Sputtering Hollow Cathode Discharges designed for Laser Applications; Experiments and Theory

PROEFSCHRIFT

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to my family
Summary

This thesis presents experimental and modelling studies of hollow cathode discharges (HCDs). This type of plasma source has several applications, for instance in the field of atomic spectrometry, vacuum microelectronics, UV generation, ion sources, plasma processing, surface treatments and laser technology. More specifically, this study is devoted to the application of HCDs in laser technology. In this application field the HCD acts both as a metal vapour source, by means of cathode sputtering, and as an active medium to excite metallic laser transitions. To improve and optimise the laser performance a proper understanding is needed of the various processes and mechanisms that take place in the hollow cathode plasma and how these depend on the settings of the external parameters.

Experiments: Several laser tubes were designed, built and studied experimentally. The impact of the external parameters settings on the laser operation were investigated by monitoring the behaviour of the IR copper ion line (780.8 nm), the transition with the highest gain. In this way the features of the optimal conditions and the best geometry could be determined experimentally.

Modelling: The PLASIMO-MD2D platform was used to construct a model that facilitates in-depth studies of the plasma behaviour and that can be used to find the optimum location in parameter space for the operation of these discharge types. The versatility of the model makes it possible to simulate the plasma processes in different HCD geometries, gas compositions and operation parameters.

Analytical model: An analytical model was constructed with the aim to interpret the numerical results. The model is focused on the main plasma properties such that plasma density, potential, current, etc. It is based on some elementary theories of glow discharges and extends them to account the axial non-uniformity of longitudinal HCDs.

The modelling results were validated with experimental observations. This comparison is guided by the analytical model that was constructed for that purpose. The insights obtained by this validation have lead to an improvement of the numerical model that can now successfully be employed as an optimisation tool of HCD lasers in particular, and more in general, for the improvement of various other HCD applications.
Samenvatting

Dit proefschrift bespreekt experimenteel en modelmatig onderzoek aan holle-kathodeontladingen (Engels: hollow cathode discharges, HCDs). Dit type plasmabron heeft diverse toepassingen, bijvoorbeeld in atomaire spectroscopie en vacuüm micro-elektronica, voor de productie van ultraviolette straling en ionenbronnen, en binnen de plasma processing, oppervlaktebehandeling en lasertechnologie. Deze studie richt zich in het bijzonder op de toepassing van HCD’s in lasertechnologie. In deze toepassing draagt de HCD zowel zorg voor de introductie van metaaldamp in het plasma middels sputtering van kathodemateriaal, als voor de aanslag van metaaldeeltjes naar toestanden die voor laserwerking kunnen zorgen. Om de laser-performance te verbeteren is een grondig inzicht vereist in de processen en mechanismen die plaatsvinden in de HCD en hoe deze afhaken van de keuze voor de systeemparameters.

**Experimenten:** Diverse laserbuizen zijn ontworpen, gebouwd en experimenteel onderzocht. De invloed van de systeemparameters op de lasereigenschappen zijn bestudeerd door het gedrag van de infra-rood koper-ion overgang (780.8 nm), die de grootste laser-versterkingsfactor bezit, in kaart te brengen. Aldus zijn de optimale geometrie en andere systeemparameters bepaald.

**Numerieke Modellering:** Het PLASIMO-MD2D modelleerplatform is gebruikt voor de constructie van een model dat een gedetailleerde numerieke plasmastudie toestaat. Het kan worden gebruikt ter bepaling van de optimale keuze voor de instelparameters voor de gasontladingslasers. De flexibiliteit van het model heeft het mogelijk gemaakt om verscheidene HCD-geometriën, gassamenstellingen en andere systeemparameters aan onderzoek te onderwerpen.

**Analytische Modellering:** Teneinde de numerieke resultaten te interpreteren is een benaderend analytisch ontladiingsmodel opgesteld. Dit richt zich op de primaire plasmaparameters zoals de elektronendichtheid en de elektrische potentiaal en stroom. Het is gebaseerd op een uitbreiding van gangbare elementaire gasontladingstheorieën die rekening houdt met de axiale variaties in longitudinale HCD’s.

De modelresultaten zijn gevalideerd door deze te vergelijken met de experimentele waarnemingen. Hierbij heeft het analytische model een ondersteunende rol gespeeld. De hieruit verkregen inzichten hebben weer geleid tot verbeteringen van het model. Dientengevolge kan dit nu niet alleen worden ingezet voor de optimalisatie van HCD-lasertoepassingen, maar ook voor de studie naar verscheidene andere toepassingen van holle-kathodeplasmas.
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1

General introduction

“Laser... inter eximia naturae dona numeratum plurimis compositionibus inseritur.”

Laser is one of the most miraculous gifts of nature and lends itself to a variety of applications.

*Plinius, Naturalis Historia, XXII, 49 (1st century A.D.)*
Chapter 1.

1.1 Introduction

“Far out in the uncharted backwaters of the unfashionable end of the Western Spiral Arm of the Galaxy lies a small unregarded yellow sun. Orbiting this at a distance of roughly ninety-two million miles is an utterly insignificant little blue-green planet...”\(^1\)

To the knowledge of (some of) the inhabitants of this little blue-green planet, the yellow sun is a so-called plasma. It is often stated that more than 95% (or 99%, depending on whom they are trying to impress) of the visible matter of the Universe is in the plasma state, the rest being mysterious dark matter or dark energy. We are not going to hitchhike in the Universe to verify this statement (this seems to be difficult), but rather we will descend to the little blue-green “mostly harmless” planet Earth to find other examples of this state of matter.

A plasma is a gas in which a considerable fraction of the particles is electrically charged. These electrons and ions make the gas electrically conductive so that it can and will respond to electromagnetic fields. Therefore, plasmas have properties that are substantially different from those of solids, liquids, or gases. This makes the plasma state to be considered as a distinct state of matter.

In contrast to the large scales plasmas like stars, nebulae and galaxies that are mainly driven by nuclear, gravitational and radiation power, the small-scaled man-made plasmas are mainly sustained by electrical energy.

There is a large variety of technological plasmas, employed in many different applications: plasmas are widely used for surface treatment, for cutting and welding and they are employed for etching and deposition of materials; processes that are for instance used for the fabrication of computer chips. Apart from that, plasmas are used for lighting, as e.g. generated by discharge lamps and plasma display panels.

Plasmas also play an important role in laser technology. Unlike any other light source, laser light is highly concentrated in space, time (coherence) and spectral range (monochromaticity). The light of a laser differs from that of an ordinary light source more or less as sound differs from noise (or Chopin from Eminem). Lasers are the brightest and strongest light sources ever created by mankind.

The invention of the laser initially denoted as a “solution of a problem that do not exist”, eventually turned out to be able to solve many problems almost in any field of science, industry and daily life. The huge number of available lasers nowadays have resulted in many applications in very different fields and many other utilizations are to be expected in near future.

This chapter gives a general introduction to gas discharges, briefly describes the specific type of plasmas that is used for laser applications, and discusses the type of lasers studied in this thesis. The description is intended to be as detailed as needed to make this thesis self-contained. At the end of this chapter the scope and outline of the remainder of the thesis will be presented.

\(^{1}\) “Hitchhiker’s guide to the galaxy”, by Douglas Adams.
1.2 Gas discharges

Gas discharge lasers form a class of gas lasers that are excited by a sufficiently large current, direct (DC) or pulsed, passing through the gas and creating a plasma. The plasma can be created in a pure gas, inert or reactive, atomic or molecular, in mixtures of gases, or in mixtures of gases and metal vapours. Due to the large variety of chemical compositions, the gas discharge lasers can offer laser oscillations spread out over the whole optical range — from the VUV to the far IR spectral range in a continuous or pulsed mode.

The description of the specific types of plasmas used in this thesis is facilitated by making a comparison with the classical glow discharge. To that end a glow discharge is described first.

Figure 1.1 gives an outline of the different regions of a canonical DC gas discharge [1]. As an example we take a tube with a radius of about 3 cm and a length of 30 cm filled with helium gas at low pressure in the order of $10^{-2}$ mbar. Between the electrodes, which are placed at opposite ends, a voltage of approximately 400 V is applied such that a current flows through the gas without the assistance of any external ionising agent: it is a self-sustained discharge. A number of regions can be distinguished in figure 1.1. Globally starting at the cathode we encounter the cathode fall (CF) region, the negative glow (NG), the positive column (PC) and anode layer. Most of the applied voltage is concentrated

\begin{figure}[h]
\centering
\includegraphics[width=\linewidth]{figure1.png}
\caption{Top: different regions of a canonical gas discharge; bottom: the voltage distribution as a function of the axial position; notations: the Aston dark space (ADS), cathode glow (CG) and cathode dark space (CDS) form the cathode fall (CF) region. Beyond the CF are successively the negative glow (NG), Faraday dark space (FDS) and positive column (PC). Close to the anode are the anode dark space (ADS) and the anode glow (AG).}
\end{figure}
Chapter 1.

in the CF region where the strong electric field accelerates ions toward the cathode and electrons into the discharge. In general this leads to the formation of successive dark and glow regions in the CF: the Aston dark space, the cathode glow and the cathode dark space.

We will now follow the electrons in their journey from the cathode to the anode. In the Aston dark space, the electrons that have just been emitted from the cathode do not have enough energy to excite atoms. As their energies increase with the distance from the cathode, at some point the electrons are able to excite buffer gas atoms and a luminous region is formed, which is called the cathode glow. Beyond the cathode glow region, the electron energy is increased to such an extend that the excitation cross-section diminishes. Consequently, a region of low luminosity is formed called the cathode dark space.

Next to the CF we find the negative glow (NG), where the electrons spend the energy gained in the CF to excite and ionise atoms. This makes the NG the most luminous region of the discharge. Most of the ions produced in the NG are lost from the discharge due to diffusion in radial direction. Others are accelerated towards the cathode where they, upon impact, cause the ejection of electrons. These secondary electrons are needed to continue the process. Hence the CF and NG cooperate together to sustain the discharge: the electrons launched by the CF create ions in the NG that via the CF are accelerated towards the cathode where new electrons are liberated. So in contrast to the positive column (see below), the synergetic combination of CF & NG is needed in DC discharges. However, there are large differences between the CF and NG. The NG is a region characterised by charge neutrality; essentially it is a field-free region. The CF on the other hand, is dominated by a positive space charge density, which gives the potential the typical shape shown in figure 1.1.

As the electrons dissipate their energy in the NG, events of excitation and ionization become less and less frequent, because electrons do not gain new energy in the weak field. This leads to a region beyond the NG with a low luminosity called Faraday dark space (FDS). In this region the longitudinal field gradually increases to a value that is needed to support the energy losses in the positive column (PC). In the PC electrons have gained sufficient energy to excite gas atoms. The electrons in PC have a near Maxwellian energy distribution. The PC can be arbitrarily long depending among others on the voltage as offered by the power supply.

Not all the regions given above are present in every DC discharge. The exclusion of plasma regions or the modification of others can be realised by varying the conditions such as pressure, electrode distance and electrode material, the applied voltage and gas mixture.

Usually, an increase of pressure causes the CF, NG and FDS to be compressed towards the cathode. A decrease of pressure naturally causes the reverse effect: these zones expand and their boundaries become more diffuse. The positive column is driven into the anode and disappears altogether when the pressure is sufficiently low. The rise of the potential has more or less the same effect as increasing the pressure. In reality there is also an increase in the current through the discharge and consequently a general increase in the brightness of the luminous parts. The voltage that is necessary to maintain the discharge depends also on the cathode material: lower voltages are needed when the cathode is a good emitter of electrons under bombardment by positive ions or photons.
General introduction

Shortening the distance between the anode and the cathode of the discharge tube eventually leads to the disappearance of the PC, leaving only the cathode dark space, negative glow and Faraday dark space. The anode can be positioned adjacent to the negative glow so that the entire PC and FDS are absent without unduly affecting the discharge, since the NG and CF are the region which maintain the discharge. Current is carried by an ion flow from the NG to the cathode and an electron flow from the NG to the anode. Thus what mostly remains is the bright region of the negative glow that gave the name *glow discharge* to this type of discharge as a whole.

If the anode is moved even closer to the cathode, the length of the NG is shortened but sufficient ionization may still occur to maintain the discharge. If the anode is moved even further to the cathode, the NG will be shortened too much and the ionization may be insufficient to maintain the discharge. If this happens, we speak of an *obstructed discharge*. Gas discharge devices often utilise the obstructed discharge in certain regions to ensure that parasitic discharges do not develop in those regions.

As seen, the discharge can in principle be sustained without a PC or FDS. In a DC discharge the most important part is the CF region, where most of the potential drop between the electrodes occurs. However, the cathode fall is not necessary if there is an other mechanism at the cathode that creates sufficient electrons. An example is the hot cathode discharge, where electrons are released by thermionic emission so that the action of ions or photons is not needed.

The various plasma applications make profit of the different plasma regions or combinations thereof. For example the fluorescent lamps makes use of the PC discharge. The same applies to microwave plasmas or other high-frequency high-current plasmas. For these plasmas there is no need to liberate the electrons from the cathode, the CF is not needed and these plasmas can be operated electrode-less.

Gas discharge lasers can also be based on the employment of different plasma regimes: laser action can for instance be performed in the positive column, or in the negative glow. A typical representative of a PC laser is the well-known He-Ne laser; this is the first laser in history based on the excitation of atoms by means of electrons generated in the PC of a gas discharge. The 633 nm Ne laser line is probably the most popular and most widely used laser line of He-Ne lasers. The PC is also used by the Ar *ion laser* which can oscillate at a number of wavelengths; the most intense are the blue line at 488 nm and the green line at 514.5 nm. Unlike the He-Ne laser, the laser action involves transitions between ion levels. Lasing of ion transitions has been obtained on hundreds of lines of different elements in various gas mixtures.

When the active medium for laser excitation is a mixture of rare gases (for instance He, Ne, Ar, Xe) and metal vapours, we speak of *metal vapour lasers* (MVLs). The MVLs oscillating on ion transitions at the metal levels will be subject in this thesis.
1.3 Hollow cathode discharges

Laser transitions in metallic ion-systems were for the first time obtained in the positive column (PC) of a gas discharge. But it immediately became clear that better laser characteristics could be obtained by employing the negative glow (NG) instead of the PC. However, as stated in the previous section, in a discharge with the configuration shown in figure 1.1 a substantial fraction of the charged particles created in the NG are lost by radial diffusion so that only a minor fraction of the ions travels towards the cathode where they contribute to secondary electron emission — the mechanism that is needed to sustain a DC discharge.

If the planar cathode is replaced by a cylinder with a length and inner diameter comparable to the width of the CF region, the NG will be confined within the cathode. Such a cathode is called a hollow cathode (HC). Discharges equipped with this configuration primarily consist of two distinct regions: the regions of the NG and the CF and, as we have seen, these regions feed each other to sustain the discharge.

It is clear that in this hollow cathode configuration many of the ions produced in the NG that in ordinary discharges would have been lost by wall recombination will be accelerated to the cathode. There they will greatly contribute to the current by secondary emission processes. This implies that for a given discharge voltage, the current density in a HCD can be orders of magnitude larger than in a conventional discharge. This is known as the hollow cathode effect (HC effect).

Most properties of the HC effect result from the more efficient use of electrons and ions generated in that closed configuration. The minimised losses of charged particles leads to a relatively low discharge voltage, which depends on the diameter $d$ of the hollow cathode and the gas pressure $p$. Figure 1.2 shows the dependence of the voltage on the parameter $pd$.

We will follow the changes that take place when the pressure is reduced for a hollow cathode with a given diameter $d$ and constant current. At high pressure $p$, the sizes of the CF and NG are small compared to $d$; the NG is a bright ring near the cathode surface whereas the centre of the cathode, being ruled by the FDC, is darker. With decreasing

![Figure 1.2: The voltage-pressure characteristic of a hollow cathode discharge at constant current.](image)
pressure, the thickness of the CF and NG increases, as we have seen, so that a larger and larger part of the bore will be filled with NG plasma. As a consequence the discharge becomes more efficient and the voltage decreases to a minimum. At a certain pressure-value the voltage is at minimum and the discharge is brightest in the centre. This is the situation for which the hollow cathode effect is strongest. At still lower pressures the tube voltage begins to rise, as the size of the CF becomes comparable to the NG, i.e. the discharge becomes obstructed. It is clear that the optimum pressure, for which the voltage is lowest, depends on the diameter $d$ of the cathode so that the transitions described above depend on the parameter $pd$. The optimum in $pd$ depends on the gas-type. For gases with low ionization potential this optimum $pd$-value is lower.

There is a wide variety of HC geometries that lead to a myriad of applications, for instance in the field of atomic spectrometry, vacuum microelectronics, UV generation, ion sources, plasma processing and surface treatments. In this thesis the plasma created in a HCD will be used as active medium of so-called hollow cathode lasers.

### 1.4 Hollow cathode laser constructions

To obtain high laser powers one needs to increase the discharge volume, the current density as well as the pressure. To optimise the discharge configuration, a wide variety of laser hollow cathode designs have been built and studied. Some examples of HC laser geometries are shown in figure 1.3.

In order to provide a useful gain-length in the NG, the optical axis of the laser is always chosen to be parallel to the cathode surface, while the cathode is made as long as possible. The anode can be placed parallel to the cathode such that the plasma current is transverse to the optical axis as shown in figure 1.3(a). In such a transverse HCD geometry the discharge length is unlimited, in principle. However, the corresponding high discharge current densities lead to arcing, which not only stops the laser operation but may also damage the discharge tube. Another electrode-arrangement is the so-called longitudinal configuration, where the optical axis is parallel to the current (figure 1.3(b)). In this case the chance of arc-formation is minimised but strong axial inhomogeneities may arise that limit the increase of the active volume. Different variations of these two types exist — with outside or inside anodes; or with coaxial electrodes. A variant of the longitudinal HC is the so-called flute type (figure 1.3(c)). As mentioned before, the laser performance also depends on the efficiency of the excitation of the ion levels, which can be improved by increasing the “beam” of fast electrons produced in the cathode fall. This can be realised by increasing the voltage which is possible in the so-called high-voltage (HV) constructions. The first HV construction was developed by Rozsa et al [2] (figure 1.3(d)). Other variants of HV constructions are the helical HCs, shown in figures 1.3(g) and 1.3(h) [3].

Laser oscillations in HC lasers have been obtained on almost 300 transitions in more than 20 metals. One of the most developed and studied laser is the He-Cd ion laser. The major visible lines of Cd$^+$, the red (636 nm), green (537 nm) and blue (442 nm) are sufficiently close to the primary colours to give the possibility for “white-light” lasers.
Figure 1.3: Various hollow cathode laser constructions.
The applications include colour holography, colour separation and printing, film-to-video conversion and colour microscopy.

Apart from oscillations in the visible spectral range, the hollow cathode MVLs are attractive due to the possibility to create new reliable sources for lasing in UV with a high degree of monochromaticity. At present only two types of laser systems are known that provide direct laser oscillation in the UV: the excimer and the metal vapour laser. Excimer lasers are the most powerful sources, but operate only in a pulsed mode with comparatively low repetition rates. Moreover, they suffer from low coherence. Metal vapour lasers are the only direct sources of (quasi-) continuous laser oscillation. Although they have comparatively low powers (up to 1 W), the quality of the laser beam is very high, with high degrees of coherence in the UV spectral range below 300 nm. This makes the HC lasers suitable for many applications in biotechnology, micro-lithography, UV Raman spectroscopy, for the pumping dye lasers and to save information on photo-materials with high resolution and speed [4]. In medicine, MVLs are used for eye surgery.

The first demonstration of direct UV laser action was by McNeil et al [5], who used a slotted HC (figure 1.3(a)) in Ne buffer gas. Continuous wave laser actions was obtained from Cu ion transitions at 259.1 and 259.9 nm, quasi-continuous action was obtained at 248.6 and 250.6 nm. Laser oscillation in HC in the 200–300 nm spectral range is obtained on Cu$^+$ (11 lines), Ag$^+$ (2 lines) and Au$^+$ (7 lines). The main representatives of the UV HC lasers are the Ne-Cu$^+$, He-Ag$^+$ and He-Au$^+$ lasers. The most energetic transitions are obtained in the Ag$^+$ system (224.3 and 227.7 nm) [5].

1.5 Laser excitation mechanisms

Depending on the combination of buffer gas and metal vapour, one can distinguish different excitation processes that can generate population inversion. The main excitation mechanisms, also shown schematically in figure 1.4, are:

- **Asymmetric charge transfer.** In a metal-ion laser asymmetric charge-transfer usually takes place between a buffer gas ion B$^+$ and a metal atom M and proceeds according:
  \[ B^+ + M \rightarrow B + M^{++} + \Delta E, \]  
  \[(1.1)\]
  where $\Delta E$ is the difference between the ionization potentials. This process is quasi-resonant and takes place with high probability at $\Delta E = 0.2 − 0.5 \, \text{eV}$.  

- **Radiative cascade.** In many cases it is possible to obtain selective population of energy level with strong radiation transitions (in many cases also laser transitions) from higher levels, which are populated via charge transfer.

- **Penning ionization.** In this process, metal atoms are ionised in collisions with metastable buffer gas atoms:
  \[ B^* + M \rightarrow B + M^{++} + e(\Delta E). \]  
  \[(1.2)\]
  This process is effective when the energetic difference ($\Delta E$) is up to $2 − 3 \, \text{eV}$. 

In some cases other processes like direct or stepwise electron excitation can contribute to population inversion, but usually their contribution is negligible. In HCDs the most effective process is the charge transfer reaction of the type given by equation (1.1).

1.6 Metal vapour production

One of the main issues for MVLs is to bring metal vapours into the discharge in a way that leads to uniform distributions.

Traditionally, metals vapours are introduced by thermal evaporation, which requires high temperatures, in some cases higher than 1000°C; hence great technical difficulties in the design of MVLs must be overcome.

