitstekend overeen met die van simulaties. Deze simulaties zijn van groot belang voor de interpretatie van de metingen.

Een puls van een terawatt-laser kan ook gebruikt worden om een vonkbrug te sluiten, wat leidt tot ultrasnelle schakeling. Experimenteel blijkt echter dat niet alle spanning die over de vonkbrug staat wordt geschakeld, in het bijzonder niet voor lagere spanningen. We hebben de vonkbrug analytisch en numeriek gemodelleerd om een verklaring voor deze spanningsval te vinden. Het bleek dat de spanningsval veroorzaakt wordt door de eindige geleidbaarheid van het boogplasma dat de elektrodes verbindt. Verder is inzicht verkregen in het gedrag van dit sterk veranderlijke plasma.

Diffuse interstellaire banden zijn absorptielijnen in het interstellaire spectrum die breder zijn dan normale absorptielijnen. Ze zijn bijna een eeuw onverklaard gebleven. Recentelijk is er op NASA Ames een opstelling gebouwd waarin een mogelijke oorzaak van de diffuse interstellaire banden, namelijk polycylyclische aromatische koolwaterstoffen (PAHs) en hun kationen. Deze opstelling, die in feite een spuitstuk met daarin een gepulste ontlading is, wordt PDN genoemd, en is in staat geëxciteerd PAH en PAH-kationen te genereren in een omgeving die lijkt op het interstellaire medium. Zij kunnen bestudeerd worden met spectroscopische techniqueen. Wij hebben deze PDN bestudeerd, en vastgesteld dat het een bron is van koude, metastabiele atomen van het draaggas, die de gewenste deeltjes kunnen produceren door zachte Penning-ionisatie. Verder is een uitstekende overeenkomst tussen de resultaten van metingen aan de PDN en resultaten van deze simulaties gevonden. De ontlading is geclassificeerd als een gloeiontlading. Het idee om de afstand tussen de elektrodes in het apparaat te vergroten om zo de opbrengst van de PAHs in de gewenste toestand te vergroten is onderzocht. Dit heeft opgeleverd dat er geen significante toename van de productie van metastabiele optreed door het verlengen van de afstand tussen de elektrodes, en dat dit zelfs contra-productief kan zijn. Wel er sterk bewijs gevonden dat de ontlading inderdaad een gloeiontlading is.

De laatste applicatie van het raamwerk die in dit werk beschreven wordt is de modellering van een cascadeboog die als lichtbron gebruikt wordt. Dit is een intense bron van straling, waarvan de emissie het hoogst is in het blauwe en ultraviolette deel van het spectrum. Wat nieuw is aan deze boog is een vernauwing van de boog, wat leidt tot een sterke locale verhoging van de dissipatie en van de lichtopbrengst. Een model van dit plasma is gemaakt om de werking van de boog, en dan met name de plasma-eigenschappen en de lichtopbrengst, te onderzoeken. Het model komt uitstekend overeen met de trends die experimenteel gevonden worden. Omdat het model dat voor de vonkbrug en de gepulste capillaire ontlading in essentie hetzelfde is, is dit verder bewijs voor de validiteit van de resultaten die voor deze toepassingen verkregen zijn.
Related Publications

Peer-reviewed articles

- B. H. P. Broks, J. G. Keizer, R. A. B. Zijlmans, and J. J. A. M. van der Mullen, Modeling of a pinched cascaded arc light source using argon, *to be submitted to Plasma Sources Science and Technology*


- B. H. P. Broks, J. van Dijk, H. M. J. Bastiaens, K. J. Boller, and J. J. A. M. van der Mullen, Study of a pulsed capillary discharge waveguide with a modulated radius, *Accepted for publication in Journal of Physics D: Applied Physics*

- B. H. P. Broks, W. van Dijk, and J. J. A. M. van der Mullen, Parameter study of the plasma and optical guiding properties of a pulsed capillary discharge waveguide, *Accepted for publication in Journal of Physics D: Applied Physics*


**Invited lectures**

• B. H. P. Broks, J. van Dijk, and J. J. A. M. van der Mullen, A method of enhancing the output of a high-harmonics extreme ultra violet source, COST 529 Meeting, Mierlo, The Netherlands, March 30- April 2 2006, oral contribution

• B.H.P. Broks, E. Stoffels, R. E. J. Sladek, J. van Dijk, W. J. M. Brok, and J. J. A. M. van der Mullen, Modeling of two biomedical plasma sources, to be given at the HTPP-9 conference, May 29 - June 4, 2006, St. Petersburg, Russia, oral contribution

**Peer-reviewed conference proceedings**

• B. H. P. Broks, and J. J. A. M. van der Mullen, Creating a global plasma model using Distured Bilateral Relations, *Accepted for publication in Journal of Physics:Conference Proceedings*