Another method to produce the needed metal vapours of hard evaporating metals is to bring them into the discharge as carried by their easily evaporating volatile compounds (i.e. CuBr, CuCl), which are dissociated in the plasma while producing free atoms.

Last but not least, we mention the method which has a key position in this thesis
and that is known as cathode sputtering. The cathode is made of a refractory metal, usually Cu, Ag, Au, Al, which by means of sputtering generates the necessary metal atom concentration in the discharge.

Sputtering is a stochastic process in which every time when an energetic particle impinges the cathode, ejection of atom(s) occurs with a certain probability. Generally it is caused by collisions between the incoming particles and the atoms in the (near) surface layer(s) of the solid. It is a typical multiple collision process involving a cascade of moving target atoms. The net result is usually expressed in terms of the sputtering yield, which is defined as the number of sputtered atoms per particle impinging the surface.

The sputtering yield depends on the properties of the incoming particle (mass, energy and incidence angle), the properties of the solid (structure and orientation of the solid), the surface binding energy, the number atoms set in motion, the type of collisions, the beam-target geometry, etc. The calculation of the sputtering yield is not simple due to the complexity of the problem so that a number of assumptions have to be made. There are considerable differences in the basic processes that the various authors take as responsible for sputtering [6, 7, 8, 9]. Figure 1.5(a) reproduces the sputtering yield calculated using Yamamura’s formula [8] and the model of Mahan et al [9]. Figure 1.5(b) shows the sputtering yield of various gas and metal ions. Since the sputtered metal atoms can also be ionized in the glow by means of electron collisions or charge exchange, the formed metal ions can produce sputtering as well — this is called self-sputtering. The metal ions play a

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2 The word “sputter” appeared in the English language as early as 1598 and is adapted from the imitative words “sputteren” in Dutch and “sputterje” in West Frisian.

The dictionary (wiktionary) says: “to spit or to emit saliva from the mouth in small, scattered portions, as in rapid speaking; to utter words hastily and indistinctly, to speak so rapidly as to emit saliva; to throw out anything, as little jets of steam, with a noise like that made by one sputtering”.

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Figure 1.5: Sputtering yield of a copper cathode; 1.5(a) sputtering yield for Ar ions on Cu target, as a function of ion energy according to the Yamamura [8] and Mahan [9]; 1.5(b) sputtering yields due to bombardment by He, Ar and Cu particles on Cu target as a function of the projectile energy, calculated with the empirical formula of [8].
quite important role in sputtering, especially at high enough current densities [10]. This is mainly attributed to the fact that the target and projectiles have the same mass meaning that the energy transfer when bombarding the cathode is most efficient.

In many applications, sputtering is an undesired effect that contaminates plasmas and destroys electrodes and grids. Nevertheless, sputter plasmas become more and more of interest. Sputtering is used for many applications and has become an indispensable process in modern technology. It is widely applied for surface cleaning and etching, thin film deposition, depth profiling of thin films, surface and surface layer analysis, for the creation of ion beams, and for the production of metal vapours.

In MVLs sputtering is the easiest way to bring metal vapours into the discharge. The minimised diffusive losses of charged particles in a HCD enable efficient cathode sputtering. The main advantage is that lower temperatures can be used, which simplify the experimental design. However, the process of sputtering destroys the cathode, reducing the lifetime of the laser tube. Another disadvantage is that the discharge current and the metal vapour pressure can not be controlled independently.

The first metal ion laser oscillation on Cu$^+$ 780.8 nm was discovered by chance by Csillag et al (1974) [11] during experiments with a He-Kr$^+$ laser, which utilised a slotted Cu cathode. Since that time, sputtering has been used for metal seeding of metal vapour lasers and laser oscillation has been observed on more than 130 lines; 46 of them being from Cu$^+$. In this thesis we will employ the sputtering of Cu cathode for laser oscillation of the Cu$^+$ 780.8 nm laser line. This transition ($6s^3D_3 - 5p^3F_4$) is of interest because the upper level is also the upper level of a number of potential laser transitions in the UV.

1.7 Modelling of HCDs

The method of modelling is complementary to experimental work and theoretical studies. By bringing a set of equations describing the system to a solution, the model can give insights in the plasma. In contrast to the experimental techniques, the converged solution gives insight in several relevant quantities and their mutual dependences. It is possible to study how they influence one another by artificially manipulating them. The model results can directly be compared to experimental data and this can increase the confidence in the validity of the model. Finally, model results can initiate new experiments and help to interpret observations.

Analytical models have been used since centuries. They can be accurate and give useful results if they are based on valid approximations. As such they can generate direct insights into the fundamentals of complicated problems. However, when many physical processes conspire it can be difficult to provide an analytical solution. With the development of computer technology, numerical modelling has come up as a tool for more accurately describing systems that could not easily be treated analytically. A wide variety of numerical models has been developed nowadays and continue to be developed for a wide variety of purposes. Some models represent all major aspects of the plasma behaviour self-consistently, others focus on certain aspects by solving specific problems.
The challenge of plasma modelling is to represent all the physical aspects that are relevant for a given purpose and configuration. The complexity of the numerical description of plasmas is due to the fact that many different mechanisms may be at play simultaneously, like the electromagnetic coupling with an external source, the myriad of elementary reactions that take place between particles, diffusive, radiative and convective transport, multi-fluidity and so on.

At Eindhoven University of Technology the modelling toolbox PLASIMO has been developed. It can be used to construct simulation models for a wide variety of plasmas. Sub-models are available for various electromagnetic problems and for plasmas with different degrees of equilibrium. Moreover, PLASIMO offers modules for calculating various plasma aspects such as temperature and flow fields and radiation transport. More recently, the Monte-Carlo sub-model has been developed, which allows kinetic simulations. In the near future it will be able to couple this MC sub-model to the fluid part of PLASIMO, so that hybrid modelling can be performed. In this thesis we will report on the usage of the MD2D sub-model, which can handle drift-diffusive plasmas, to describe the HC discharge numerically.

1.8 This thesis

This thesis presents experimental, numerical and analytical studies of sputtering hollow cathode discharges (HCDs). Special attention is devoted to the HCD as active medium for metal vapour lasers. In this application the HCD plasma acts both as a metal vapour source, by means of cathode sputtering and as an active medium to excite metallic laser transitions. The hollow cathode laser construction under study belongs to the longitudinal type (figure 1.3(b)).

The final goal of this study is to optimise the operating conditions and the hollow cathode configuration in order to improve the laser performance and to build a realible and efficient sputtering metal vapour laser. To that end, a proper understanding is needed of the various processes and mechanisms that take place in the hollow cathode plasma and how these depend on the settings of the external control parameters.

Chapter 2 gives an overview of experimental explorations of sputtering hollow cathode Cu⁺ ion lasers. The performance of various discharges is studied experimentally by investigating the axial current distribution, spontaneous emission and laser performance and how these are influenced by the control parameters such as input current, gas composition and electrode geometry.

Chapter 3 describes the PLASIMO sub-model MD2D and outlines the extensions that have been realised for the simulation of sputtering HCDs in the framework of this project. Chapter 4 describes the construction of a lean and realible model for the chemistry in the discharges under study. Chapter 5 shows that the model can be used as a versatile tool for the optimisation of sputtering metal vapour discharges. Chapters 6 and 7 are devoted to the optimisation of the geometry of such discharges in the context of laser applications. In particular the influence of the length (6) and diameter (7) of the hollow cathode is
discussed.

An analytical model (chapter 8) is developed with the aim to interpret the numerical results and to gain insight in the causal relations between the various mechanisms. The analytical and numerical modelling results are compared with each other and with the experimental data and observations.

Chapter 9 is devoted to recent developments and discusses plans for future work. Finally, conclusions are presented in chapter 10.

References

Experimental exploration on sputtering hollow cathode Cu ion lasers

Abstract. This paper gives an overview of systematic experimental studies on the longitudinal sputtering hollow cathode discharge (HCD) with the aim to investigate in detail the process of laser oscillation in these devices. As the main problem of the longitudinal HCD lies in the axial inhomogeneity we have studied the current and plasma species distribution along the cathode length for different discharge conditions, gas compositions and cathode geometries. The optimal conditions for lasing and the optimal cathode dimensions have been determined experimentally. A reliable configuration of a sputtering metal vapour laser is found to be such that a nearly homogeneous axial plasma species distribution in the cathode volume is created. As a result the optical losses in the discharge are reduced and a more efficient laser excitation is achieved.
2.1 Introduction

Hollow cathode discharges (HCD) are widely used as active media for the excitation of laser transitions of non-volatile metals such as Cu, Au, Ag, etc. They are promising sources of (quasi-) continuous laser oscillation in the UV and deep UV (DUV) spectral range. At present only two types of laser systems are known that provide direct laser oscillation in the UV: the excimer and the metal vapour laser. Excimer lasers are the most powerful sources, but they operate only in pulsed mode at comparatively low pulse repetition rates and low coherence. Metal vapour lasers are the only direct sources of (quasi-) continuous laser oscillation within narrow line widths and high degrees of coherence in the UV spectral range below 300 nm. Moreover apart from oscillation in the UV they can generate visible and IR radiation with a high degree of monochromaticity.

Different methods for producing metal vapours in glow discharges are known: thermal evaporation of the metal or metal-compounds in external reservoirs or in the cathode cavity, or by sputtering of the cathode material. The latter method is applicable to metals such as Cu, Ag, Au etc., that have a very low vapour pressure. At the same time these metals have a high sputtering rate [1] so that by sputtering sufficient metal vapour can be brought into the discharge.

Cathode sputtering, as a method to produce free metal atoms in the HCD for laser oscillation, was demonstrated for the first time by Karabut et al [2]. The method became popular after the report of Csillag et al [3] who demonstrated oscillation of the Cu$^+$ 780.8 nm line. In a short period after this publication, oscillations on more than 130 lines of the ion spectra of 12 metals (Cu, Ag, Au, Al, Ni, Ir, Sn, In, Ga, Bi, Ge, and Te) were reported in the range from DUV to the near IR [4]. The interest in this method of producing metal vapours in the discharge was provoked by the prospect of achieving laser action in the DUV range with very good beam quality. Oscillations on 22 lines of the ion spectra of Cu, Ag and Au were obtained in the spectral range below 300 nm [5, 6, 7, 8, 9, 10], while laser powers of almost 1 W at multi-line operation of Cu ion lines in the DUV was achieved [11, 12]. Potential oscillations in the vacuum UV range (below 200 nm) on Cu [13, 14] and Al [15] ion transitions were discussed as well.

As the laser gain in the UV is low compared to that in the visible spectral range, different methods to increase the excitation efficiency and hence the laser power have been studied. The investigations are carried out in three main directions: 1) increasing the input power; 2) optimising the gas composition and 3) improving the design of the electrode geometry.

Improving the laser power by increasing the input power is an obvious choice since the laser lines are mainly excited by means of charge transfer between the buffer gas ions and metal atoms [5]. The density of both these interacting species will be enhanced by increasing the input power of the discharge. In general the HCDs are characterised by a large increase of current while the discharge voltage remains nearly constant if all the other operation parameters are kept constant. Thus the increase in power can mainly be achieved by increasing the current. However the possibility is limited by a critical current value that depends on the discharge conditions and electrode configuration. Above this value a saturation of laser power is observed [8, 16, 17]. In many cases the saturation
is attributed to an excess of metal atoms at high current values [12], but there is no experimental evidence for this assumption. Previous attempts [18] to describe laser power saturation by modelling were also not successful.

Another possibility to increase the input power is to look for discharge configurations for which the voltage can be increased. This is possible with the so-called high-voltage HCD (HV HCD), where the discharge is obstructed such that, depending on the discharge geometry an increase of voltage can take place. Different HV geometry designs were studied: hollow-anode-cathode [19], segmented [20] and helical [21, 22]. Using the high-voltage HCD higher laser power and better laser performance is obtained.

Apart from increasing the power, the metal atom density can also be increased by adding small admixture of gases with better sputtering yields to the main gas. A well-known example is the admixture to He discharges. With a pure He discharge it is not possible to achieve laser oscillation. The reason is that due to low mass of He ions the sputtering rate is low. By adding several percents of Ar, Ne, Xe or Kr the laser operation can be improved significantly [3, 5]. With respect to Ne discharges the situation is less clear. The Ne ion sputtering efficiency is much better than that of He. Nevertheless it was reported that the addition of small quantities of Ar to Ne discharge improves the laser performance [23]. However, according to other authors [11] the addition of Ar to the Ne discharge decreases the laser output power.

Apart from the laser power, other important issues related to the laser performance are the discharge stability and efficiency. In HCDs these features strongly depend on the shape and the dimensions of the electrodes. Depending on the relative position of the electrodes — the hollow cathode and the anode — the HCDs can be divided in two groups: longitudinal and transverse HCDs. Both types, as well as their variants, are successfully used for the excitation of laser lines in various gas mixtures.

The transverse HCD possesses very good axial homogeneity. The most popular variant of this HCD-type is the so-called slotted discharge. Examples are shown schematically in figure 2.1. The cathode design shown in figure 2.1 is used in the only available commercial

Figure 2.1: Schematic drawing of the cross section of transverse HCD geometries. The hollow cathode and the anodes are parallel and perpendicular to the plate of drawing, so that the transport of charged particles is transverse to the optical axis.
sputtering hollow cathode laser [24]. The active volume is a rectangular slit cut along a metal rod while the anode is placed parallel to the slit, above it. The dimensions of the slit are optimised empirically. This structure has been used in many experiments [25, 26]. Due to the cathode wall sputtering, the rectangular shape of the cathode changes in time, which deteriorates the laser operation conditions. The main problem of the transverse design is that it is sensitive to arc formation, especially at high input power levels which are needed for sputtering HCD metal vapour lasers. To overcome these problems, different variants of the transverse HCD have been studied. The best results are obtained using the high-voltage variants — the hollow-anode-cathode [19] and the segmented HCD [27]. The transverse segmented HCD has been studied in detail both experimentally and by modelling [27, 28] and the optimal ratio of the cathode and anode surfaces is determined [29].

This paper is devoted to the longitudinal HCD, which has a structure consisting of successive hollow cathodes and hollow anodes. Such HCD geometry is typical for longitudinal sputtering metal vapour lasers [16, 17, 30, 31]. The discharge stability of this laser class is much better than that of the transverse HCD. On the other hand, it is known that these devices may suffer from axial non-uniformities, which may strongly affect the excitation efficiency and thus the laser operation.

In a series of publications, Mizeračzyk et al [32, 33, 34] reported the plasma parameters of transverse and longitudinal HCDs and measurements of the axial current distribution and plasma parameters in longitudinal HCDs [32, 33]. They showed that the plasma properties of this type of discharge may vary strongly along the cathode axis and that, due to the inhomogeneity of most of the plasma parameters, a non-uniform excitation of laser lines along the cathode axis can be expected. All these measurements were performed on pure helium discharges created in a steel hollow cathode with negligible sputtering. In [35] it was shown that in case of sputtering HCDs the erosion rate of the cathode varies non-linearly $\sim j^{5/2}$ with the current density $j$ so that, as a result of axial current inhomogeneity, the cathode geometry will change during the course of laser operation. This causes changes of the discharge characteristics and a decrease of laser power. Moreover, after a certain operation period the cathode aperture can be blocked by the deposited metal [36, 37]. Various longitudinal designs were tested with the aim to overcome this problem: for instance spherical cathodes and “tulip-like” cathodes [8, 30, 31, 38]. It was found that for some discharge conditions and cathode dimensions, the changes in cathode geometry are of minor importance; this increases the laser lifetime significantly [35, 39, 40].

This paper gives an overview of systematic experimental studies on the longitudinal sputtering HCD with the aim to investigate in detail the process of laser oscillation in these devices. The final goal is to come to a construction of a reliable configuration of a sputtering metal vapour laser. As the main problem of the longitudinal HCD lies in the axial inhomogeneity we have studied the current and plasma species distribution along the cathode length for different discharge conditions, gas compositions and cathode geometries. The optimal cathode dimensions have been determined, and they were found to be such that a nearly homogeneous axial plasma species distribution in the whole volume is created. As a result the optical losses in the discharge are reduced and a more efficient laser excitation is achieved.
All experiments were done with a copper cathode, so that the sputtering of Cu (in He and He-Ar discharges) could be investigated in more detail. Special attention is paid to the oscillation of the 780.8 nm Cu ion line, which has the highest gain of all Cu ion laser lines in the near IR spectral range. The corresponding advantage is that the lasing starts at comparatively low discharge current and due to the high gain of this line, lasing can be obtained for comparatively short laser tubes, making it more convenient to study a great variety of HCD geometries. Because the same mechanism of laser excitation, namely the charge exchange from buffer gas ions to Cu atoms is at play:

$$B^+ + Cu \rightarrow B + Cu^{++}$$  \hspace{1cm} (2.1)

We may regard the oscillation of Cu ion lines in the near IR as a template for the lasing of a Ne discharge in the UV. The study may even indicate how to achieve lasing in the vacuum UV.

2.2 Experimental setup

The experimental investigations were performed using various discharge tubes that were designed, built and studied for this purpose. They can be classified in three main types: the segmented discharge tube (SegmDT), the slotted discharge tube (SlotDT) and the long segmented tube (LongST). The SegmDT was mainly constructed to study the axial distribution of the cathode current, the SlotDT was devoted to the investigation of the axial behaviour of plasma spectroscopic properties, while the design of the LongST was aimed at studying the laser performance. Hereafter, we will outline the three main constructions.

2.2.1 Segmented discharge tube; axial current distribution

The design of the segmented discharge tube (SegmDT) was aimed at studying the axial current distribution. It comprises 19 annular identical electrode segments, each with a length of 3 mm, an inner diameter of 4 mm and an outer diameter of 15 mm (figure 2.2). All the segments are made of high quality oxygen-free copper and isolated from each other by 0.5 mm thick quartz rings. The electrode segments are mounted in a quartz tube with

**Figure 2.2:** Segmented discharge tube design (SegmDT): 19 annular electrode segments, each with a length of 3 mm, inner diameter of 4 mm and outer diameter of 15 mm. Each electrode can be connected independently to the electric power supply, allowing to measure the current through the individual segments.
Chapter 2.

15 mm inner diameter hence the discharge can burn only inside the copper rings. Each electrode can be connected independently to the electric power supply, allowing individual ballasting of the electrodes. This gives a better discharge stability and makes it possible to measure the current through the individual segments. By connecting together several electrode segments, it is possible to configure longitudinal HCDs with different cathode lengths and different anode-cathode (A-C) sequence patterns. With this discharge tube, three main anode-cathode patterns were studied: the asymmetric pattern (AC) — one end segment is an anode and all other segments are inter-connected forming one cathode; the symmetric pattern (ACA) — the two end segments are anodes and all other segments are inter-connected to one cathode; and the successive pattern discharges A(CA)_n.

2.2.2 Slotted discharge tube; spectroscopic measurements

Figure 2.3 represent the slotted discharge tube (SlotDT) used for spectroscopic studies. It consists of a hollow cathode copper cylinder with two copper ring anodes, placed at both ends of the cathode. Along the length of the cathode a 0.5 mm wide slit is cut. The electrodes are mounted in a quartz tube that has a side quartz window adjacent to the cathode slit. This window enables side-on observations of the plasma through the cathode slit so that the plasma emission can be monitored as a function of position along the cathode axis.

The measurement setup is shown schematically in figure 2.4. The light that is emitted through the cathode slit passes through a 1 mm diaphragm and is focused by a lens onto the front end of an optical fibre. The other end of the fibre is mounted at the entrance slit of a Bentham grating monochromator. The signal of the photomultiplier on the selected emission line is monitored by a 200 MHz Tektronix TDS420A oscilloscope and stored on a computer. The diaphragm, the lens and the front-end of the optical fibre are mounted on a table, which can be translated along the discharge tube, parallel to the cathode slit, with steps of 1 mm.

The SlotDT design allows measuring the axial profiles of the spontaneous emission of a number of He, Ar and Cu atom and ion lines.
Variants of these discharge tubes with different cathode lengths (from 10 to 70 mm) and different diameters (from 2 to 6 mm) were constructed. In this way the influence of the hollow cathode dimensions on the spectroscopic properties of the discharge could be studied.

### 2.2.3 Long segmented tube; laser performance

The long segmented tube (LongST) is designed with the purpose to measure the laser output power and to investigate how this parameter depends on the different configurations and control parameters. The LongST is comparable with the SegmDT in the sense that it is flexible in choosing the cathode length and anode-cathode patterns, but it has a longer size (figure 2.5). It comprises a series of 31 cylindrical electrodes (16 electrodes of 10 mm length and 15 electrodes of 20 mm length) with total length of 460 mm. Again all the electrodes can be connected independently to the power supply. The investigated electrode configurations, composed of A(CA)$_n$ sequence patterns with different length of the cathodes, are shown in table 2.1.

The two ends of the quartz tube are cut at Brewster angles and a 1.0 m long laser resonator is formed by two highly reflecting mirrors for the 740 – 800 nm spectral range.
Table 2.1: The investigated electrode configurations. Each configuration \((L_1 \ldots L_8)\) is of the form \(A(CA)_n\) and thus consists of repetition of \(n\) number of cathode segments. The length of the cathode segments \(l\) is varied, so that the total cathode length is also changed \((L = l \times n)\).

<table>
<thead>
<tr>
<th>Laser tube configuration</th>
<th>(L_1)</th>
<th>(L_2)</th>
<th>(L_3)</th>
<th>(L_5)</th>
<th>(L_8)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Single cathode length (l), mm</td>
<td>10</td>
<td>20</td>
<td>30</td>
<td>50</td>
<td>80</td>
</tr>
<tr>
<td>Number of cathodes (n)</td>
<td>14</td>
<td>15</td>
<td>10</td>
<td>7</td>
<td>5</td>
</tr>
<tr>
<td>Full active length (L = l \times n), mm</td>
<td>140</td>
<td>300</td>
<td>300</td>
<td>350</td>
<td>400</td>
</tr>
</tbody>
</table>

In all cases the discharge is excited by 3 ms sinusoidal current pulses with a pulse repetition rate of 3 Hz. The peak values of the laser output power and spontaneous emission intensity at each point are averaged over 30 pulses. All pulses were recorded by a digital 200 MHz Tektronix oscilloscope (TDS 420A) with a 20 MHz cutoff filter and are stored on a computer. A Digikrom spectrograph equipped with a 1200 g/mm grating (blaze wavelength 500 nm) and a CCD camera have been employed to record the laser spectrum in the IR spectral range.

2.3 Results

In search for the optimal operating conditions and discharge geometry, various parameters are determined while their influence on the axial current distribution, the discharge emission and the laser power is investigated. This is done for different values of the total input current and gas pressure, in various gas mixtures and for different HC geometries.

The measurements are done at conditions typical for laser oscillation: an input current density up to 1 A/cm\(^2\) and pressure ranging from 0.9 kPa to 3.2 kPa.

2.3.1 Axial current distribution

Influence of the input current

Figure 2.6 shows the axial current distribution for different values of the input current in pure He discharges. The measurements are done in the segmented discharge tube (see figure 2.2), where a simple AC discharge configuration is constructed, i.e. the first electrode is connected as anode while all the remaining electrodes are connected to the same potential; they act as one and the same cathode with total length of 50 mm.

Figure 2.6 shows that the axial cathodic current distribution is strongly inhomogeneous. It has a maximum near the anode end of the cathode at a distance of about 5 mm from the anode end. The maximum position does not depend on the value of the total input current. Figure 2.6(b), presenting the normalized axial current distribution, shows that increasing the total discharge current does not change the relative current distribution. Most of the current remains concentrated in the first \(\sim 20\) mm of the cathode in the region close to
Figure 2.6: Axial current distribution in the cathode at different values of the total input current in AC discharge in pure He at a constant pressure of 1.5 kPa. Position $x = 0$ is the cathode edge close to the anode.

Influence of the gas mixture

Usually a heavier gas is added to the main gas He in order to increase the sputtering efficiency. As a consequence the Cu atom density and hence the laser power \[3, 5\] will increase. We have investigated the influence of Ar addition on the axial current distribution. Figure 2.7 shows that the addition of Ar does not change the axial current distribution: the same non-uniformity is observed as in the pure He case, exhibiting a maximum in the region close to the anode. We have also measured the axial current distribution in a Ne discharge for the same discharge conditions (current density and gas pressure). Again the same axial inhomogeneity of the discharge current is observed. The current is concentrated in the region close to the anode similarly to He and He-Ar discharges. To conclude: the shape of the axial current distribution does not depend on the chemistry of the discharge.

Different construction patterns

As shown, the axial current distribution in AC construction is concentrated in the region close to the anode in the first 20 mm. It is independent on further increasing of the cathode length. For comparison we also investigated a symmetric (ACA) discharge, meaning that the two end electrodes are inter-connected as anodes while all remaining electrodes in between are connected as cathodes with total length of 50 mm. The current distributions in both configurations (AC and ACA) are presented in figure 2.8(a) and 2.8(b) for different values of the total input current in He-Ar mixture with 5% Ar concentration.
As mentioned before in the AC discharge the maximum cathode current is in the region close to the anode independently on the gas mixture and total input current. It can be seen that in the ACA discharge again the maximum current is concentrated in the regions close to the anodes. Thus the symmetric ACA discharge can be regarded as being composed of two AC discharges, which apparently do not affect each other.

Besides the two main electrode configurations (AC and ACA), a third electrode configuration comprising two successive symmetric discharges (ACACA) is investigated. This is done by connecting the middle segment to the anode potential so that the length of one ACA discharge is decreased. The axial current distribution in this configuration is presented in figure 2.8(c). Since the maximum is located at a distance from the anode, typically 5 mm, shortening the cathode lengths leads to maximum in the middle of the cathode. This suggests that the cathode length should not be too short in order to obtain uniform or nearly uniform distribution. The optimum is reached when the discharges from both edges start to merge.

We can conclude this subsection with the statement that there is an optimum in the cathode segment length. In the case of a diameter 4 mm the segment length is about 20 mm. As we will see later it depends on the diameter of the cathode.

### 2.3.2 Spontaneous emission

In order to study the influence of the axial current non-uniformity on the excitation efficiency of the laser lines, we have measured the spontaneous emission of different He, Ne, Ar and Cu lines through the cathode slit, along the tube length (see figure 2.3).

Figure 2.9 presents the axial sidelight emission intensity distribution along the cathode.
length for Cu and He atom lines in a He-Ar discharge with an AC configuration. The total gas pressure is 2.3 kPa and the constant input current equals 2.5 A. For comparison, the axial current distribution is shown as well, measured in the SegmDT at the same operating conditions, meaning that apart from the pressure and chemical composition, the dimensions of the cathode for both tubes are the same: a cathode length of 50 mm and a diameter of 4 mm.

The axial shape of the helium atom line intensity almost coincide with that of the longitudinal current distribution, reaching half of its intensity at the middle of the cathode. The inhomogeneity of the Cu atom line intensity profile is stronger, exhibiting a maximum at 3 – 5 mm from the anode, decreasing to almost zero at the middle of the cathode.

The well-pronounced maximum of the Cu atom line close to the anode indicates that the sputtering efficiency in that region is higher and that it decreases with increasing distance.
Figure 2.9: Axial emission intensity profiles for Cu and He atom lines in AC configuration of a SlotDT compared to the current distribution in the SegmDT. The pressure is 2.3 kPa and the input current is 3 A.

to the anode. Due to the intensive sputtering in this region the side quartz window, through which the sidelight emission is monitored, is covered with a thin copper layer after several hours of operation. The thickness of this layer has more or less the same longitudinal distribution as the copper atom line intensity. This confirms that the sputtering of the cathode material is strongly non-uniform along the cathode length.

As a conclusion, the axial profile of the buffer gas atom line intensity coincides with the axial current profile, showing a linear dependence on the current distribution. For the axial metal atom profile however, the dependence is not linear: the emission profile decreases faster than the current profile with increasing distance from the anode. A non-linear dependence of the sputtering and current density was previously discussed by J. Yu et al [35], as mentioned in the introductory section. They have shown that the sputtering rate varies non-linearly as $j^{5/2}$ with current density $j$.

Influence of the gas mixture

In order to check if the axial emission distribution is influenced by the addition of Ar we have measured the axial emission intensity for different Ar concentrations. In figure 2.10 the axial profiles of different Cu, He and Ar lines are shown for Ar concentrations from 0 to 10%. Increasing the Ar fraction the Ar emission intensity increases, as expected (figure 2.10(c)). But also the intensity of Cu lines rises (figures 2.10(b) and 2.10(d)). This proves that the sputtering is mainly by Ar ions [41]. Contrary to the increased emission intensity of Ar and Cu, the intensity of He lines decreases.

Recently, these observations were studied by means of numerical modelling [42] (chapter 5 of this thesis). Both experimental and modelling results confirm that the addition of
heavier atoms, such as Ar, is required to produce efficient sputtering. Increasing the Ar concentration leads to an increase of \( \text{Ar}^+ \), Cu atom and Cu ion densities and thus, to potentially higher population of the upper laser level. However, the \( \text{He}^+ \) density decreases. When the role of Ar ions becomes too dominant, the rate of the asymmetric charge transfer processes (equation (2.1)) between Cu atoms and He ions decreases, hence the population of the upper laser level decreases. The results predict that there must be an optimal Ar concentration above which the laser power decreases. These model predictions are in agreement with experimental observations.

Influence of the geometry

In He-Ar mixtures, as shown above, a strong nonuniform current distribution is observed at cathodes with \( l > 20 \text{ mm} \). Similar distributions of the axial spontaneous emission intensity of the 587.6 nm HeI and 510.6 nm CuI lines are observed, as shown in figure 2.11 for a
ACA discharge at different cathode lengths. In order to compare cathodes with different dimensions the current density of 0.5 A/cm² is kept constant. For longer cathodes, the emission intensity shows well-pronounced maxima close to the anodes while the emission decreases rapidly with increasing distance to the anodes. As a result, the plasma is most intense in the regions close to the anodes, while the central part of the cathode cavity remains inactive and does not contribute efficiently to the excitation of the laser lines. Shortening the cathode length results in a more uniform current distribution and also in a flattening of the intensity profiles. The two separated intense regions approach each other, reducing the concavity in the middle of the cathode. In cathodes of 10 mm and 20 mm length the axial distributions of the intensity of both He and Cu atoms is more or less homogeneous.

In order to study the influence of the cathode diameter on the discharge uniformity we have compared the axial spontaneous emission profiles for different diameters (figure 2.12) in He-Ar discharges. The length of the cathode is 70 mm in all cases, while the parameter pd is kept constant equal to 9 kPa cm, meaning that with increasing diameter d the pressure p must be decreased.

For small diameters, a strong axial non-uniformity is observed, similar to what is observed for longer cathodes. Again, the plasma is more intense close to the anode edges and can be regarded as two separate discharge regions that do not affect each other. Increasing the diameter leads to an approach (merge) of the two intensive regions from both sides, reducing the concavity in the middle of the cathode. Increasing the cathode diameter has similar effect on the plasma as decreasing the length of the cathode — both lead to a more uniform plasma distribution. This demonstrates that the plasma uniformity depends on the aspect ratio of length and diameter, rather than on length or diameter solely.
2.3.3 Laser action

In spite of the relatively short (only 50 mm) length of the active medium we could obtain lasing of the 780.8 nm IR Cu ion line in the SegmDT (figure 2.2). Lasing with the SegmTD was achieved using a simple ACA configuration filled with a He-Ar mixture with an Ar concentration in the range 1 – 10% and with gas pressures between 1 kPa and 3 kPa. The laser oscillation of the 780.8 nm IR Cu ion line is easily achieved due to its high gain. That is why this line is often used in optimisation studies of the discharge geometry and excitation conditions.

In figure 2.13 the laser output power is shown as a function of the gas pressure at a constant current of 3 A in a He-Ar mixture. The optimal gas pressure for the SegmDT is

![Graph showing laser output power dependence on gas pressure.](image)

**Figure 2.13:** Laser output power dependence on the gas pressure.
Figure 2.14: Laser output power dependence on the discharge current at different Ar concentration for AC.

found to be in the range from 1.7 to 2.5 kPa.

Figure 2.14 shows the dependence of the output laser power on the discharge current at different concentrations of Ar. The total pressure is constant and equals 2.3 kPa, which is in the range of the optimal pressure. The laser oscillation starts at $\sim 1.1$ A and the laser power increases almost linearly with current. However, for current values above $\sim 3 - 4$ A a tendency to saturate is observed. The same behaviour is observed in AC configuration, but the threshold for lasing is higher ($\sim 1.5$ A) and saturation is observed at lower values of the input current ($\sim 2.5$ A).

The fact that the ACA configuration offers a better laser performance than the AC configuration can be understood from the fact that in the AC case only about 10 mm of the cathode length contributes to the lasing, while for the ACA discharge the active laser length is nearly doubled (see figure 2.8). For both discharges the current density at which laser output power begins to saturate is about 1 A/cm$^2$.

Below 1% of Ar admixture laser oscillation is not observed. This indicates that addition of heavier atoms is required for lasing since they are needed for sputtering to bring more Cu atoms into the discharge. The highest laser power is observed at an Ar concentration of 5% which suggests that the presence of too much Ar is destructive for the laser action. As mentioned above, the modelling results [42] (chapter 5 of this thesis) demonstrate that the He/Ar ratio has a significant impact on the plasma characteristics. They also show that, apart from enhancing the sputtering rate and thus increasing the Cu atom density, the addition of Ar will also lead to a reduction of the He ion density. This tends to reduce the rate of the asymmetric charge transfer from He ions and Cu atoms:

$$\text{He}^+ + \text{Cu} \rightarrow \text{He} + \text{Cu}^{++}.$$
Thus, this leads to a reduction of the population of the upper laser level. So apparently the optimum Ar concentration is the value at which the product of the concentrations of Cu atoms and He ions is highest.

The existence of optimal conditions (input current, pressure and Ar addition) limits the possibilities to increase the laser power by increasing the input power, or by increasing the sputtering efficiency (by increasing pressure or adding heavier atoms). The most effective way to increase the laser power is by increasing the active volume.

In order to study the influence of the cathode length on the laser power we employed the long segmented tube (see figure 2.5). This LongST, being longer than the SegmDT (460 mm versus 50 mm), is specially designed for this purpose. Simultaneous laser oscillation of 7 IR Cu ion lines is observed at conditions typical for lasing: a total gas pressure of $1 - 3.0 \text{kPa}$ with an Ar concentration between $4 - 7\%$. All laser lines respond in the same way to changes in the operating parameters, although they may have different threshold current for lasing. The strongest line in this spectral range, the Cu ion line (780.8 nm), has the lowest threshold, so that the further investigation will be devoted to the behaviour of this line.

Lasing is observed in all studied electrode configurations with cathode segment lengths of: $l = 10$, 20, 30, 50 and 80 mm (see table 2.1). Figure 2.15 presents the V-I characteristics for all the configurations of the LongST (see figure 2.5, table 2.1).

The discharge voltage depends on the configuration. For configurations with longer cathodes ($l = 20 - 80 \text{mm}$) the V-I characteristics have a shape that is typical for conventional hollow cathode discharges: the voltage is nearly constant and changes from 360 V for $l = 80 \text{mm}$ to 400 V for $l = 20 \text{mm}$. However, the V-I characteristic of the $l = 10 \text{mm}$ configuration is different. It increases from 400 V at $0.15 \text{A/cm}^2$ up to 500 V at $0.6 \text{A/cm}^2$. 

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**Figure 2.15:** V-I characteristics at different length of the cathode segments $l$. 

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Such a V-I characteristics is comparable to that of a so-called high-voltage construction [19, 20, 21, 22] as discussed in the introductory section. Therefore, the $l = 10\,\text{mm}$ configuration can be regarded as a transition to a high-voltage discharge operation.

Figure 2.16 presents the laser power as a function of the gas pressure for different configurations. The curves are in all cases typical for HCD lasers, showing that an optimal gas pressure exists for the specific discharge configuration. The behaviour of the $l = 80\,\text{mm}$ configuration is not given in this figure. The reason is that despite the lower voltage the discharge tends to arc at low input powers and lasing could only be achieved in a very narrow pressure range around 1.6\,kPa.

Figure 2.17 gives the output laser power as a function of the input power density for the studied constructions for a pressure of 1.6\,kPa. The comparison is made at equal power density because, as already shown, the voltage depends on the configuration (cathode size). The laser power rises almost linearly with input power density and no tendency to saturate is observed up to the limit of the power supply.

The discharge stability is influenced by the cathode segment length. The discharge is most stable at small $l$, while as mentioned before, at $l = 80\,\text{mm}$ the laser output power is limited due to arc formations. The highest laser power is observed at $l = 20\,\text{mm}$, where the spectroscopic measurements show axially uniform distributions of the current and the spontaneous emission. The laser power at longer cathodes is lower, which also indicates that some part of the HC medium does not participate effectively in the excitation of the laser levels. The lowest laser power obtained at $l = 80\,\text{mm}$ also indicates that apparently additional optical losses are introduced, due to the longer inactive volume.
2.4 Conclusions

Results of a systematic experimental study on the longitudinal sputtering HCD for laser application is presented. The final goal is to optimise the operating conditions and HC geometry in order to build a reliable and efficient sputtering metal vapour laser. The experimental observations can be compared with modelling results. The model validated in this way can be used as a design tool to study that part of the parameter space that has been explored experimentally.

The discharge performance is experimentally studied in detail by investigating the axial current distribution, the spontaneous emission distribution and the laser performance and how these are influenced by the operating parameters such as input current, gas composition and electrode geometry. The experimental investigations were performed using various discharge tube designs that were built for this purpose.

The results show strong axial inhomogeneities of the discharge current, the plasma distribution and the spontaneous emission.

The cathode current is concentrated in the regions close to the anodes and the position of the maximum is at a distance (around 5 mm) from the anode edge. The axial current non-uniformity can not be reduced by increasing the total input current and the shape of the axial current distribution does not depend on the chemistry of the discharge. So that the axial current profile is independent on the input power and gas composition.

The non-uniform axial current distribution has influence on the discharge excitation efficiency. The measured intensity of different lines is axially inhomogeneous exhibiting a high intensity in the regions close to the anodes which decreases with increasing the distance from the anode. The axial profile of the buffer gas atom line intensity coincides with the axial current profile, showing a linear dependence on the current distribution. For
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the axial metal atom profile however, the dependence is not linear: the emission profile decreases faster than the current profile with increasing distance from the anode. This is in agreement with previous observations by J. Yu et al [35], where they have shown that this non-linearity of the sputtering rate varies as \( j^{5/2} \) with current density \( j \).

The non-uniformities are mainly influenced by the electrode configuration and dimensions. Decreasing the cathode length results in decreasing the concavity in the middle of the cathode so that a nearly uniform plasma distribution can be obtained. The same non-uniform distribution is observed at small diameters. Decreasing the diameter has the same effect as increasing the cathode length — the concave non-uniformities increase. This suggest that the plasma uniformity depends on the aspect ratio of length and diameter, rather than on length or diameter independently.

These observations are confirmed by the laser oscillation of the 780.8 nm Cu ion line in a He-Ar gas mixture. The optimal gas pressure for lasing is determined to be 2.3 kPa. The He/Ar ratio has a significant impact on the plasma characteristics. The addition of heavier atoms is required for lasing but the presence of too much Ar is destructive for the laser action. The highest laser power is observed at an Ar concentration of 5%. The existing optimal concentration of Ar limits the possibility to increase the laser power by increasing the sputtering efficiency. Moreover, it was found that an optimum current density of \( \sim 1 \text{ A/cm}^2 \) exists, above which the output power tends to saturate. This limits the increase of the laser power by increasing the input excitation power. Another possibility to increase the laser power is to build a laser with a longer active length. But due to the axial inhomogeneity of the longitudinal discharge resulting from the different distance between the anode and the respective cathode point, an optimal cathode length for each particular discharge conditions exists, which depends on the cathode diameter.

Summarising, the existence of optimal conditions (input current, pressure and Ar addition) limits the possibilities to increase the laser power by increasing the input power, or by increasing the sputtering efficiency (by increasing pressure or adding heavier atoms). The most effective way to increase the laser power is by increasing the active volume. Based on the results of these experiments we have demonstrated that the most efficient laser oscillation is achieved when the laser active volume comprises a series of anodes and cathodes, each cathode with a length not longer than 20 mm for a cathode diameter of 4 mm.

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References

Chapter 2.

Abstract. In this thesis a multi-fluid approach is employed to model the plasma created in a hollow cathode. The model used is the PLASIMO’s sub-model MD2D. This is a time-dependent two-dimensional fluid code based on the balance equations derived from the Bolzmann equation — the continuity equations for the evolution of the relevant active species and the electron energy balance. These fluid equations are coupled with the Poisson equation for computation of the electric field. This chapter gives a brief overview of the physics behind the model, describes the discretisation aspects of the code, and outlines the extensions and improvements done to the code.
Chapter 3.

3.1 Introduction

A model in physics is a system of mathematical equations that are adapted, combined and solved to describe some aspects of reality. The solution of the model equations is intended to reproduce observed and measured phenomena, thus explaining these in terms of fundamental physics. Another task of models is to predict phenomena, so that they can be used to guide experimental procedures. Several types of models can be distinguished. Here we mention the division into Grand and specific models. A Grand model simulates the plasma as a whole in conjunction with its direct environment (e.g. the input and output). The input of the model is, just as in practice, formed by the set of plasma control parameters such as the value of the electrical current, the mass flow, the inlet pressure, fill-gas composition and the vessel configuration. The output is the set of internal plasma properties like the distribution of the voltage and electron temperature, the density fields of the electrons, radicals and light emitting species. From that information the plasma products can be deduced; for instance the efflux of heat, light and radicals. In a Grand model the route from input to output is guided by the laws of nature while the transport coefficients and source terms are based on well documented collision theories and experimental data. There is no place for fudge factors! A specific model normally deals with the relation between two or more (set of) internal quantities. For instance the determination of rates of (composed) reactions assuming known values of the electron density and temperature. Another example is a Boltzmann solver that provides the electron energy distribution function (EEDF) and certain transport coefficients for a situation with given electric field strength and degree of ionization.

The PLASIMO platform developed at the Eindhoven University of Technology can be seen as a toolkit that facilitates the construction of specific, but also Grand models. The code MD2D is a part of PLASIMO dedicated to the construction of Grand models for plasmas for which the gas heating and gas flow are less or not important. This study is mainly based on models derived with MD2D.

The majority of gas discharge (plasma) models are built upon the Boltzmann equation. In principle, the combination of the Boltzmann equation, together with the Maxwell equations, needed for computation of the electromagnetic field, describes the physics of many discharges completely provided that this set of equations is equipped with suitable boundary conditions. In practice, however, the Boltzmann equation is unwieldy and cannot easily be solved without making significant simplifications.

One of the methods for solving the Boltzmann equation is the kinetic approach. Kinetic models are time and spatially dependent solutions of the Boltzmann equation, which produce electron and ion velocity distributions by applying statistical techniques such as the Particle-in-Cell or Monte-Carlo method. These models give kinetic information by following the trajectories of a large number of individual particles. However, most discharges contain too many particles to follow them all individually. An often used technique is to follow a set of particles that is taken to be representative. The kinetic method is capable of treating the behaviour of a set of different species with a minimal number of assumptions, but this is in the most cases associated with the cost of a significant calculation time.
Another approach to describe plasmas is with the \textit{fluid} approach. Fluid models describe the plasma species in terms of smoothed average hydrodynamic quantities like the particle, momentum and energy densities. Fluid models solve the moments of the Boltzmann equation in time and space. The EEDF can be calculated off-line. This provides the electron transport and rate coefficients that can be coupled to the fluid model.