• J. J. A. M. van der Mullen, and B. H. P. Broks, Distured bilateral relations: A guide for plasma characterization and global plasma models, *Accepted for publication in Journal of Physics:Conference Proceedings*

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• W. J. M. Brok, B. H. P. Broks, J. Remy, and J. J. A. M. van der Mullen, Model Study of the Pulsed Discharge Nozzle, 58th Gaseous Electronics Conference, October 16?20, 2005, San Jose, California


Conference proceedings

• B. H. P. Broks, W. van Dijk and J. J. A. M. van der Mullen, Parameter study of the slow pulsed capillary discharge, 8th Euregional WELTPP, Rolduc, The Netherlands, November 24-25 2005, oral contribution


• B. H. P. Broks, and J. J. A. M. van der Mullen, Nonlocal-thermal-equilibrium model of a pulsed capillary discharge waveguide, 7th Euregional WELTPP, Rolduc, The Netherlands, 25-26 November 2004, oral contribution


• B. H. P. Broks, and J. J. A. M. van der Mullen, Modeling a capillary discharge in PLASIMO, Spring Meeting of the DPG/IOP/DPSP/BPS, Germany, 24-28 March 2003, poster contribution
Dankwoord

De mythe dat wetenschap iets is dat je alleen doet is zeker niet van toepassing op dit werk, dat het resultaat is van de vruchtbare samenwerking met een groot aantal mensen. Zonder daarbij de velen die achter mij hebben gestaan en dit werk mogelijk hebben gemaakt iets te kort te willen doen, wil ik er graag een aantal personen in het bijzonder uitlechten.

Dit werk draagt het stempel van mijn promotor Joost van der Mullen, onder wiens bezielende en inspirerende begeleiding ik heb gepoogd me het vak van wetenschapper eigen te maken. De vele verhelderende discussies, goede ideeën, en de vraagstukken die hij aandroeg hebben dit proefschrift in een belangrijke mate geschapen. Mijn oprechte dank daarvoor. Verder wil ik ook de leden van mijn promotie-commissie van harte bedanken voor de uitgebreide en nuttige discussies.

Het overgrote deel van dit proefschrift bestaat uit het modelleren van plasma’s met een numerieke code. Deze code is voor het overgrote deel niet door mij geschreven, maar het resultaat van het werk van velen, in het bijzonder Bart Hartgers, Jan van Dijk, Gerjan Hagelaar en Wouter Brok. Zonder hun kennis, vaardigheid en behulpzaamheid zou dit werk nooit in deze vorm tot stand kunnen zijn gekomen. Ook de andere leden van het team, Michiel van den Donker, mijn voormalige afstudeerder Mark Beks, Colin Johnston en Kurt Garloff ben ik dank verschuldigd voor hun bijdragen tot de code, maar ook voor de gezellige sfeer die ze hebben weten te creëren.

Bijna alle publicaties in dit proefschrift zijn tot stand gekomen in wetenschappelijke samenwerkingen met groepen binnen en buiten de universiteit. Voor hun bijdrage aan het werk aan de capillaire ontladingen wil ik Walter van Dijk en Reda Akdim in het bijzonder bedanken. Furthemore, I would like to extend my gratitude to the group of Simon Hooker, and in particular Tony Gonsalves, at Oxford University for their hospitality and cooperation. Het werk aan de sparkgap is gedaan in nauwe samenwerking met Wouter Brok, Jimi Hendriks en Seth Brussaard, en ik wil ze bedanken voor de goede discussies en hulp bij het schrijven van het artikel. Het werk aan de PDN is gedaan in samenwerking met Wouter Brok, Jérôme Remy, Ludovic Biennier, Abdessamad Benidar en Farid Salama. Ik ben hun dank verschuldigd voor de goede discussies en het geven van ondersteuning in de vorm van aanpassingen aan de code en noodzakelijke invo-
ergegevens. Tot slot wil ik Rens Zijlmans en mijn stagair Joris Keizer bedanken voor hun bijdrage aan het werk aan de cascadebogen.

De afgelopen 5 jaar heb ik het genoegen gehad lid te zijn van de groep EPG. De leden van deze groep hebben gezorgd voor een prettige werksfeer, en daarvoor wil ik ze graag bedanken. In het speciaal wil ik Rina Boom in het zonnetje zetten, die het papierwerk stipt en correct afgehandeld heeft en daarmee een belangrijke bijdrage heeft geleverd aan het doorlopen van het promotie-traject.

Tenslotte wil ik Dirk bedanken voor zijn hulp bij het typesetten van het proefschrift en voor het maken van de kaft.
Curriculum Vitæ

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