A more general fluid approach is the two-fluid or \textit{multi-fluid} plasma method. It is based on the fact that the electrons behave differently from the atoms, molecules or ions and the species are described separately. The (multi-) fluid approach, although being less accurate than the kinetic method, usually has shorter computational times. That allows higher dimensionality (2D, 3D) and the treatment of more different physical aspects with the model.

Fluid models are often accurate when sufficient collisions take place, i.e. when the mean free path of the particles is much smaller than the plasma size. On the other hand, a kinetic description is often necessary in cases where the mean free path exceeds the plasma size.

The advantages of both type of models can be combined in so-called \textit{hybrid models}. Hybrid models use the kinetic approach to handle the non-local transport of electrons and ions and to derive transport coefficients of charged species. The fluid approach is simultaneously applied in order to provide the density of charged species and the electric field distribution. Hybrid models have, among others, been developed to simulate plasmas of complex chemical compositions and to describe discharges composed of regions ruled by different mechanisms.

In this work we choose to describe the plasma in a HCD using the \textit{multi-fluid} approach. In the future this fluid model can be coupled with a kinetic Monte-Carlo model to describe highly non-equilibrium regions of the plasma more precisely.

We use PLASIMO’s MD2D sub-model. The Micro-Discharge 2-Dimensional (MD2D) code is a two-dimensional fluid code that is suitable for the simulation of plasmas that deviate from thermal equilibrium. Moreover it allows to describe plasmas that are not quasi-neutral.

The code was initially created by Hagelaar [1] for the simulation of the micro-plasmas used in display technology. A significant reorganisation and extension was performed by Van Dijk and Brok [2]. The code has been used for many different plasma modelling studies, such as the ignition of fluorescent lamps [3], the plasma needle for biomedical applications [4], simulations of plasma breakdown of a parabolic electrodes configuration [5], DBD discharges [6, 7, 8, 9, 10], and the pulsed discharge nozzle [11].

This chapter gives a brief overview of the physics behind the model, describes discretisation aspects of the code and gives references to relevant documentation and publications.

In section 3.2 the general transport equations and their physical interpretation are briefly presented. Section 3.3 gives the transport equations in the form as they are used in the model, together with the assumptions made in the derivations. The next sections present the numerical solution of the transport equations.
3.2 Transport equations

The model is based on the balance equations for the particle, momentum and energy densities. These can be derived from the Boltzmann equation. Details of this procedure can be found in many textbooks, such as [12] and [13]. For completeness we will first present the general forms of the transport equations and proceed by giving the assumptions that lead to the equations that are used in MD2D.

The Boltzmann equation describes the spatial and temporal evolution of the density \( f_p(x, v, t) \) of particles of type \( p \) in the phase space, which is spanned by the configuration space, denoted by the vector \( x \), and velocity space, represented by \( v \). It can be written as [14, p. 48]:

\[
\frac{\partial f_p}{\partial t} + v \cdot \nabla_x f_p + a \cdot \nabla_v f_p = \left( \frac{\delta f_p}{\delta t} \right)_{CR},
\]

where \( a \) is the acceleration of the particles under the influence of external forces. The right hand side of the equation denotes the effect of collisions and radiation on the phase space density function.

The first three moments of the Boltzmann equation are obtained by multiplying it by \( m_p, m_p v \) and \( \frac{1}{2} m_p v^2 \) respectively, and integrating the result over the entire velocity space. This gives the equations of mass, momentum and energy conservation.

Integrating equation (3.1) over velocity space results in the zeroth moment, the so-called particle balance equation. For a species \( p \) it reads:

\[
\frac{\partial n_p}{\partial t} + \nabla \cdot \Gamma_p = S_p,
\]

where \( n_p \) is the density of species \( p \), \( \Gamma_p \) its flux density, and \( S_p \) the net source.

The first moment of the Boltzmann equation, the momentum balance equation, is obtained by multiplying equation (3.1) with the species’ momentum \( m_p v \) and integrating the result over velocity space. Considering that the acceleration of particles is under the influence of electric field only, \( a = q_p E/m_p \), gives:

\[
\frac{\partial n_p m_p u_p}{\partial t} + \nabla \cdot (n_p m_p u_p u_p) = -\nabla \cdot P_p + q_p n_p E + R_p,
\]

where \( u_p \) is the average velocity \( u_p = \langle v \rangle_p \), \( P_p \) the pressure tensor, \( q_p \) and \( m_p \) the charge and mass of the species and \( E \) the electric field. The friction vector \( R_p \) denotes the rate of change of the mean momentum per unit volume due to collisions. The reduced form of this equation, which is used in MD2D, will be discussed in section 3.3.

Multiplication of the Boltzmann equation (3.1) with the kinetic energy \( \frac{1}{2} m_p v^2 \) and integrating the result over the velocity space results in the second moment, the energy balance:

\[
\frac{\partial n_p \varepsilon_p}{\partial t} + \nabla \cdot n_p \varepsilon_p u_p + \nabla \cdot (P_p \cdot u_p) + \nabla \cdot q_p = \varepsilon_s,
\]
where $\varepsilon_p$ is the mean kinetic energy of species $p$. We can define the temperature $T_p$ of species $p$ by equating $\varepsilon_p = \frac{3}{2} k_B T_p$. In case that the kinetic energy of particles is distributed according to Maxwell, $T_p$ is the usual thermodynamic temperature. The first term on the left hand side of equation (3.4) represents the total rate of change of the kinetic energy density of species $p$, the second is the change in the kinetic energy density due to macroscopic transport, the third term gives the work done by the kinetic pressure and viscous dissipation, and the fourth term represents the change in the energy density due to heat flux. At the right hand side we find $S_\varepsilon$ that represents the energy source density due to collisions and EM heating.

3.3 MD2D

The balance equations given in the previous section will now be simplified and prepared for their use in the model.

One of the basic assumptions of MD2D is that the gas heating is not important, meaning that the buffer gas, formed by the neutral majority species, is stationary in time and uniform in space; i.e. the gas temperature $T_g$, pressure $P_g$ and density $n_g = P_g/k_B T_p$ are independent on the time and spatial position. That means that the features of the buffer gas do not need to be calculated with the three balance equations given above.

The defined active species for which moments of the Boltzmann equation are needed, are the electrons, ions, atoms and (excited) atoms of the minority species. The number densities of these active species is described by the relevant species balance equations.

The momentum balance of the active species will be simplified to the drift-diffusion approach. The energy description of the plasma is based on a splitting between the electrons and the heavy particles. For the heavy particles we assume that they will attain the temperature of the background gas. So only the energy balance for the electrons needs to be solved. This energy equation – the electron energy equation – takes account of the fact that the electrons will carry most of the current in the plasma. Consequently they will receive most of the heat generated by Ohmic dissipation. Moreover, the thermal exchange between electrons and heavy particles is inefficient as only a fraction of the thermal energy difference of the order of $m_e/m_h$ is exchanged per collision. This means that the electrons will have a temperature that is much higher than that of the heavy particles $T_e \gg T_h$.

Particle balance

For each active species $p$ the time evolution of the density is described by the particle balance in the same form as (3.2)

$$\frac{\partial n_p}{\partial t} + \nabla \cdot \Gamma_p = S_p,$$

(3.5)
where \( \Gamma_p = n_p u_p \) is the flux density. The net source term \( S_p \) is determined by the reactions occurring in the discharge:

\[
S_p = \sum_r c_{p,r} R_r, \tag{3.6}
\]

where \( c_{p,r} \) is the net stoichiometric number of particles of species \( p \) created in one reaction \( r \). The value \( c_{p,r} \) is positive if the reaction leads to the production of \( p \), negative if particle \( p \) is destroyed and zero if the reaction does not change the number density of \( p \).

The reaction rate \( R_r \) is proportional to the densities of the reacting particles: for two body collisions \( R_r = k^{(2)} n_p n_q \), for three body collisions \( R_r = k^{(3)} n_p n_q n_l \) and similarly for radiative decay \( R_r = k^{(1)} n_p \). The proportionality constant \( k \) is the reaction rate coefficient.

### Momentum balance

The flux density in equation (3.5) is solved in the drift-diffusion approach, resulting from the momentum balance equation (3.3) after some simplifying approximations.

Firstly, the viscosity term is neglected and pressure is assumed to be isotropic. This allows to replace the tensor \( \mathcal{P}_p \) by the scalar pressure \( P_p \), which can be related to the density \( n_p \) and temperature \( T_p \) of the species via the ideal gas law.

The time dependent term in equation (3.3) can be assumed to be zero on time scales much larger than the collision time. Furthermore, we neglect the inertia term, the second term at the left hand side of equation (3.3). With these simplifying approximations the momentum equation becomes:

\[
- \nabla (n_p k_B T_p) + q_p n_p E + R_p = 0. \tag{3.7}
\]

An expression for the momentum transfer rate by collisions is given by [12]:

\[
R_p = -n_p m_{pq} \sum_p \nu_{pq} (u_p - u_q), \tag{3.8}
\]

which assumes that the force per unit volume exerted on the particle of type \( p \) due to collisions with particle of another type \( q \) is proportional to the difference between the mean velocities of these particles. In this proportionality we find \( \nu_{p,q} \), is the momentum transfer frequency of the particles of type \( p \) for collision with particles of type \( q \). In view of the dominance of the buffer gas species and taking into account that this background gas has no velocity, so that \( u_0 = 0 \), equation (3.8) reduces to

\[
R_p = -n_p u_p (m_{p0}\nu_{p0}), \tag{3.9}
\]

where the properties of the buffer gas are labelled with zero as subscript.

Assuming that the temperature is uniform (\( \nabla T = 0 \)), equation (3.7) gives the drift-diffusion equation [15]:

\[
n_p u_p = \frac{q_p}{m_{p0}\nu_{p0}} E n_p - \frac{k_B T_p}{m_{p0}\nu_{p0}} \nabla n_p. \tag{3.10}
\]
The first term at the right hand side gives the flux due to the electric field (drift) and the second term represents the flux due to density gradients (diffusion). This equation can be written in the more common form:

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

(3.11)

where two transport coefficients have been introduced: the mobility $\mu_p$ and the diffusion coefficient $D_p$:

$$\mu_p = \frac{q_p}{m_p \nu_0}, \quad D_p = \frac{k_B T_p}{m_p \nu_0}.$$  

(3.12)

Note that $\mu_p$ is a signed quantity; it is negative for negatively charged species.

Equations (3.12) automatically leads to the Einstein relation:

$$D_p = \frac{k_B T_p \mu_p}{q_p},$$

(3.13)

which can be used to deduce the diffusion coefficient from the mobility. Note that it has been derived under the assumption that the species have Maxwellian energy distribution functions. In that case for the electrons we have seen (in section 3.2) that

$$k_B T_e = \frac{2}{3} \varepsilon.$$  

(3.14)

For ions in a strong electric field, the distribution function is usually not Maxwellian, but equation (3.13) can still be used if $T_p$ is replaced by an effective temperature [16]:

$$k_B T_i = k_B T_g + \frac{m_i + m_g}{5m_i + 3m_g} m_g (\mu E)^2,$$

(3.15)

where $T_g$ is the gas temperature, $m_i$ and $m_g$ the ion and gas particle masses, respectively.

For positive ions, the local field approximation is used. This assumes a direct relation between the particle energy distribution and the local electric field, hence the transport coefficients are regarded as being functions of the reduced electric field $E/N$:

$$\mu_i = \mu_i(E/N), \quad D_i = D_i(E/N).$$

(3.16)

A compilation of measured ion mobilities in various pure gases is given in Ref. [16] as a function of $E/N$. The mobility in gas mixtures is given by Blanc’s law [17]:

$$\frac{1}{\mu} = \sum_i \frac{1}{f_i \mu_i},$$

(3.17)

which relates the mobility $\mu$ in a mixture to the pure gas mobilities $\mu_i$ and to the fractional gas concentrations $f_i$. 

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For electrons, substantial deviations from the local field approximation can be observed as a result of the poor energy transfer in electron-neutral collisions, which is due to the huge mass difference. Therefore, rather than using relation (3.16), the electron transport coefficients are taken to be functions of the mean electron energy:

$$\mu_e = \mu_e(\varepsilon), \quad D_e = D_e(\varepsilon).$$  \hspace{1cm} (3.18)

The functions (3.18) can be obtained from cross section data by solving the electron Boltzmann equation or from Monte-Carlo simulations. In the present model these functions are obtained from the freeware Boltzmann equation solver BOLSIG+ [18], which solves the Boltzmann equation using the two-term approximation. These input data for the fluid model are presented in the form of lookup tables as a function of the mean electron energy.

Energy balance

Using the electron energy density $n_\varepsilon = n_e \varepsilon$, where $\varepsilon$ is the mean electron energy, the electron energy equation becomes:

$$\frac{\partial n_\varepsilon}{\partial t} + \nabla \cdot n_\varepsilon \mathbf{u}_e + \nabla \cdot P_e \mathbf{u}_e + \nabla \cdot \mathbf{q}_e = S_\varepsilon.$$  \hspace{1cm} (3.19)

where $\mathbf{u}_e$ is the mean electron velocity and $\mathbf{q}_e$ the thermal flux density. The effective source term $S_\varepsilon$ for electron energy is given by:

$$S_\varepsilon = \Gamma_e \cdot \mathbf{E} - n_e \sum_r \varepsilon_r k_r n_r - L_\varepsilon,$$  \hspace{1cm} (3.20)

where the first term represents heating by the electric field and the second the energy loss in inelastic collisions. The summation runs only over the electron impact reactions, with $n_r$ the density of the target particles and $\varepsilon_r$ the threshold energy. The last term $L_\varepsilon$ in the equation represents the energy loss due to elastic collisions.

Combining the divergence terms into an energy transport term casts the electron energy balance in the same form as the particle balance:

$$\frac{\partial n_\varepsilon}{\partial t} + \nabla \cdot \Gamma_\varepsilon = S_\varepsilon,$$  \hspace{1cm} (3.21)

where the transport term is:

$$\Gamma_\varepsilon = n_\varepsilon \mathbf{u}_e + P_e \mathbf{u}_e + \mathbf{q}_e.$$  \hspace{1cm} (3.22)

Using the relation between the mean electron energy and the electron pressure $P_e = \frac{2}{3} n_e$ and substituting $\mathbf{u}_e = \Gamma_e/n_e$, the first two terms in equation (3.22) can be combined as:

$$n_\varepsilon \mathbf{u}_e + P_e \mathbf{u}_e = \frac{5}{3} \varepsilon \Gamma_e.$$  \hspace{1cm} (3.23)
The heat flux density (the last term in equation (3.22)) is assumed to be proportional to the gradient of the electron mean energy, according to [19] and [12]:

$$q_e = -\frac{5}{3} n_e D_e \nabla \varepsilon.$$  \hfill (3.24)

Substituting the flux density for electrons (equation (3.11)) into equation (3.23) and combining the result with equation (3.24), gives, after some algebra, the expression for the electron energy flux density in the form as used in MD2D:

$$\Gamma_\varepsilon = \frac{5}{3} \mu_e E n_e - \frac{5}{3} D_e \nabla n_e.$$  \hfill (3.25)

The transport equations — the continuity equation (3.5), the momentum balance equation (3.11), and the energy balance equation (3.21) — are coupled to Poisson equation for computation of the electric field:

$$\nabla \cdot (\varepsilon E) = -\nabla \cdot (\varepsilon \nabla V) = \rho,$$  \hfill (3.26)

where $\varepsilon$ is the dielectric permittivity, $V$ the electrostatic potential, and $\rho$ the space charge density given by:

$$\rho = \sum_p q_p n_p.$$  \hfill (3.27)

Note that $\rho$ may be discontinuous and that surface charges may be present.

3.4 Numerical solution of the fluid equations

3.4.1 Grid layout

MD2D uses the control volume method for solving the species densities (equation (3.5)), the electron energy (equation (3.21)) and the electrostatic potential (equation (3.26)).

We use a 2-dimensional uniform grid consisting of grid cells with size $\Delta x$ and $\Delta y$ in $x$- and $y$- directions. In every individual grid cell we define material. It can be plasma, an electrode material with a certain voltage or a dielectric material with a certain permittivity. The type of the material in the cell is represented by an index in the input file. The transport equations are solved only for the plasma region. The Poisson equation is solved in the entire grid, except the electrode regions. Then from the material grid thus constructed, a nodal grid is derived.

Figure 3.1 shows the material grids and layout of the computational nodal grid. The physical grid cells are marked with dashed lines with size $(\Delta x, \Delta y)$. Nodal points are marked with points that lie at the corners of the material cells. Around each nodal point a control volume cell (CV) is constructed, so that the nodal point represents the centre of the CV.
Given a nodal point C, the nodal west and east neighbours, denoted as W and E lie at a distance $\Delta x$ from the nodal point C. In addition, the control volume boundary points, called w and e, are located halfway between the nodal points. In the figure they are marked as arrows. The same applies for the south and north neighbours (S and N) and the boundaries of the control volume (s and n). Note that capital letters always refer to nodal points, whereas lowercase letters refer to locations on the cell boundary grid.

The values of the scalar quantities are evaluated at the nodal points. The vector quantities are evaluated at the boundaries of the control volume. The last nodal point lie on the physical boundary at a wall (for example E-point in figure 3.1). This boundary point can be seen as the centre of control volume with half size. The same applies for the other boundaries. In this way, two extra nodal points are placed at the boundaries in $x$- and $y$- directions and each boundary nodal point is a centre of CV cell with half size.

When the control volume method is used, equations are solved in integral form, rather than in differential form. As an example, let us consider the particle balance (3.5)

$$\frac{\partial n_p}{\partial t} + \nabla \cdot \Gamma_p = S_p.$$  

Integrating over a control volume $V$ with boundary surface $A$ and applying Gauß divergence theorem yields

$$\frac{\partial}{\partial t} \int_V n_p d^3V + \int_A \Gamma_p \cdot e_n d^2A = \int_V S_p d^3V,$$  

(3.28)
where $e_n$ is the unit normal vector to the surface. This equation can be written \[20\]:

$$V \frac{\partial}{\partial t} n_p + A_e \Gamma_e - A_w \Gamma_w + A_n \Gamma_n - A_s \Gamma_s = V S_p.$$ (3.29)

The remaining issue is how to express the time-derivative and flux density in terms of the present and future values of the densities $n_p$ in the grid points. This question will be answered later in this section; first we will look how an MD2D simulation proceeds.

### 3.4.2 Simulation overview

The variables involved in the model are the electrostatic potential $V$, the densities $n_p$ of the active species and the electron energy density $n_\varepsilon$. Time-integration is done using a Gummel scheme, in which the variables are solved consecutively; this procedure can be summarised as follows:

1. Set $t = t_0$ and choose initial values for $n_p(x, t_0)$ and $n_\varepsilon(x, t_0)$;
2. Determine the time step $\Delta t$;
3. Calculate the transport coefficients and reaction rate coefficients;
4. Update the electrode potentials and calculate the electrostatic potential $V(x, t + \Delta t)$ and electric field $E(x, t + \Delta t)$;
5. Calculate the species densities $n_p(x, t + \Delta t)$;
6. Calculate the electron energy source term and calculate $n_\varepsilon(x, t + \Delta t)$;
7. If $t$ is smaller than the requested simulation time, set $t \leftarrow t + \Delta t$ and continue with step 2. Otherwise, the simulation is finished.

We will now discuss the discretisation of the equations that are used in the steps 4, 5 and 6.

### 3.4.3 The electrostatic potential

Let us consider the calculation of the electrostatic potential and field. The field $E$ is governed by Poisson equation (3.26):

$$\nabla \cdot \epsilon E = \rho,$$

where the charge density is given by

$$\rho = \sum_p q_p n_p.$$ (3.30)

Let us use superscripts $k$ and $k + 1$ to denote values at times $t^k$ and $t^{k+1} = t^k + \Delta t$, respectively. The field $E^{k+1}$ at the target time depends on the electrode potentials and
charge density at time \( t^{k+1} \). Since \( \rho^{k+1} \) depends on the species densities \( n_p^{k+1} \), which have not yet been calculated when the Poisson equation is solved, one may choose to use \( \rho^k \) instead. However, this limits the time step \( \Delta t \) to the Maxwellian relaxation time, which can be prohibitively small in plasmas with not too low charged species densities [21]. In order to overcome this limitation, Ventzek et al [21] suggest to use an estimate for the charge density \( \rho^{k+1} \) instead. This estimate is based on the current continuity equation,

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot J = 0, \quad (3.31)
\]

which allows us to write

\[
\nabla \cdot \epsilon E^{k+1} = \rho^{k+1} = \rho^k + \Delta t \frac{\partial \rho}{\partial t} = \rho^k - \Delta t \nabla \cdot J.
\]

The current density is given by \( J = \sum_p q_p \Gamma_p \), from equation (3.11) we see that

\[
J^k = \sum_p q_p \Gamma_p(n^k, \mu^k, D^k, E^k).
\]

If we replace \( E^k \) with \( E^{k+1} \) in this expression and linearise the result, a term which is linear in \( E^{k+1} \) arises, which can be moved to the left hand side of the Poisson equation, where it occurs as an implicit contribution to the permittivity. On the right hand side, an implicit charge density is generated. So that the equations becomes:

\[
\nabla \cdot (\epsilon E^{k+1}) = \rho^k - \Delta t \nabla \cdot \Gamma_p(n^k, \mu^k, D^k, E^{k+1}).
\]

Both implicit terms vanish for \( \Delta t = 0 \), as well as under steady-state conditions. Details of this technique, which allows much larger time steps, can be found in [21, 22, 23].

In MD2D, the Poisson equation is expressed in terms of the electrostatic potential, which is related to the electric field via \( E = -\nabla V \). The resulting equation for \( V \) is solved in the integral form, which is again obtained by applying Gauß theorem to each control volume. The electric field is discretized using the central difference scheme, taking into account the surface charges that might exist on material interfaces. On electrode surfaces, the potential is prescribed as a function of time (Dirichlet conditions), on other boundary points \( \nabla V \cdot e_n = 0 \) is assumed (homogeneous Neumann conditions), where \( e_n \) is the unit normal vector to the surface. The resulting set of equations is solved for \( V \) with a modified version [1, section 3.7] of the MSI linear equation solver [24]. For details concerning the discretisation procedure we refer to Ref. [1].

### 3.4.4 Discretisation of the transport equation

Let us first consider the evolution of the particle and electron energy densities. If we approximate the time derivative as \( \partial n_p / \partial t \approx (n^{k+1}_p - n^k_p) / \Delta t \), the continuity equation (3.2) for the particles and for the electron energy (3.21) becomes

\[
\frac{n^{k+1}_p - n^k_p}{\Delta t} + \nabla \cdot \Gamma(n^{m_w}_p, \mu^{m_e}_p, D^{m_D}_p, E^{k+1}) = S^{m_p}_p. \quad (3.32)
\]
Note that, in view of the order of calculation, which has been discussed in section 3.4.2, the flux is always calculated in terms of the electric field \(E^{k+1}\) at the target time \(t^{k+1}\). The indices \(m\) can be either \(k\) or \(k+1\). For \(m = k\), an explicit equation for the densities at the target time step \(t^{k+1}\) is obtained. While easy to implement, such explicit scheme has the disadvantage that the time step is severely limited by the Courant-Friedrichs-Lewy criterium \([25, 26]\). For this reason, in MD2D a semi-implicit scheme that does not have this restriction has been adopted. In this scheme we choose \(m_n = k + 1\). For the other indices we choose \(m = k\), with one exception: for the electron energy density equation we choose \(m = k + 1\); this implicit source term for the electron energy density will be discussed in section 3.4.5.

The application of the control volume method requires that the perpendicular components of the flux densities are discretized on the control volume boundary surfaces. As an example, consider the flux density \(\Gamma_e\) in the point \(e\) in figure 3.1. In MD2D, Scharfetter-Gummel’s \([27]\) exponential scheme is used, in which case the flux \(\Gamma_e\) is expressed in terms as the neighbouring nodal density values \(n_C\) and \(n_E\) as

\[
\Gamma_e = \frac{1}{\Delta x} D_e [f_2(z_e)n_C - f_1(z_e)n_E],
\]

where the functions \(f_1(z)\) and \(f_2(z)\) are defined by

\[
\begin{align*}
    f_1(z) & = z/(\exp(z) - 1), \\
    f_2(z) & = f_1 + z,
\end{align*}
\]

and the grid Péclet number \(z\) is given by

\[
z_e = \frac{\mu_e E_e}{D_e} \Delta x.
\]

Similar expressions can be derived for the (outward) flux in the points \(w, n,\) and \(s\). These can then be substituted in the integral form (see equation (3.29)) of equation (3.32). The result is an algebraic equation that couples the values of \(n_p\) in the point \(C\) to the values in the neighbouring points. When supplemented with boundary conditions, the set of equations for all nodal points can be solved for the field \(n_p\). In MD2D the latter is done with a modified version of MSI, see the references at the end of section 3.4.3. For a thorough discussion of the boundary conditions for the species densities and electron energy density, we refer to Ref. [1].

### 3.4.5 The electron energy source term

In the previous section we have mentioned that the solution of the electron energy density equation is similar to that of the species densities equations. However, an explicit treatment of the electron energy source term tends to produce instabilities because it depends strongly on the mean energy, mainly due to inelastic energy losses. To avoid instabilities at larger time steps, the electron energy source term is evaluated semi-implicitly, rather
than explicitly. This way, larger time steps are possible than is the case with an explicit formulation which is based only on the old values of the densities, transport coefficients and fields. The procedure is explained in detail in Ref. [1], we will briefly discuss the implicit evaluation and its improvement.

The derivation starts with the expression for the electron energy source, which was first presented in equation (3.20),

$$S_\varepsilon = \Gamma_e \cdot E - n_e \sum_r \varepsilon_r k_r n_r - L_\varepsilon.$$  

The discussion of the linearisation of this source term in Ref. [1] is based on the assumption that the electrons satisfy the Einstein relation (see section 3.3). In the present text we will develop a result that is also valid in the case that the Einstein relation does not apply. To this end we consider the Ohmic heating term in the previous equation,

$$S_{\varepsilon,em} = \Gamma_e \cdot E.$$  

In the nodal point of a control volume this is made up by four contributions from east, west, south and north. The source term is made semi-implicit by first linearising each contribution in terms of the mean electron energy $\bar{\varepsilon}$,

$$S_{\varepsilon,em}^{k+1} = S_{\varepsilon,em}^k + \left. \frac{\partial S_{\varepsilon,em}}{\partial \varepsilon} \right|_k (\varepsilon^{k+1} - \varepsilon^k).  \quad (3.37)$$

Let us look at the contribution $\Gamma_{e,e} E_e$ from the eastern boundary. Since the electric field is not $\varepsilon$-dependent, we have

$$\frac{\partial \Gamma_{e,e} E_e}{\partial \varepsilon} = E_e \frac{\partial \Gamma_{e,e}}{\partial \varepsilon}.$$  

So, in order to linearise the energy source term we must find the derivative of the flux components with respect to the electron energy. The electron flux density depends on the electron density, mobility and diffusion coefficient and on the electric field. Since only the electron mobility and diffusion coefficient are functions of the electron energy, we get

$$\frac{\partial \Gamma_{e,e}}{\partial \varepsilon} = \frac{\partial \Gamma_{e,e}}{\partial \mu_{e,e}} \frac{d \mu_{e,e}}{d \varepsilon} + \frac{\partial \Gamma_{e,e}}{\partial D_{e,e}} \frac{d D_{e,e}}{d \varepsilon}.$$  

Using the flux expression from equation (3.33), a little amount of algebra yields

$$\frac{\partial \Gamma_{e,e}}{\partial \varepsilon} = -\frac{1}{\Delta x} h(z) (n_{e,E} - n_{e,C}) \mu_{e,e} \frac{d}{d \varepsilon} \left( \frac{D_{e,e}}{\mu_{e,e}} \right) + \frac{\Gamma_{e,e}}{\mu_{e,e}} \frac{d \mu_{e,e}}{d \varepsilon},  \quad (3.38)$$

where the function $h(z)$ is defined, using equation (3.34) and (3.35), as:

$$h(z) = f_1(z) - \frac{\partial f_1(z)}{\partial z} z = f_2(z) - \frac{\partial f_2(z)}{\partial z} z = \frac{z^2 \exp(z)}{(\exp(z) - 1)^2}.  \quad (3.39)$$
The expression (3.38), and similar expressions for the electron flux derivative with respect to energy at the other sides can be substituted back into the equation for the source derivative (equation (3.37)) in order to obtain the implicit source term. For details we refer to Ref. [1].

In the cited work, the expression (3.38) cannot be found. Rather, the result of a more restricted derivation which assumes the Einstein relation is presented. Given the present formulation, and assuming that the Einstein relation is valid, so \( D_e/\mu_e = -2\varepsilon/3e \), we get

\[
\frac{\partial \Gamma_{e,e}}{\partial \varepsilon} = +\frac{2\mu_{e,e}}{3e} \frac{1}{\Delta x} h(z) (n_{e,E} - n_{e,C}) + \frac{\Gamma_{e,e}}{\mu_{e,e}} \frac{d\mu_{e,e}}{d\varepsilon},
\]

(3.40)

which indeed corresponds to equation 3.33 in Ref. [1].

### 3.4.6 Wall interaction

At the open boundaries (symmetry axis for example) homogeneous Neumann boundary conditions are employed, meaning that the derivatives of the quantities in the directions perpendicular to these boundaries are set to zero: for all the active species \( \nabla n_p \cdot e_n = 0 \), where \( e_n \) is the unit normal vector to the surface; \( \nabla n_e \cdot e_n = 0 \) for the electron energy density. For the physical boundaries the boundary conditions for \( n_p \) and \( n_e \) are given by expressions for the flux densities \( \Gamma_p \) and \( \Gamma_e \). For the detailed description of the boundary conditions we refer to [1].

The difference from the cited work, lies in the boundary conditions for the neutral particles. This has been modified such that sputtering process at the metallic boundaries is possible. The process of sputtering can be treated as a probability process in which every time when an energetic particle impinges the cathode, ejection of atoms can occur with a certain probability. The rate of sputtering is usually expressed in terms of the sputtering yield \( \xi_p \), giving the mean number of atoms removed per incident particle.

The source \( S_{sp} \) of sputtered metal atoms at the metallic boundaries is given by:

\[
S_{sp} = \sum_p \xi_p \Gamma_p \cdot e_n,
\]

(3.41)

that is the sum of the product of the flux density of the bombarding energetic particles \( \Gamma_p \) and the corresponding sputtering yield \( \xi_p \). Here \( p \) runs over the collection of energetic particles.

The boundary conditions for the sputtered species is given by [1, eqn. 2.28]:

\[
\Gamma_{sp} \cdot e_n = \frac{1 - r_{sp}}{1 + r_{sp}} \frac{1}{2} v_{th,sp} n_{sp} - \frac{2}{1 + r_{sp}} S_{sp},
\]

(3.42)

where \( r_{sp} \) is the reflection coefficient, \( v_{th,sp} \) is the thermal velocity.

\(^1\)Apart from a minus sign, which is due to the fact that in the cited work the absolute value of \( \mu_e \) is used throughout.
In the present work we opted to calculate the sputtering yield using the Yamamura’s empirical formula [28]. The formula gives the sputtering yields of pure materials as a function of the bombarding energy and could predict the energy dependent yields for any ion-target combination.

References


Towards a reduced chemistry module of a He-Ar-Cu hollow cathode discharge

Abstract. This study is aimed to find a reduced chemistry module for hollow cathode discharge excited in a He-Ar-Cu mixture. This enables to construct lean and reliable models that can be used as a part of the design tool of hollow cathode discharges (HCDs). To this end estimative calculations and numerical simulations are performed at optimal conditions for lasing. An analysis of the species behaviour and reactions is made and as a result the model is simplified by means of reducing the number of species and reactions. The consequences of these reductions is justified by comparing the results of the simplified models with those of a more complete one. This study delivers a model that is chemical lean and thus, much less time consuming. It can be used in optimization studies to find the optimum in the plasma control parameter-set of HCDs.
4.1 Introduction

The interest in complex chemistry plasmas is increasing rapidly during the last decade. This is among others boosted by the booming attention paid to plasmas for biomedical applications and environmental technology. In all these modern applications the classical atomic plasma composed of one single noble gas species is replaced by plasmas created in rich chemical fillings. As a consequence of plasma activities these fillings are transformed into a myriad of excited atoms, molecules, radicals and various types of ions. This gives firm ground to the statement that the problem of plasma physics nowadays mostly lies in plasma chemistry.

One of the serious problems is manifest by the increase in the number $N$ of relevant species. For a complex plasma this can easily go beyond $N = 100$. Even more seriously is the number of conversion reactions between these species for which we can expect a scaling with something like $N^2$. A further complicating factor is the large variety in reaction speeds. Here we can easily find differences in the order of $10^3$. This makes that complex chemistry plasma models can become extremely stiff.

In search for the most appropriate strategy to model these plasmas the question rises whether it is really needed to treat all the species individually. Should we compute for all the species their densities and transport fluxes? Or, do there exist systematic methods that can lead to a reduced chemistry? A reduced chemistry module is a module that is small enough to make the grand plasma model lean while the computation of the main plasma properties is not in jeopardy.

This study is devoted to the search of a reduced chemistry module for hollow cathode discharges (HCDs) used for laser applications. Such a module enables to construct a lean and reliable model that can be used as part of a design tool of HCD lasers. The aim of this tool is to find, by modelling, the optimum in the plasma control parameters-set of these plasmas. Since the control parameter space is enormous it is requested that the model converges easily so that the exploration of this hyper-space is not too time consuming. With this chemical lean model it is then possible to find the best version of the geometrical construction, the anode cathode patterns, the current settings, the fill-chemistry, etc.

This study starts with a model that describes a longitudinal sputtering HCD used for laser application. It has a cylindrical geometry with two anodes at both sides as shown in figure 2.3 in chapter 2. The sputtered cathode material is Cu and the operating gas is a mixture of He with 5% of Ar. The total gas pressure equals 2.3 kPa and the discharge is excited by a 3 ms current pulse.

The model used for this study is the time-dependent fluid model MD2D described in detail in chapter 3. The simulation model is based on a set of 10 species and 23 reactions. For these active species the balance equations are solved each of them describing the competition between the transport and chemistry on a per-species base. The results of the simulation includes the spatial and temporal evolution of densities, the reaction rates and transport fluxes of the relevant species. These are used to get volume-averaged values of these quantities. From this output a detailed analysis of the contribution of production/ destruction and transport for each species can be obtained so that the most important species
and the relevant conversion channels can be determined. The insight obtained can subsequently be used to construct a grand model based on a reduced chemistry. To justify the possible consequences of the chemistry reduction we will compare the results of the simplified models with those of the initial more complex chemistry model.

The paper is organised as follows: in section 4.2 the hierarchy in processes is explored and estimative calculations are made. Section 4.3 describes the set of species and reactions used in the numerical model. A detailed analysis is made and presented in section 4.4. Based on this analysis simplified models are constructed and the comparison with the more complete model is presented in subsections. Finally, section 4.5 summarises the observations.

4.2 Hierarchy in processes

The analysis of the hierarchy of chemical species and conversion processes will be guided by the general form of the particle balance showing the competition between chemistry (production and destruction processes) and transport. For the particle $p$ it reads:

$$\frac{\partial n_p}{\partial t} + \nabla \cdot \Gamma_p = S_p,$$  \hspace{1cm} (4.1)

where $\Gamma_p$ is the flux density and $S_p$ is net source term. The time-derivative can be omitted since the duration of the excitation current pulse, being 3 ms, greatly exceeds the time-scales of the plasma processes. This implies that steady state (SS) is obtained during the pulse. Omitting the transient term, the continuity equation reads:

$$\nabla \cdot \Gamma_p = S_p.$$ \hspace{1cm} (4.2)

We start with an exploration of the left hand side, the transport side, of this equation. After that the right hand side will be dealt with.

By writing the transport term as $\nabla \cdot \Gamma_p = n_p \nu_p^t$, we introduce the transport frequency $\nu_p^t$ of species $p$. The flux density $\Gamma_p$ is given by:

$$\Gamma_p = \mu_p E n_p - D_p \nabla n_p,$$ \hspace{1cm} (4.3)

where the mobility $\mu_p$ and diffusion coefficient $D_p$ of species $p$ are determined by collisions of these species with buffer gas atoms. We see that in general two driving forces are present: the drift generated by the electric field and the diffusion set up by gradients. It is clear that neutral particles are driven by the diffusion only. Due to the additional drift term, the ions will have higher velocities in general. We will come back to this later.

The right hand side of equation (4.2), the source term $S_p$ is determined by the reactions occurring in the discharge:

$$S_p = \sum c_{p,r} R_r,$$ \hspace{1cm} (4.4)
where \( c_{p,r} \) is the net stoichiometric number of particles of species \( p \) created in one reaction \( r \) while \( R_r \) is the corresponding reaction rate. The sum runs over all the reactions. The source term \( S_p \) consists of positive \( c_{p,r} > 0 \) (production) and negative \( c_{p,r} < 0 \) (loss) contributions. So that it can be split in two terms — the production \( \mathcal{P}_p \) and loss rate \( \mathcal{L}_p \):

\[
S_p = \mathcal{P}_p - \mathcal{L}_p. \tag{4.5}
\]

Note that \( \mathcal{P}_p \) is the total production rate: it is a sum over all the reactions in which \( p \) is created. The same applies for the loss rate \( \mathcal{L}_p \); it is the total loss due to radiative and collision-induced transitions. The production rate can be written as:

\[
\mathcal{P}_p = \sum_r n_q \mathcal{D}_{q,p}, \tag{4.6}
\]

where \( \mathcal{D}_{q,p} \) is the frequency of the destruction processes of particle \( q \) that lead to the production of \( p \). The sum runs over all the reactions that lead to the production of \( p \). The destruction of the same level \( p \) is:

\[
\mathcal{L}_p = n_p \mathcal{D}_p, \tag{4.7}
\]

where \( \mathcal{D}_p = \sum_r \mathcal{D}_{p,r} \) is the destruction frequency of particle \( p \), the so-called destruction factor of \( p \).

Thus, we can write the general form of the particle balance as:

\[
n_p \nu_{p,t} = \mathcal{P}_p - n_p \mathcal{D}_p
\]

\[
n_p \frac{\nu_{p,t}}{\mathcal{D}_p} = \frac{\mathcal{P}_p}{\mathcal{D}_p} - n_p. \tag{4.9}
\]

In the case that the transport frequency \( \nu_{p,t} \) is much smaller than the destruction frequency, i.e. \( \frac{\nu_{p,t}}{\mathcal{D}_p} \ll 1 \), equation (4.9) becomes:

\[
n_p = \frac{\mathcal{P}_p}{\mathcal{D}_p}. \tag{4.10}
\]

So that the density of \( p \) is completely determined by local reactions. The species will be designated by local chemistry (LC) species. Combining equations (4.10) and (4.7) leads to the criteria that holds for LC species:

\[
\frac{\mathcal{P}_p}{\mathcal{L}_p} = 1. \tag{4.11}
\]

The density of these LC species is generally much lower than that of the ground state atoms and ions. The latter, in general large density reservoirs, are apart from chemistry, also ruled by transport. These species are denoted by transport sensitive (TS) species.
Reduced chemistry module

buffer gas atoms in ground state are excluded, since they are assumed to be stationary in space and time.

The above shows that the dimensionless quantity $\nu_t^l/D_p$ can be employed as a tool to distinguish between LC and TS species. For the last category the competition between transport and chemistry is important meaning that we have to solve the corresponding transport equations while for the first that is not needed and the continuity equation can be simplified. This ratio can be determined by running the grand model and to find $\nu_t^l/D_p$ (or $\mathcal{D}_p/\mathcal{L}_p$) by post processing. Before doing so, we will perform some estimative calculations. We distinguish the following species:

- The buffer gas particles, the ground state atoms of He and Ar are assumed to be constant in time and space so they will not be equipped with a particle balance.

- For excited neutral species we can assume that the transport is purely diffusive and given by $\nu_t^l = D_p/R^*^2$, where $R^*$ is in the order of the radius of the discharge. For the diffusion coefficient $D_p$ we use the expression $D_p = k_B T_g/(M_p \nu_m)$ with $\nu_m$ frequency of momentum transfer from the species to the buffer gas atoms, in this case He. We can use $\nu_m = n_{\text{He}} \langle v \rangle \sigma$, where $\langle v \rangle = (k_B T_g/M_{\text{He}})^{1/2}$ is the thermal velocity of the He atoms; this is justified since the relative velocity is mostly determined by thermal velocity of the lightest particle (He). That means that the diffusion coefficient reads:

$$D_p = \frac{(k_B T_g)^{3/2} \sqrt{M_{\text{He}}}}{M_p P \sigma m_p} = \frac{0.4}{M_p} m^2 s^{-1}, \quad (4.12)$$

where $M_p^*$ is the mass number of the atom, $m_p$ the proton mass and $\sigma$ the cross section for momentum transfer. Inserting $T_g = 1000 K$, $P = 2.3 kPa$ and $\sigma = 10^{-19} m^2$, we get the value given at the right hand side. Of course the $\sigma$-value depends on the couple of interacting species but to get insight in the order of magnitude we took $\sigma = 10^{-19} m^2$. Inserting for $R = 2 mm$ we find for the Cu and Ar atoms values in the order of $\nu_t^l = 2.10^3 s^{-1}$. For excited He atoms this is more or less 40 times larger. For the excited atomic species we can use for the destruction frequency $\mathcal{D}_p = 10^7 s^{-1}$, which is taken equal to a typical value of the radiative decay frequency. So for excited species we get $\nu_t^l/\mathcal{D}_p$ values in the order of $10^{-3}$ to $10^{-2}$, meaning that they can be considered as being determined by local chemistry.

- For the excited ions we only have to consider $\text{Cu}^{+*}$, which will represent the upper laser level. Apart from diffusion, this species will also be transported by drift. To get an estimate of the combination of drift and diffusion we can assume that the net effect leads to ambipolar diffusion. This implies a diffusion enhancement of the form $D_a = D_p(1 + T_e/T_h)$, meaning that the transport frequency will be about 50 times higher. Inserting the radiative decay frequency in order of $10^8 s^{-1}$ we find that $\nu_t^l/\mathcal{D}_p$ is in the order of $10^{-3}$. So that also the $\text{Cu}^{+*}$ species are determined by local chemistry.
For the atomic and molecular ions we have to compare the ambipolar diffusion with recombination processes. Several recombination channels have to be considered, which can be classified in 2-particle or 3-particle recombination channels. Due to the (global) charge neutrality the destruction of ions is closely related to that of electrons and as the plasma is strongly ionising we can expect that electrons are TS. So the same must apply to the collection of ions. However, to understand the role of the various different ions we have to consult the model results in order to determine the LC ions.

Before to run the model some considerations can be made on the transition frequencies and the nature of the underlying processes. For the transition frequency related to the conversion of species \( p \) into \( q \) the transition frequency \( D_{p,q} \) was introduced. In the case of radiative decay \( D_{p,l} = A_{p,l} \), being the probability of the radiative decay of \( p \) to a lower level \( l \). In the case of that 2 particles are involved we have \( D_{p,q} = n_q k_{p,q}^{(2)} \) where \( n_q \) is the density of the particles colliding with \( p \). For 3-particles interaction we have \( D = n_x n_y k_{p,q}^{(3)} \).

A further classification can be made on the role of the excitation agent, the particle of type \( x \) and \( y \) as given above the expressions of the frequencies. We start with the electrons. These are found to be of main importance, especially there, where the active zone plasma is created. These endothermic processes, including excitation and ionization, are denoted by electron excitation kinetics (EEK). Once the radicals (the excited particles, ions) are formed, they can interchange there internal energy by means of charge or excitation transfer. The collection of these processes is denoted by heavy particle excitation kinetics (HEK). The excess of internal energy will be used to heat the gas. If the internal energy is transferred to the electrons we talk about MEK: mixed excitation kinetics. An example of a MEK process is Penning ionization. Finally we have the exothermal processes, the collection of recombination and deexcitation processes.

### 4.3 Species and reactions included in the model

The buffer gas atoms are uniformly distributed in time and space, i.e. the gas temperature, the gas pressure and density are independent on the spatial position. The gas temperature is assumed to be 1000 K.

Table 4.1 lists the set of species for which the balance equations (equations (4.1) and (4.3)) were solved, together with the transport coefficients used in the model and the corresponding references.

The Cu\(^{+}\) in table 4.1 represents the excited ionic upper laser level, it is situated 24.5 eV above the ground state of atomic Cu.

The molecular ions are included as their influence could be of importance at the conditions under study. The intermediate states, the excited molecules, are not included in the model.

We assume that for the charged particles, the Einstein relation can be used to link the
Reduced chemistry module

<table>
<thead>
<tr>
<th>Species</th>
<th>$\mu_p \times p$ (Torr m$^2$V$^{-1}$s$^{-1}$)</th>
<th>$D_p \times p$ (Torr m$^2$V$^2$)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>e</td>
<td>$f_1(\varepsilon)$</td>
<td>equation (4.13)</td>
<td>[1]</td>
</tr>
<tr>
<td>Ar$^+$</td>
<td>$f_2(E/N)$</td>
<td>equation (4.13)</td>
<td>[2]</td>
</tr>
<tr>
<td>He$^+$</td>
<td>$f_3(E/N)$</td>
<td>equation (4.13)</td>
<td>[2]</td>
</tr>
<tr>
<td>Cu$^+$</td>
<td>$f_4(E/N)$</td>
<td>equation (4.13)</td>
<td>[3]</td>
</tr>
<tr>
<td>Cu$^{+*}$</td>
<td>$f_5(E/N)$</td>
<td>equation (4.13)</td>
<td>[3]</td>
</tr>
<tr>
<td>Ar$_2^+$</td>
<td>$f_6(E/N)$</td>
<td>equation (4.13)</td>
<td>[2]</td>
</tr>
<tr>
<td>He$_2^+$</td>
<td>$f_7(E/N)$</td>
<td>equation (4.13)</td>
<td>[2]</td>
</tr>
<tr>
<td>Ar$^*$</td>
<td>-</td>
<td>20</td>
<td>[5]</td>
</tr>
<tr>
<td>He$^*$</td>
<td>-</td>
<td>15.35</td>
<td>[5]</td>
</tr>
</tbody>
</table>

Table 4.1: The list of species for which the balance equation is solved, including the used transport coefficients and the corresponding references. The mobility coefficient for the electrons is specified as a function of the mean electron energy $f(\varepsilon)$. For the ionic species it is given as a function of the reduced electric field $f(E/N)$. The diffusion coefficients are deduced from the mobility using the Einstein relation (4.13).

diffusion coefficients to the mobility:

$$D_p = \frac{k_B T_p \mu_p}{q_p},$$  \hspace{1cm} (4.13)

where $q_p$ is the elementary charge, and $T_p$ the particle temperature, corresponding to the energy of the random particle motion.

For the ions the local field approximation is used. This assumes a direct relation between the particle energy distribution and the electric field, hence the transport coefficients are regarded as being functions of the reduced electric field. These functions can be found in literature as a result of experiments and classical theories [2, 3]. The mobility of He$^+$ and Ar$^+$ ions in their pure parent gases as a function of the reduced electric field are adopted from [2]. From this the mobilities in a He-Ar gas mixture are calculated using Blanc’s law (see equation (3.17)).

For electrons, however, substantial deviations from the local field approximation can be expected for typical HCD conditions. Due to the small mass as compared to that of the heavy particles, electrons created in a high $\mathbf{E}$-field region can easily be launched to regions where the $\mathbf{E}$-field is small while keeping (part of) the initially gained high energy. Therefore, rather than using a dependence of the electron properties on $(E/N)$, we have to solve the electron energy balance. This gives the mean electron energy $\varepsilon(x)$ as a function of position. Subsequently the electron transport coefficients can be computed using the local $f(\varepsilon)$ values. The data are obtained from the free-ware Boltzmann equation solver BOLSIG+ [1].

Since the individual metastable helium states participate in similar reactions with very similar rate coefficients [6], they are combined into one effective level, lying at 20.215 eV
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Table 4.2: Reaction rate coefficients used in the model. The electron impact reactions are specified as a function of the mean electron energy \( k_r(\varepsilon) \).

<table>
<thead>
<tr>
<th>Nr.</th>
<th>Reaction</th>
<th>Rate coefficient, ( k_r )</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>( e + Ar \rightarrow Ar^* + e )</td>
<td>( k_1(\varepsilon) ) m(^3) s(^{-1})</td>
<td>[1]</td>
</tr>
<tr>
<td>2</td>
<td>( e + Ar^* \rightarrow Ar^* + 2e )</td>
<td>( k_2(\varepsilon) ) m(^3) s(^{-1})</td>
<td>[1]</td>
</tr>
<tr>
<td>3</td>
<td>( e + Ar \rightarrow Ar^+ + 2e )</td>
<td>( k_3(\varepsilon) ) m(^3) s(^{-1})</td>
<td>[1]</td>
</tr>
<tr>
<td>4</td>
<td>( e + Cu \rightarrow Cu^+ + 2e )</td>
<td>( k_4(\varepsilon) ) m(^3) s(^{-1})</td>
<td>[1]</td>
</tr>
<tr>
<td>5</td>
<td>( e + He \rightarrow He^* + e )</td>
<td>( k_5(\varepsilon) ) m(^3) s(^{-1})</td>
<td>[1]</td>
</tr>
<tr>
<td>6</td>
<td>( e + He^* \rightarrow He^* + 2e )</td>
<td>( k_6(\varepsilon) ) m(^3) s(^{-1})</td>
<td>[1]</td>
</tr>
<tr>
<td>7</td>
<td>( e + He \rightarrow He^* + 2e )</td>
<td>( k_7(\varepsilon) ) m(^3) s(^{-1})</td>
<td>[1]</td>
</tr>
<tr>
<td>8</td>
<td>( Ar^* + Cu \rightarrow Ar + Cu^+ )</td>
<td>( 3.16 \times 10^{-16} ) m(^3) s(^{-1})</td>
<td>[7]</td>
</tr>
<tr>
<td>9</td>
<td>( He^* + Cu \rightarrow He + Cu^{++*} )</td>
<td>( 1.0 \times 10^{-15} ) m(^3) s(^{-1})</td>
<td>[1]</td>
</tr>
<tr>
<td>10</td>
<td>( Ar^* + 2Ar \rightarrow Ar^+_2 + Ar )</td>
<td>( 9.66 \times 10^{-6} ) m(^3) s(^{-1})</td>
<td>[8]</td>
</tr>
<tr>
<td>11</td>
<td>( He^* + 2He \rightarrow He^+_2 + He )</td>
<td>( 6.8 \times 10^{-6} ) m(^3) s(^{-1})</td>
<td>[9]</td>
</tr>
<tr>
<td>12</td>
<td>( Ar^* + Ar^* \rightarrow Ar^* + Ar + e )</td>
<td>( 1.17 \times 10^{-15} ) m(^3) s(^{-1})</td>
<td>[8]</td>
</tr>
<tr>
<td>13</td>
<td>( Ar^* + Ar^* \rightarrow Ar^+_2 + e )</td>
<td>( 2.03 \times 10^{-15} ) m(^3) s(^{-1})</td>
<td>[8]</td>
</tr>
<tr>
<td>14</td>
<td>( Ar^* + Cu \rightarrow Ar + Cu^* + e )</td>
<td>( 4.4 \times 10^{-16} ) m(^3) s(^{-1})</td>
<td>[7]</td>
</tr>
<tr>
<td>15</td>
<td>( He^* + He^* \rightarrow He^* + He + e )</td>
<td>( 0.87 \times 10^{-15} ) m(^3) s(^{-1})</td>
<td>[9]</td>
</tr>
<tr>
<td>16</td>
<td>( He^* + He^* \rightarrow He^+_2 + e )</td>
<td>( 1.5 \times 10^{-15} ) m(^3) s(^{-1})</td>
<td>[9]</td>
</tr>
<tr>
<td>17</td>
<td>( He^* + Ar \rightarrow He + Ar^* + e )</td>
<td>( 1.68 \times 10^{-16} ) m(^3) s(^{-1})</td>
<td>[7]</td>
</tr>
<tr>
<td>18</td>
<td>( He^* + Cu \rightarrow He + Cu^* + e )</td>
<td>( 9.48 \times 10^{-16} ) m(^3) s(^{-1})</td>
<td>[7]</td>
</tr>
<tr>
<td>19</td>
<td>( Ar^* + 2e \rightarrow Ar + e )</td>
<td>( k(\varepsilon) ) m(^3) s(^{-1})</td>
<td>[1]</td>
</tr>
<tr>
<td>20</td>
<td>( Ar^+_2 + e \rightarrow Ar^* + Ar )</td>
<td>( k_8(\varepsilon) ) m(^3) s(^{-1})</td>
<td>[8]</td>
</tr>
<tr>
<td>21</td>
<td>( He^+_2 + e \rightarrow He^* + He )</td>
<td>( k_9(\varepsilon) ) m(^3) s(^{-1})</td>
<td>[9]</td>
</tr>
<tr>
<td>22</td>
<td>( He^* + 2e \rightarrow He + e )</td>
<td>( k_{10}(\varepsilon) ) m(^3) s(^{-1})</td>
<td>[1]</td>
</tr>
<tr>
<td>23</td>
<td>( Cu^{++} \rightarrow Cu^* + h\nu )</td>
<td>( 0.23 \times 10^{9} ) s(^{-1})</td>
<td>[10]</td>
</tr>
</tbody>
</table>

The reactions listed in Table 4.2 are used in the model: the first 7 lines give the reactions induced by EEK, the group 8 – 11 are HEK, the group 12 – 18 are MEK. The next group of reactions from 19 to 22 are the recombination processes followed by the radiative process 23. Above the ground level. The same applies to the Ar metastable levels: the metastable levels are combined into one effective level lying at 11.55 eV above the argon ground state. Other excited states are not included in the model. In section 4.4.3 we will investigate what happens to the main plasma properties when the total excitation to all the excited levels is being set equal to ionization. By doing so, we in fact involve all the excited levels as stages in the ladder-climbing ionization process. This can be seen as giving the maximum in effective ionization. Excited states of sputtered Cu atoms are not taken into account since we do not expect that the (de)excitation of these levels will affect the electron energy and particle balance substantially.
As stated before, the buffer gas atoms, He and Ar in ground state, are assumed to be uniformly distributed in the discharge whereas the ground state copper atoms are produced by sputtering only. The atoms in ground state are ionized by electron collisions: direct ionization (reactions 3, 4 and 7) and excited to the lumped levels Ar$^*$ and He$^*$ (reactions 1 and 5). These levels can be converted into ions by means of electron collisions (reactions 2 and 6) and Penning ionization (reactions 12, 14, 15, 17 and 18). The HEK includes charge transfer reactions between the buffer gas ions and metal atoms in ground state (reactions 8 and 9). The processes of molecular ion formation include associative ionization (reactions 13 and 16) from the metastable levels and ion conversion (reactions 10 and 11), while the destruction process is dissociative recombination (reactions 20 and 21). The density of the excited Cu ionic level is mainly generated by charge transfer of He ions to Cu atoms (reaction 9) and decay spontaneously to the ground ionic level of Cu (reaction 23).

4.4 Results and discussion

The calculation results include the spatial distribution of the densities and fluxes of the species, as well as the rates of their production and loss processes. These are used to get volume averaged values of these quantities.

4.4.1 Chemistry analysis

Table 4.3 presents the reaction rates of each reaction averaged over the volume. From this table the following information can be deduced.

- The buffer gas atoms (Ar and He) are ionized mainly by electron impact; this can be due to direct ionization or stepwise ionization, that is ionization intermediated by excitation. From the table it can be seen that the rates of excitation and ionization of Ar are much larger than those of He. This is because the ionization (excitation) potential of Ar is lower (24.58 versus 15.76 eV for ionization and 20.215 versus 11.62 eV for excitation). This implies that the Ar ion is the main ionic species in the discharge in spite of the fact that Ar consists of only 5% of the gas mixture. Comparing the electron excitation kinetics EEK with HEK shows that the electron impact processes have the highest rates. This makes that the system can be seen as an ionising EEK plasma.

- The rates of the backward processes, atomic recombination (reactions 19 and 21), are in the order of $10^{16} \text{ m}^{-3} \text{ s}^{-1}$ for He$^+$ and Ar$^+$. Since they are 8 orders of magnitude lower than the forward process, it is obvious that they can be neglected.

- The excited ion level Cu$^{++}$ is only produced by charge transfer with He atoms (reaction 9) and decay spontaneously to Cu$^+$ (reaction 23). It can be seen from table 4.3 that the rates of these two reactions are the same, meaning that the criteria (4.11) holds for Cu$^{++}$. This confirms what was found before: Cu$^{++}$ is a LC species.
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<table>
<thead>
<tr>
<th>Nr.</th>
<th>Reaction</th>
<th>Rate ( \text{m}^{-3} \text{s}^{-1} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>( \text{e} + \text{Ar} \rightarrow \text{Ar}^* + \text{e} )</td>
<td>( 7.752 \times 10^{24} )</td>
</tr>
<tr>
<td>2</td>
<td>( \text{e} + \text{Ar}^* \rightarrow \text{Ar}^+ + 2\text{e} )</td>
<td>( 6.769 \times 10^{24} )</td>
</tr>
<tr>
<td>3</td>
<td>( \text{e} + \text{Ar} \rightarrow \text{Ar}^+ + 2\text{e} )</td>
<td>( 2.159 \times 10^{24} )</td>
</tr>
<tr>
<td>4</td>
<td>( \text{e} + \text{Cu} \rightarrow \text{Cu}^+ + 2\text{e} )</td>
<td>( 1.421 \times 10^{24} )</td>
</tr>
<tr>
<td>5</td>
<td>( \text{e} + \text{He} \rightarrow \text{He}^* + \text{e} )</td>
<td>( 4.465 \times 10^{23} )</td>
</tr>
<tr>
<td>6</td>
<td>( \text{e} + \text{He}^* \rightarrow \text{He}^+ + 2\text{e} )</td>
<td>( 1.73 \times 10^{23} )</td>
</tr>
<tr>
<td>7</td>
<td>( \text{e} + \text{He} \rightarrow \text{He}^+ + 2\text{e} )</td>
<td>( 3.708 \times 10^{22} )</td>
</tr>
<tr>
<td>8</td>
<td>( \text{Ar}^+ + \text{Cu} \rightarrow \text{Ar} + \text{Cu}^+ )</td>
<td>( 4.864 \times 10^{22} )</td>
</tr>
<tr>
<td>9</td>
<td>( \text{He}^+ + \text{Cu} \rightarrow \text{He} + \text{Cu}^{++} )</td>
<td>( 3.439 \times 10^{21} )</td>
</tr>
<tr>
<td>10</td>
<td>( \text{Ar}^+ + 2\text{Ar} \rightarrow \text{Ar}_2^+ + \text{Ar} )</td>
<td>( 4.719 \times 10^{19} )</td>
</tr>
<tr>
<td>11</td>
<td>( \text{He}^+ + 2\text{He} \rightarrow \text{He}_2^+ + \text{He} )</td>
<td>( 3.348 \times 10^{20} )</td>
</tr>
<tr>
<td>12</td>
<td>( \text{Ar}^* + \text{Ar}^* \rightarrow \text{Ar}^* + \text{e} + \text{Ar} )</td>
<td>( 1.298 \times 10^{23} )</td>
</tr>
<tr>
<td>13</td>
<td>( \text{Ar}^* + \text{Ar}^* \rightarrow \text{Ar}_2^* + \text{e} )</td>
<td>( 2.197 \times 10^{23} )</td>
</tr>
<tr>
<td>14</td>
<td>( \text{Ar}^* + \text{Cu} \rightarrow \text{e} + \text{Ar} + \text{Cu}^+ )</td>
<td>( 1.325 \times 10^{23} )</td>
</tr>
<tr>
<td>15</td>
<td>( \text{He}^* + \text{He}^* \rightarrow \text{e} + \text{He} + \text{He}^* )</td>
<td>( 6.731 \times 10^{19} )</td>
</tr>
<tr>
<td>16</td>
<td>( \text{He}^* + \text{He}^* \rightarrow \text{He}_2^+ + \text{e} )</td>
<td>( 1.161 \times 10^{20} )</td>
</tr>
<tr>
<td>17</td>
<td>( \text{He}^* + \text{Ar} \rightarrow \text{He} + \text{Ar}^+ + \text{e} )</td>
<td>( 2.61 \times 10^{23} )</td>
</tr>
<tr>
<td>18</td>
<td>( \text{He}^* + \text{Cu} \rightarrow \text{e} + \text{He} + \text{Cu}^+ )</td>
<td>( 8.586 \times 10^{21} )</td>
</tr>
<tr>
<td>19</td>
<td>( \text{Ar}^+ + \text{e} + \text{e} \rightarrow \text{Ar} + \text{e} )</td>
<td>( 1.337 \times 10^{16} )</td>
</tr>
<tr>
<td>20</td>
<td>( \text{Ar}_2^* + \text{e} \rightarrow \text{Ar}^* + \text{Ar} )</td>
<td>( 1.04 \times 10^{23} )</td>
</tr>
<tr>
<td>21</td>
<td>( \text{He}^+ + \text{e} + \text{e} \rightarrow \text{He} + \text{e} )</td>
<td>( 6.042 \times 10^{16} )</td>
</tr>
<tr>
<td>22</td>
<td>( \text{He}_2^* + \text{e} \rightarrow \text{He}^* + \text{He} )</td>
<td>( 2.563 \times 10^{19} )</td>
</tr>
<tr>
<td>23</td>
<td>( \text{Cu}^{++} \rightarrow \text{Cu}^+ + \text{h}\nu )</td>
<td>( 3.2 \times 10^{21} )</td>
</tr>
</tbody>
</table>

Table 4.3: Calculated reaction rates averaged over the volume.

- The production processes of molecular ions include ion conversion (reactions 10 and 11) and associative ionization (reactions 13 and 16). The calculated density of \( \text{He}_2^+ \) is less than 1000 smaller than the density of \( \text{He}^+ \). The same holds more or less for the ratio \( \text{Ar}_2^+ / \text{Ar}^+ \).

For \( \text{Ar}_2^+ \) the dominant production process in the associative ionization with a rate in the order of \( 10^{23} \text{ m}^{-3} \text{ s}^{-1} \) whereas the ion conversion rate is in order of \( 10^{19} \text{ m}^{-3} \text{ s}^{-1} \).

Due to the low densities of \( \text{He}_2^+ \) and \( \text{Ar}_2^+ \) and consequently low fluxes to the cathode wall, the contribution of the molecular ions to sputtering can be neglected.
Table 4.4: Calculated contribution in percents of every reaction to the production/destruction rate for every species presented.
Table 4.4 presents the calculated contribution in percents of every reaction to the production/destruction rate for each relevant species. The ratio between the production and destruction rate $P_p/L_p$ is shown as well. This helps to determine the locality/non-locality of the species.

- **Electrons**
  The most important electron production processes are the direct ($\sim 20\%$), and the stepwise ionization ($\sim 60\%$) of Ar. Ionization of Cu contributes with about 13\%. This is a substantial contribution especially in view of the fact that the Cu atom density forms less than 0.1\% of the gas mixture. The reason lies again in the ionization potential; this is lower than that of Ar and He. The Penning processes contribute with a only a few percents to the creation of electrons.

  From table 4.4 can be seen that the loss rate of electrons is two orders of magnitude lower than the production rate, so that $P_p/L_p \gg 1$, As it was expected the electrons are TS species.

- **Excited species**
  The Ar excited states are produced mainly (98.7\%) by electron impact excitation of the ground state and are predominantly lost by ionization (89.1\%). Only a few percents is lost by MEK processes (reactions 12, 13 and 14).

  For He$^*$ the production is 100\% due to electron impact excitation of the ground level. The Penning process that transforms Ar ground atoms into Ar$^+$ (reaction 17) appears to be the dominant loss process of He$^*$ (58.9\%); another substantial destruction channel is that of ionization (39\%).

  From table 4.4 it can be seen that the excited gas species are completely determined by the reactions occurring in the discharge: $P/L = 1$, which completely agrees with the considerations given in section 4.2. However, despite of the locality, they give an important contribution to the (stepwise) ionization. Around 80\% of the He ions are produced by stepwise ionization versus 20\% by direct ionization. For Ar$^+$ the ratio is more or less the same. As discussed in section 4.2, for LC species it is not necessary to solve the transport equation (4.3). This speed up the calculations. Concluding we may state that there is no need to employ the particle balance in full glory for the computation of the density of excited gas species Ar$^*$ and He$^*$. As LC species local production/destruction balances suffice. However, the species can not be removed since they serve as important intermediate stages in the ionization processes.

- **Molecular ions**
  For He$_2^+$ the production is mainly ($\sim 74\%$) due to ion conversion process and associative ionization ($\sim 26\%$). They are lost by dissociative recombination. The calculated He$_2^+$ density is in the order of $10^{14}$ m$^{-3}$. From table 4.4 it can be seen that these processes give minor contributions for the production/destruction of the other species. The He$_2^+$ are unprofitable in the discharge.
For $\text{Ar}_2^+$ the main production process (99.9%) is associative ionization (reaction 20). From table 4.4 it can be seen that this process have minor effect to the production/destruction of other species. The contribution of the ion conversion process to the production of $\text{Ar}_2^+$ is negligible (0.02%); moreover this process does not contribute to the production/destruction of any other species, hence it can be neglected.

The dissociative recombination of $\text{Ar}_2^+$ appears to be the main loss process of electrons. However, the calculated rate for this process is two orders of magnitude lower than the total ionization. Excluding this process will not affect the electron density because the ionization processes are the dominant processes in the discharge.

The ratio $\mathcal{P}_p/\mathcal{L}_p$ of $\text{He}_2^+$ is $\sim 18$. That means the $\text{He}_2^+$ is almost transport driven. This is not so surprising since $\text{He}_2^+$ has a high mobility. The ratio between the production and destruction processes for $\text{Ar}_2^+$ is 2 and indicates that the density of $\text{Ar}_2^+$ is predominantly determined by the local reactions in the discharge. At the condition under study, the locality of the $\text{Ar}_2^+$ allows even to ignore the presence of the Ar molecular ions; the density is small and the contribution to production or destruction for other species is negligible. The elimination of the molecular ions is discussed in next subsection.

### 4.4.2 Molecular ions

To analyse the possible consequences of ignoring molecular ions and the related reactions, two models were constructed: the first one is equipped with the complete set of species and reactions as shown in the tables 4.1 and 4.2; the second model is a simplified model with reduced chemistry according to the observations above, i.e. excluding molecular ions and the relevant reactions, and atomic recombination processes. For both models the power is set to 300 W while the pressure is chosen to be 2.3 kPa.

Figure 4.1 compares the electron, $\text{He}^+$ and $\text{Ar}^+$ densities obtained by the two models. It is clear that the presence of molecular ions does not affect the density distribution of other species behaviour in the discharge. They can be excluded in the reduced chemistry model.

### 4.4.3 Ladder climbing

In section 4.2 was shown, and in subsection 4.4.1 confirmed by the model, that the metastable levels are LC species. They are created predominantly by excitation and immediately lost by ionisation, contributing greatly to the ionization degree. However, in practice EEK processes to higher excited levels can also contribute to the ionization degree; the so-called ladder-climbing processes might be of importance [11]. In order to check the importance/unimportance of these processes, we will construct another model and again compare the results with the complete model.

From the same considerations as described above, we can expect that species in all the possible excited states are LC species and that they, after being created, are immediately
Figure 4.1: Comparison of the axial density profiles of two models with and without molecular ions.

ionized. So that we can consider that the total excitation rate equals to the total ionization rate. With this assumption, the reduced chemistry model can be further simplified excluding completely the metastable levels and hence omitting the intermediate step of excitation. So, the reactions left in the reduced chemistry model are presented in table 4.5. The EEK rate coefficients as a function of the mean electron energy are replaced with the rate coefficient for the total excitation. From the HEK processes we have left only the charge transfer processes of metal atoms with gas ions.

Table 4.6 presents the densities of the active species, averaged over the volume in comparison with the complete model. Table 4.6 shows small differences in the averaged values of the densities. The densities are slightly higher for the simple model. The densities of electrons and Ar ions are higher for the simplified model, while the density of He ions is lower. The reason is that the increased ionization in the simplified model results in lower electron energy. The ionization rate will be higher for species with lower ionization potential. For Cu ions the difference is more pronounced also because the increased Ar
Reduced chemistry module

Table 4.5: Reaction rate coefficients used in the simple model.

<table>
<thead>
<tr>
<th>Nr.</th>
<th>Reaction</th>
<th>Rate coefficient, $k_r$ (cm$^3$ s$^{-1}$)</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$\text{Ar} + e \rightarrow \text{Ar}^+ + 2e$</td>
<td>$k_{\text{exc}}(\varepsilon)$</td>
<td>[1]</td>
</tr>
<tr>
<td>2</td>
<td>$\text{Cu} + e \rightarrow \text{Cu}^+ + 2e$</td>
<td>$k_{\text{exc}}(\varepsilon)$</td>
<td>[1]</td>
</tr>
<tr>
<td>3</td>
<td>$\text{He} + e \rightarrow \text{He}^+ + 2e$</td>
<td>$k_{\text{exc}}(\varepsilon)$</td>
<td>[1]</td>
</tr>
<tr>
<td>4</td>
<td>$\text{Ar}^+ + \text{Cu} \rightarrow \text{Ar} + \text{Cu}^+$</td>
<td>$3.16 \times 10^{-10}$</td>
<td>[1]</td>
</tr>
<tr>
<td>5</td>
<td>$\text{He}^+ + \text{Cu} \rightarrow \text{He} + \text{Cu}^{++}$</td>
<td>$1.0 \times 10^{-9}$</td>
<td>[7]</td>
</tr>
</tbody>
</table>

Table 4.6: Species densities averaged over the volume. Comparison between the complete model and the simple one.

<table>
<thead>
<tr>
<th>Species</th>
<th>Density, m$^{-3}$</th>
<th>Complete Model</th>
<th>Simple Model</th>
</tr>
</thead>
<tbody>
<tr>
<td>e</td>
<td>$7.848 \times 10^{-18}$</td>
<td>$9.135 \times 10^{-18}$</td>
<td></td>
</tr>
<tr>
<td>$\text{Ar}^+$</td>
<td>$7.286 \times 10^{-18}$</td>
<td>$8.814 \times 10^{-18}$</td>
<td></td>
</tr>
<tr>
<td>$\text{He}^+$</td>
<td>$2.031 \times 10^{-17}$</td>
<td>$1.76 \times 10^{-17}$</td>
<td></td>
</tr>
<tr>
<td>$\text{Cu}^+$</td>
<td>$1.677 \times 10^{-17}$</td>
<td>$2.48 \times 10^{-17}$</td>
<td></td>
</tr>
<tr>
<td>Cu</td>
<td>$3.583 \times 10^{-19}$</td>
<td>$4.522 \times 10^{-19}$</td>
<td></td>
</tr>
</tbody>
</table>

ion density results in increased sputtering, hence the density of Cu atoms in the discharge. Nevertheless, there is not considerable change in the species densities.

Figure 4.2 presents the axial profiles of the electron density and the ion densities. It can be seen that the axial profiles of the main charged particles remains more or less the same. The difference lies in the regions where the plasma density is highest. Apart from the slightly changes in the maximum values of the densities, the spatial density distribution is not changed significantly. We have to conclude that the contribution to the ionization degree due to ladder-climbing is negligible at the conditions under study.

4.5 Conclusions

In a search of a reduced chemistry module for the description of the plasma behaviour in a HCD with a He-Ar-Cu mixture, estimative calculations and numerical simulations are performed. This reduced chemistry model aims to construct a lean and reliable model that can be used as part of a design tool of HCD lasers.

Analysing the various species and their behaviour we determined the species that are not essential for the discharge, meaning that the presence of these species does not affect the main plasma properties (spatial values of the electron density and mean energy) so that they are not essential for the plasma application.

Based on the estimative calculations, numerical simulations and the post processing of the modelling results, we can summarise the following:
The HCD plasma is characterised as an ionising electron excitation kinetics (EEK) plasma; the main ionic species is Ar\(^+\), despite the fact that Ar constitute only 5% of the gas mixture.

Being an ionising plasma, the atomic recombination processes can be neglected.

Electrons and ions are transport sensitive (TS) species, while excited species (atom and ion) are local chemistry (LC) species. This observation results in simplification of the continuity equations for the LC species as the transport term is zero.

The influence of the molecular ions is negligible at the conditions under study. Although in many cases the molecular ions could appear as an important recombination channel, it was shown that for the conditions under study all the recombination processes can be neglected with respect to the ionization processes.
• The contribution to the ionization degree due to ladder-climbing is negligible at the conditions under study.

Taking the statements above into considerations, enables to construct a lean and reliable model that can be used as part of a design tool of HCD lasers. With this chemical lean model it is than possible to find the best of the geometrical construction, the anode cathode patterns, the current settings, the fill-chemistry, etc.

References

A flexible platform for simulations of sputtering hollow cathode discharges for laser applications

Abstract. The PLASIMO modelling platform, extended with a cathode wall sputtering module is used to study the discharge processes and to optimise the design parameters of a sputtering hollow cathode discharge (HCD). We present PLASIMO simulations of a HCD used for laser applications. A time dependent sub-model is used to describe the behaviour of the plasma species. The sputtering yield at the metallic boundaries is calculated using an empirical formula. Copper is chosen as the cathode material and the discharge operates in helium with a small admixture of argon for more efficient sputtering. The optimal conditions for lasing of the infra-red (IR) copper ion line (780.8 nm) that have been determined experimentally are used as input conditions for the simulation model. Calculations are made for various gas mixtures. The observed quantities are compared with the experimental data obtained for the same discharge geometry and operating conditions. The agreement between the measured data and the results from modelling indicates that the main reactions in the model are correctly described. Therefore, it is believed that the present model can be used as a design tool in optimizing discharge studies for various applications based on the sputtering of the cathode material.
Chapter 5.

5.1 Introduction

Hollow cathode discharges (HCD) have been studied extensively for many years due to their wide range of applicability in different fields: atomic spectrometry, gas lasers, vacuum microelectronics, UV generators, plasma processing as etching, thin film deposition, surface treatment, etc. By experimental studies and theoretical modelling of the plasma processes in the HCD, a significant contribution to the understanding of the physics of processes in the discharge can be achieved. This will improve the results of various HCD applications.

Due to the high density of energetic electrons, the HCD provides favourable conditions for achieving large population inversion through charge transfer processes in rare-gas metal-vapour mixtures. The possibility to create a large longitudinally extended negative glow plasma, as well as the possibility to use the cathode sputtering as a source of metal atoms in the discharge, make the HCD suitable as an active medium for the excitation of laser transitions of non-volatile metals, such as Cu, Au, Ag, etc. [1, 2]. In so-called metal vapour ion lasers the required ground state metal atom density is commonly produced via cathode sputtering and the population inversion of the laser levels is achieved typically through charge exchange collisions with He, Ne, Ar or other rare gas ions.

Variants of hollow cathode laser geometry designs are developed and studied with the aim of improving the excitation efficiency and performance of the sputtering metal vapour ion lasers: transverse, longitudinal, high-voltage, etc. [1, 2, 3, 4]. By numerical modelling of different electrode configurations, information for various plasma discharge parameters has been obtained [5, 6, 7, 8, 9]. Recently, a hybrid modelling network consisting of several Monte-Carlo and fluid models was used to describe a longitudinal HCD at typical conditions for laser oscillation [10]. A reasonable agreement has been reached with the experimentally measured data [11, 12].

In spite of the decades of research and use of HCDs, there is still no general model that can be used as a tool to optimize the various discharge parameters and the HCD geometry by easily manipulating them. The aim of this work is to develop such a comprehensive simulation model for a hollow cathode glow discharge in a gas mixture of inert gas atoms and metal atoms, produced as a result of cathode sputtering, for an arbitrary geometry of the electrodes and for various discharge parameters. For this reason we are extending the PLASIMO modelling platform [13] to be able to handle that task by the implementation of the cathode wall sputtering module.

PLASIMO has been designed as a modelling toolbox, rather than as a monolithic code for one particular application. It comes as a set of C++ class libraries that can be used for the construction of (plasma-)physical models. At present, sub-models are available for various electromagnetic problems and for plasmas with various degrees of equilibrium. Moreover, PLASIMO offers modules for calculating temperature fields, the electron energy equation, the barycentric flow field and radiation transport. Finally, an electron Monte-Carlo module allows kinetic or hybrid simulations. The configurability of PLASIMO makes it a helpful tool in improving the understanding of the plasma medium and the various plasma processes. Moreover its graphical user interface offers the opportunity to involve non-software-experts in plasma model studies.
In the present work, we have used the PLASIMO’s MD2D sub-model. Originally MD2D was developed for the simulation of microdischarges for display technology [14, 15]. Later the model was incorporated in PLASIMO which allowed MD2D to profit from some PLASIMO features like its convenient (graphical) user interface and its build and test infrastructure. Since MD2D does not contain any hard-coded configuration data, the user is allowed to specify the complete configuration in an input file. Among the configurable items are the geometry, the electrode potentials, the species properties, the pressure and composition of the background gas, the transport coefficients and the reaction rate coefficients. It is also possible to construct a model using a graphical user interface, whose hierarchical structure guides the simulation, controls the calculation and allows the user to monitor the behaviour of the variables involved in the plasma model. As a result it is easy to test various discharge designs, which makes MD2D a convenient engineering tool in optimization studies of HCDs in general.

In this paper we present PLASIMO/MD2D simulations of a HCD used for laser applications. Cu is chosen as the cathode material, and the discharge operates in He with a small admixture of Ar for more efficient sputtering. The optimal conditions for lasing of the infra-red (IR) copper ion line (780.8 nm) as determined experimentally in the past [16] are used as input data in the model. The upper laser level is supposed to be excited mainly by asymmetric charge transfer between He\(^{+}\) ions and Cu atoms.

In section 5.2, a short general model description will be given and the extensions of the model that were realised will be outlined. The used input data and the main assumptions done in the simulations will be discussed in section 5.3. The modelling results and qualitative comparison with the experimental data will be presented in section 5.4. Finally, conclusions will be given in section 5.5.

### 5.2 Description of the model

To numerically describe the behaviour of the plasma particles in sputtering HCD plasma, the time dependent sub-model MD2D is used. One of the basic assumptions of MD2D is that the buffer gas formed by the majority species is uniformly distributed over the vessel, i.e. the gas temperature \(T_g\), pressure \(P_g\), and density \(n_g = \frac{P_g}{k_B T_g}\) are independent of spatial position. Moreover we assume that the temperature of all heavy particle species is the same. In this study the buffer gas is a mixture of He and Ar and it is one of main aspects of this investigation to change the mixture ratio He/Ar and to study how this will affect the features of the active species which include electrons, excited atoms, ions and sputtered atoms. The behavior of these species is studied by means of a multi-fluid approach solving the relevant species balance equations. For each active species \(p\) we can describe the time evolution of the density by the particle balance:

\[
\frac{\partial n_p}{\partial t} + \nabla \cdot \Gamma_p = S_p, \tag{5.1}
\]

where \(n_p\) is the density, \(\Gamma_p = n_p \mathbf{u}_p\) the flux density, while \(S_p\) the net source of species \(p\) is determined by the reactions in the discharge. This particle balance is based on the Boltzmann
equation (BE) and can be obtained by taking the first moment; that is integrating the BE for the species $p$ over the velocity space.

The expression for the flux density $\Gamma_p$ is obtained by taking the second moment of the BE of species $p$; that is integrating over the velocity space the product of the BE and the velocity. This leads to the well-known drift-diffusion (DD) equation:

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

(5.2)

where $E$ is the electric field, $D_p$ the diffusion coefficient and $\mu_p$ the mobility of species $p$. The mobility is taken to be negative for negatively charged species and (obviously) zero for neutral particles. The first term at the right-hand side gives the flux due to the electric field (drift) and the second term represents the flux due to density gradients (diffusion). Some simplifications are made in the derivation that leads from the first moment of the BE to the DD equation. In particular, the inertial terms and the viscosity terms are neglected and it is assumed that the mobility and the diffusion coefficients of species $p$ are determined by the collisions of these species with the buffer gas solely:

$$\mu_p = q/(m_p \nu_{p0}),$$

(5.3)

$$D_p = k_B T_p / (m_p \nu_{p0}),$$

(5.4)

where $\nu_{p0}$ is the collision frequency and $m_p$ is the particle mass. Thus choosing the mean collision frequency (for momentum transfer) as $\nu_p = \nu_{p0}$, only collisions with the buffer gas particles are taken into consideration.

An additional note should be made with respect to the diffusive flux. The term $D_p \nabla n_p$ in the DD equation is derived from the gradient of the partial pressure that appears in the second moment of the BE. In general, this gradient in the pressure should be split as $\nabla P = k_B T_p \nabla n + n \nabla (k_B T_p)$, where the first term leads to density-driven and the second to temperature-driven diffusion. In our model the influence of the $\nabla (k_B T_p)$ is neglected. This can be justified by looking at relative importance of the two terms which is obtained by dividing both left- and right-hand side by $P$ giving: $\nabla \ln P = \nabla \ln n + \nabla \ln T$.

For the electrons we can expect that these gradients are mainly pointed along the radial direction and that these are largest in the region close to the electrodes. Anticipating on the results of our model and using the knowledge from comparable studies [10, 17, 9, 18], we can state that $n_e$ will change over several orders of magnitude (typically more than 5) whereas the electron temperature (not to be confused with the mean electron energy) is expected to cover less than one order of magnitude over the same distance. Moreover, in the region close to the electrodes we may expect that the drift $\mu_e E n_e$, is by far the most dominant driving force of the particle flux density $\Gamma_e$ so that the correction of the diffusive flux by including the $\nabla T_e$-term is of minor importance. In the central region where the drift and diffusion are of comparable magnitude we deal with large values of the electron density. In that situation thermal conductivity is expected to equalize temperature difference so that we can neglect the $\nabla T_e$-term there as well. So for the electrons we can in any region neglect the influence of diffusion driven by temperature gradients.
For the heavy particles we apply the assumption that all heavy particles have the same uniform temperature distribution as that of the buffer gas, so that $n \nabla (k_B T_p)$ is eliminated trivially.

Note that for the electrons as well for the heavy particles species the (partial) pressure is determined by stochastic motion solely. The effect of the directed velocity as created by the $E$-field is dealt with in the drift term.

The electron energy balance equation is obtained by multiplying the BE of the electron gas with the kinetic energy $1/2m_e v^2$ and integrating the results over the velocity space. After some approximations this gives:

$$\frac{\partial n_e}{\partial t} + \nabla \cdot n_e u_e + \nabla \cdot P_e u_e + \nabla \cdot q_e = S_e. \quad (5.5)$$

Here $n_e = n_e \varepsilon$ is the electron energy density, $\varepsilon$ is the mean electron energy, $u_e$ the mean velocity of the electrons and $q_e$ the thermal flux density. The effective source term $S_e$ includes heating of the electrons by the electric field and the energy losses due to elastic and inelastic collisions. Using the relation between the mean electron energy and the electron pressure $P_e = \frac{2}{3} n_e$ and substituting $u_e = \Gamma_e / n_e$, the second and the third term together can be expressed as:

$$\nabla \cdot n_e u_e + \nabla \cdot P_e u_e = \nabla \cdot \frac{5}{3} \varepsilon \Gamma_e, \quad (5.6)$$

The heat flux density (fourth term in equation (5.5)) is assumed to be proportional to the gradient of the electron mean energy, according to [19] and [20]:

$$q_e = -\frac{5}{3} n_e D_e \nabla \varepsilon. \quad (5.7)$$

Substitution of equations (5.6) and (5.7) in equation (5.5) casts the electron energy balance in the same form as the particle balance:

$$\frac{\partial n_e}{\partial t} + \nabla \cdot \Gamma_e = S_e \quad (5.8)$$

with

$$\Gamma_e = \frac{5}{3} \mu_e E n_e - \frac{5}{3} D_e \nabla n_e. \quad (5.9)$$

The electric field is computed using the Poisson equation:

$$\nabla \cdot (\varepsilon E) = -\nabla \cdot (\varepsilon \nabla V) = \rho, \quad (5.10)$$

where $\varepsilon$ is the dielectric permittivity, $V$ is the electrostatic potential, and $\rho$ the space charge density given by:

$$\rho = \sum_p q_p n_p. \quad (5.11)$$
Chapter 5.

The system of equations is completed with the boundary conditions (BC). The computational domain is surrounded by the symmetry axis of the device at \( r = 0 \), the planes \( b_1 \) and \( b_2 \) (figure 5.1) and the physical boundaries: the electrodes and the dielectricum in between the electrodes.

At the symmetry axis and the planes homogenous Neuman BC are employed, meaning that the derivatives of the quantities in the directions perpendicular to these boundaries are set to zero: for all the active particles \( \nabla n_p \cdot e_n = 0 \), where \( e_n \) is the unit normal vector to the surface; \( \nabla n_e \cdot e_n = 0 \) for the electron energy density; and \( \nabla V \cdot e_n = 0 \) for the potential distribution.

For the physical boundaries we apply for the potential distribution the following: the voltages at the cathode and anode are set at fixed values in the present study \( V_c = -300 \) V and \( V_a = 0 \) V. The BC for \( n_p \) and \( n_e \) at the physical boundary are given by expressions for the flux densities \( \Gamma_p \) and \( \Gamma_e \). For the heavy particles (Cu excluded) we follow [21], BC are determined by the reflection coefficients and the secondary emission coefficients. The \( \Gamma \)-value of the sputtered metal atoms at all the metallic boundaries is obtained using \( \Gamma_{Cu} = -\sum \xi_i \Gamma_i \) that is the summation of the product of the flux density of the bombarding ions \( \Gamma_i \) and the corresponding sputtering yield \( \xi_i \). Here \( i \) runs over the collection of ions. For the electrons, the \( \Gamma \)-value at the cathode is obtained via \( \Gamma_e = -\sum \gamma_i \Gamma_i \), where \( \Gamma_i \) the flux density of the ion of type \( i \) and \( \gamma_i \) the corresponding secondary emission coefficient. The BC for the electron energy density is given by relating the energy flux density to that of the electron number density using the expression \( \Gamma_\varepsilon = \frac{4}{3} \Gamma_e \varepsilon \). In the case of a Maxwellian distribution this results into \( \Gamma_\varepsilon = 2\Gamma_e T_e \).

MD2D uses the control volume method for solving the species densities (equation (5.1)), the electron energy (equation (5.8)) and the electrostatic potential (equation (5.10)). Since the equations for these quantities essentially all have the structure of a convection-diffusion equation, the implementation provides code for solving a canonical form of that equation. The method for solving this so-called \( \Phi \)-equation is essentially the same as that in Pantankar’s textbook [22].

The code uses implicit time-stepping for solving transient problems. The time step is adjusted on the basis of a preset criterion for the maximum relative change of the field variables, typically in the order of a few percent. Relatively large time steps are allowed since an implicit scheme is used for the calculation of the electrostatic potential [23, 24] and the electron energy [24].

5.3 Input data

We have chosen to simulate the plasma behaviour in the discharge geometry that is used in metal vapour lasers design as described in [12]. This choice will allow comparing the results of the calculations to the experimental results, thus checking the reliability of the present modelling. The studied discharge geometry [12] is shown in figure 5.1. It consists of a cylindrical copper hollow cathode (4 mm inner diameter and 50 mm cathode length) and two anode rings at both sides of the cathode, forming a longitudinal HCD. The fixed
discharge voltage of 300 V is applied over a gas mixture of He-Ar with 5% Ar concentration at a total constant pressure of 2.3 kPa. These operating parameters were experimentally found to be the optimal conditions for lasing of Cu$^+$ IR line (780.8 nm).

As stated before, the He and Ar gas atoms are assumed to be uniformly distributed throughout the discharge volume. The gas temperature and pressure are needed as an input in the model to determine the gas density through the ideal gas law:

\[ n_g = \frac{P_g}{k_B T_g} \]

Since \( T_g \) is not known, we follow [11] where calculations are made for similar conditions and we assume a constant value of 1000 K. Moreover, the chosen value is within the accepted range for laser application [8, 6, 9, 25].

The transport coefficients \( \mu_p, D_p \) and the reaction rate coefficients \( k_r \) are required as input data in order to solve the set of balance equations that describe the dynamics of the various particle species. The species taken into account in the model and their transport properties are summarised in table 5.1.

<table>
<thead>
<tr>
<th>Species</th>
<th>( p\mu_p \times P ) (Torr cm$^2$V$^{-1}$s$^{-1}$)</th>
<th>( D_p \times P ) (Torr cm$^2$s$^{-1}$)</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>e</td>
<td>( f(\epsilon) )</td>
<td>( f(\epsilon) )</td>
<td>[26]</td>
</tr>
<tr>
<td>He$^+$</td>
<td>( f(E/N) )</td>
<td>Einstein relation</td>
<td>[27]</td>
</tr>
<tr>
<td>He$^*$</td>
<td>–</td>
<td>( 15.38 \times 10^2 )</td>
<td>[28]</td>
</tr>
<tr>
<td>Ar$^+$</td>
<td>( f(E/N) )</td>
<td>Einstein relation</td>
<td>[27]</td>
</tr>
<tr>
<td>Cu$^+$</td>
<td>( 7 \times 10^4 )</td>
<td>Einstein relation</td>
<td>[29]</td>
</tr>
<tr>
<td>Cu</td>
<td>–</td>
<td>( 13.45 \times 10^2 )</td>
<td>[30]</td>
</tr>
</tbody>
</table>

Table 5.1: The transport coefficients used in the model. The coefficients for the electrons are specified as a function of the mean electron energy \( f(\epsilon) \), while the mobilities of the ionic species are specified as a function of the reduced electric field \( f(E/N) \).
We assume that for the charged heavy particles, the Einstein relation can be used:

\[ D_p = \frac{k_B T_p \mu_p}{q}, \]  

(5.12)

where \( q \) is the elementary charge, and \( T_p \) the particle temperature, corresponding to the energy of the random particle motion.

For the buffer-gas ions the local field approximation is used, which assumes a direct relation between the particle energy distribution and the electric field, hence the transport coefficients are regarded as a function of the reduced electric field. The relations can be found in the literature as result of experiments and classical theories [27, 29].

For electrons however, substantial deviations from the local field approximation can be observed under typical discharge conditions. Therefore, rather than using a dependency of the reduced electric field, we assume the electron transport coefficients and the rate coefficients of the electron impact reactions to be functions of the mean electron energy, as in reference [31]. The relations have been calculated from collision cross section data by solving the electron Boltzmann equation. The electron-related input data for the fluid model are obtained from the freeware Boltzmann-equation solver BOLSIG+ [26]. These input data for the fluid model are presented in the form of lookup tables as a function of the mean electron energy.

The reactions taken into account and the relevant reaction rate coefficients are shown in table 5.2. The buffer gas atoms (He and Ar) are ionised mainly by electron collisions (re-

<table>
<thead>
<tr>
<th>Nr.</th>
<th>Reaction</th>
<th>( \Delta \varepsilon ), (eV)</th>
<th>( k_r(\varepsilon) ), (cm(^3)s(^{-1}))</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>( \text{He} + e \rightarrow \text{He}^* + e )</td>
<td>20.215</td>
<td>( k_r(\varepsilon) )</td>
<td>[26]</td>
</tr>
<tr>
<td>2</td>
<td>( \text{He}^* + e \rightarrow \text{He}^+ + 2e )</td>
<td>4.365</td>
<td>( k_r(\varepsilon) )</td>
<td>[26]</td>
</tr>
<tr>
<td>3</td>
<td>( \text{He} + e \rightarrow \text{He}^+ + 2e )</td>
<td>24.58</td>
<td>( k_r(\varepsilon) )</td>
<td>[26]</td>
</tr>
<tr>
<td>4</td>
<td>( \text{He}^* + \text{He}^* \rightarrow \text{He}^+ + \text{He} + e )</td>
<td>-15.85</td>
<td>8.7 \times 10^{-10}</td>
<td>[32, 33]</td>
</tr>
<tr>
<td>5</td>
<td>( \text{He}^* + \text{He} \rightarrow \text{He} + \text{He} )</td>
<td>6.5 \times 10^{-15}</td>
<td></td>
<td>[34]</td>
</tr>
<tr>
<td>6</td>
<td>( \text{He}^* + \text{Cu} \rightarrow \text{He} + \text{Cu}^+ + e )</td>
<td>-12.495</td>
<td>9.48 \times 10^{-10}</td>
<td>[9]</td>
</tr>
<tr>
<td>7</td>
<td>( \text{He}^+ + \text{Cu} \rightarrow \text{He} + \text{Cu}^+ )</td>
<td>1.0 \times 10^{-9}</td>
<td></td>
<td>[9]</td>
</tr>
<tr>
<td>8</td>
<td>( \text{Cu} + e \rightarrow \text{Cu}^+ + 2e )</td>
<td>7.72</td>
<td>( k_r(\varepsilon) )</td>
<td>[26]</td>
</tr>
<tr>
<td>9</td>
<td>( \text{Ar} + e \rightarrow \text{Ar}^+ + 2e )</td>
<td>15.76</td>
<td>( k_r(\varepsilon) )</td>
<td>[26]</td>
</tr>
<tr>
<td>10</td>
<td>( \text{Ar}^+ + \text{Cu} \rightarrow \text{Ar} + \text{Cu}^+ )</td>
<td>3.16 \times 10^{-10}</td>
<td></td>
<td>[9]</td>
</tr>
<tr>
<td>11</td>
<td>( \text{He}^* + \text{Ar} \rightarrow \text{He} + \text{Ar}^+ + e )</td>
<td>-4.455</td>
<td>1.67 \times 10^{-10}</td>
<td>[9]</td>
</tr>
</tbody>
</table>

Table 5.2: Reactions rates and rate coefficients used in the model. \( \Delta \varepsilon \) is the energy cost of the forward process. The electron related reactions are specified as a function of the mean electron energy \( k_r(\varepsilon) \).

actions 3 and 9). The ionisation through the metastable levels (reaction 4) also contributes to the ionization degree. Since it has been demonstrated that the important metastable helium states participate in similar reactions with very similar rate coefficients [33], we
have combined them into one effective level, lying at 20.215 eV. Electron impact excitation from the metastable levels to higher levels is not taken into account, because the higher levels decay back radiatively to the metastable level, so that the net effect is small [33]. The Ar metastable levels are not taken into account in the present model because of the negligible role in the production processes of the Cu ions, compared with that of He∗, as was shown in [10].

The cathode is primarily bombarded by buffer gas ions which cause the sputtering of the cathode surface. The process of sputtering could be treated as a probability process in which every time an energetic particle impinges the cathode, ejection of atoms can occur with a certain probability depending on the properties of the incoming particle (mass, energy and incidence angle), the properties of the solid (structure and orientation of the solid) and the surface binding energy. The sputtering rate is usually expressed in terms of the sputtering yield ξi, giving the mean number of atoms removed per incident particle. We have opted to calculate the sputtering yields as a function of the bombarding energy using Yamamura’s empirical formula [35], that is based on a compilation of the available sputtering yield data and could predict the energy dependent yields for any ion-target combination. The proposed formula is for perpendicular bombardment and for sputtering of pure materials, which is a reasonable assumption for this application, because the ions are accelerated to the cathode by the electric field.

In the present model, constant values for the sputtering yields are used, calculated at constant incident ion energy. Since at present the ion energy is not calculated in the model, assumptions for the mean ion energy have to be made. The energies of the bombarding ion species in the discharge is radically different, because of the dynamics in the sheath region of the hollow cathode discharge [5, 6]. Crossing the sheath, the helium ions undergo symmetric charge transfer reactions, creating slow ions and fast neutrals. Since the cross section for this process can be large, upon reaching the cathode the He ions will have only a small fraction of the cathode fall energy. Their energy is assumed to be 50 eV giving a sputtering yield of 0.01 atoms/ion.

The argon ions, however, do not undergo significant symmetric charge transfer collisions in the cathode fall region, because the density of the parent argon atoms is low. Since the cross section of this process is low, the argon ions arrive at the cathode surface with almost full energy. As a consequence, a significant contribution to sputtering of copper atoms comes from the argon ion flux. At full cathode fall energy of 300 eV, the calculated sputtering yield is 0.8 atoms/ion.

The sputtered copper atoms can diffuse into the plasma or back toward the cathode. They can also be ionised in the glow as the ionisation mechanisms include electron impact ionisation (reaction 8), charge transfer reaction (reactions 7 and 10) and Penning ionisation (reaction 6). The formed Cu ions in the plasma can also produce sputtering. As it was first suggested in [5] and later shown in [10], Cu ions play a quite important role in sputtering (i.e. self-sputtering), which is mainly attributed to their high energy when bombarding the cathode. The metal ions, like the argon ions, do not undergo significant symmetric charge transfer collisions in the cathode fall region, because the copper neutral density is low. The only important effect that will slow down the metal ions is the elastic collisions with buffer
gas atoms. Hence, the metal ion energy at the cathode is only slightly less than the cathode fall \([5, 8]\) and in the present calculations, the Cu ion energy is assumed to be equal to the cathode fall. The sputtering yield of Cu ions is much larger due to the energy and mass difference of the ions. At 300 eV the calculated sputtering yield of Cu\(^+\) is 1.1 atoms/ion.

We use constant secondary electron emission coefficients of 0.2 for the He ions and 0.1 for the Ar ions \([36]\). For the Cu ions, the expression \(\gamma = 0.2 + 10^{-3}U\) is used \([8]\).

5.4 Results and discussion

5.4.1 Spatial structure

Figure 5.2 presents the potential distribution inside the cathode cavity. As a whole, the plasma potential inside the cathode cavity is negative. The discharge volume consists of two regions: the negative glow region which is nearly field-free with a space potential close to that of the anode, and the cathode-fall region, where all the applied voltage is concentrated. The change of the potential as a function of the radius is presented in figure 5.3. The potential is equal to \(-300\) V at the cathode wall, and rises rapidly as a function of distance away from the cathode. The cathode fall region occupies a thin layer adjacent to the cathode: its thickness changes from 0.3 mm at the anode ends of the cathode \((z=25\) mm\) to 0.5 mm at the centre \((z=0\) mm\). The electric field in this region is in order of 10 kV/cm. The electrons, leaving the cathode surface, are accelerated by the electric field in the sheath region, obtaining energies up to the cathode-fall voltage (see figure 5.4). These electrons are trapped in the negative glow region of the plasma and lose their energy in ionizing and exciting collisions in the plasma.

We compared the simulated local dependence of the mean electron energy on the reduced electric field with the \(\varepsilon(E/N)\) table that was generated by BOLSIG+. The substantial deviations indicate that the local field approximation is not applicable. This is no surprise in view of the strong electric field and energy gradients in the sheath region.

In axial direction at the discharge axis, the potential changes significantly: from slightly positive value (2 V) near the anode rings \((z=-25\) mm, \(z=25\) mm\), it decreases with the distance from the anodes to \(-60\) V in the centre of the cathode cylinder. Such potential
Influence of the He-Ar ratio

Figure 5.3: Calculated radial potential profiles for two axial positions: in the centre of the discharge (z=0) and close to the anode (z=25 mm).

Figure 5.4: Calculated radial mean energy profiles for two axial positions: in the centre of the discharge (z=0) and close to the anode (z=25 mm).

The change in axial direction results in axial electric field value of about 50 V/cm in the whole discharge volume. The latter gives rise to an electron current in the longitudinal direction, from the centre of the cathode toward the anodes. Near the anode sides the higher plasma potential gives rise to stronger electric field, resulting in higher electron energies and consequently more efficient electron impact ionisation. This leads to a higher electron density in this region. Figure 5.5 gives the contour plot of the calculated electron density spatial distribution inside the discharge cavity. The density distribution is strongly non-uniform in axial direction and it is characterised by a maximum near the anode ends of the cathode. This can be expected because of the efficient electron impact ionisation in this region. In fact, electron impact ionisation is the dominant production process for the electrons. The same non-uniformity is also observed in the other plasma species density profiles (figure 5.6). The maximum density of the sputtered atoms is again situated near the anode ends.
Figure 5.5: Calculated electron density distribution. Each grey shade denotes an interval of $2 \times 10^{18} \text{m}^{-3}$, from white (0 m$^{-3}$) to black (1.6 $\times 10^{19} \text{m}^{-3}$).

Figure 5.6: Calculated helium ion 5.6(a), argon ion 5.6(b), copper atom 5.6(c) and copper ion 5.6(d) density distributions.

The density distribution in radial direction, shown in figure 5.7 indicates also a strong non-uniformity. In our experiment done in the past, this radial non-uniformity has not been investigated, but similar results have been observed by Veldhuizen [6] and Lichtenberg [37]. The copper atom density profile (figure 5.7(a)) indicates a depletion in the centre of the glow with maximums close to the walls, as it is expected because the sputtering of the wall material. The ionic species densities (figure 5.7(b), 5.7(c), 5.7(d)) are concentrated mostly...
Influence of the He-Ar ratio

Figure 5.7: Calculated radial profiles of several species density in the centre of the copper cylinder.

5.4.2 Experimental validation

Figure 5.8 presents the calculated species density profiles plotted with the measured emission intensity of different lines. As it is difficult to measure directly the calculated plasma electron and plasma species densities, we make here a qualitative comparison with the optical emission intensities of He and Cu lines as a function of their axial position. On the other hand, in the model the emission intensity profiles cannot be calculated explicitly at present, because for such a calculation the level population of the individual excited levels are required. Since the main production process of Cu ions is the electron impact ionisation and the population of the upper level of the Cu atom transition is attributed to the same process, we assume that the measured axial spontaneous emission profile of a Cu atom line will represent with certainty the axial behaviour of the Cu ion density. Figure 5.8(a) compares the calculated Cu ion density distribution with the measured Cu atom
line (510.6 nm) intensity. There is a very good agreement between calculated and measured profiles. In figure 5.8(b) and 5.8(c) the calculated He metastable and He ion densities are compared to the measured axial intensity profiles of a He atom line (587.6 nm) and a He ion line (468.6 nm), respectively.

The agreement in figure 5.8(c) is less satisfactory, this can be attributed mainly to the accuracy of the measured results, because of the low intensity of the considered helium ion line. Although there are many uncertainties in the present model because of the numerous assumptions and the precision of the input data, the agreement between the calculated and the measured data is fairly. The general behaviour of the axial distribution is reasonable and illustrates that the main processes taken into account in the model are correctly described.
5.4.3 Influence of the Ar percentage

In order to further validate the present model with the experimental observations, calculations with different input conditions have been done.

Figure 5.9 presents the axial distribution profile of different plasma species at different Ar concentrations – from 0 to 10%. As shown in [10], the Ar concentration has a quite significant effect on the plasma characteristics and hence, on the laser excitation efficiency. Experimental studies show that the copper density needed for lasing is hardly achieved without addition of Ar and the measured laser power reaches a maximum at 5% Ar concentration.

The electron density clearly increases with the addition of Ar to the gas mixture (see figure 5.9(a)). The reason is that Ar has a lower ionisation potential than He; hence, when more Ar is present in the gas mixture, electron impact ionisation will be more efficient, leading to a higher electron density. This also leads to more efficient production of argon ions. Thus, the Ar ion density (5.9(b)) obviously increases with rising argon addition and its profile is almost identical to the electron density profile for all studied Ar concentrations above 1% Ar concentration. This shows that the Ar ions are the dominant ionic species at the discharge conditions under study, in spite of the fact that the Ar gas constitutes only a few percent of the total gas mixture.

Contrary to the efficient production of argon ions, the rate of the electron impact ionisation process of helium decreases if the Ar concentration increases. Penning ionisation by \( \text{He}^+ \) plays a quite important role in the production of ions at low Ar concentration (\(< 1\%)\), because then the \( \text{He}^+ \) density is significant. Consequently, the He ion density decreases with increasing argon concentration.

The high density of \( \text{Ar}^+ \) bombarding the cathode, play a dominant role in sputtering while the He ions are not very effective. As a result, with increasing Ar concentration, as it is expected, the density of Cu and \( \text{Cu}^+ \) increase as well (figure 5.9(d), 5.9(e)). Similar behaviour of He, Ar and Cu line intensity with Ar concentration is observed experimentally under similar discharge conditions [38].

The addition of more Ar to the gas mixture has important consequences when the discharge is used for laser application: on the one hand it is beneficial for Cu atom production in the discharge, but on the other hand the lower He ion density leads to inefficient population of the upper laser level which is populated by asymmetric charge transfer of Cu atoms with He ions. The lower population efficiency of this level might explain the experimentally observed saturation and even decrease of the laser power with further increasing Ar concentrations. Another important consequence concerns the axial inhomogeneity in the all plasma species density profiles, which increase with rising the Ar percentage. The increased axial inhomogeneity leads to more non-uniform excitation of the laser levels in the discharge volume. As a result, some part of the HCD remains inactive and cannot contribute to efficient excitation of the laser levels. Moreover, it can introduce additional optical losses in the resonator and affect the operation of the laser tube.
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Figure 5.9: Calculated density profiles in a longitudinal direction at different Ar concentrations in the He-Ar gas mixture as a function of axial position.
Influence of the He-Ar ratio

5.5 Conclusions

The PLASIMO modelling platform has been extended to enable more profound studies of the discharge processes and the optimisation of the design parameters of a sputtering hollow cathode discharge. In particular, an implementation of a wall-sputtering module has been added to the time-dependent sub-model MD2D and it is used to describe numerically the behaviour of the plasma particles. The present model allows easy configuration of discharge geometry, pressure and composition of the background gas, electrode potentials, species properties, transport coefficients and reaction rate coefficients, so it is a convenient tool for optimisation of the operating parameters in an arbitrary discharge.

The potential of the model has been demonstrated by simulating the plasma processes in a sputtering longitudinal HCD used for laser applications. The spatial distributions of various plasma quantities are calculated at discharge conditions that are typical for laser oscillation of Cu 780.6 nm laser line. The obtained quantities have been compared with the experimental data, obtained for the same discharge tube and operating conditions. The calculations suggest a strong non-uniformity of the plasma species in both axial and radial direction, which is in good agreement with the experimental data. Simulations of the plasma behaviour in a gas mixture of He with different concentration of Ar have been made. Both experimental and theoretical results indicate that the addition of heavier gas atoms such as Ar is required to produce efficient sputtering yielding higher Cu atom density and consequently the needed concentration of Cu$^+$ for efficient ion laser oscillation. The modelling results shows that the He/Ar ratio has a quite significant effect on the plasma characteristics: the sputtered Cu atom density increases while the He ion density decreases which leads to decreasing the rate of the asymmetric charge transfer process between Cu atoms and He ions, hence to non-efficient population of the upper laser level. This explains the experimentally determined optimal He/Ar ratio for laser oscillation of Cu 780.6 nm laser line. The axial inhomogeneity in the density profiles of the plasma species increases with increasing the Ar percentage. The increased axial inhomogeneity leads to less efficient excitation of the laser levels along the discharge length. As a result, some part of the HCD remains inactive and cannot contribute to the laser oscillation, reducing the active laser length.

The agreement between the measured data and the results from modelling indicates that the main reactions in the model are correctly described and the results are consistent. Due to the various approximations made and the questionable precision of the input data, the model cannot be expected to be quantitatively accurate. Nevertheless, the results turn out to be in a good qualitative agreement with the experimental data and the general behaviour of the axial distribution can be well-understood. The model can therefore be used as a tool to optimise the discharge conditions in arbitrary hollow-cathode geometries for various applications, based on the sputtering of the cathode material.
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References

Influence of the He-Ar ratio


A hollow cathode discharge for laser applications: influence of the cathode length

Abstract. The influence of the geometry of a longitudinal hollow cathode discharge (HCD) excited in Cu cathode and He-Ar mixture is studied experimentally and theoretically. Special attention is devoted to optimization of the HC length to obtain stable and uniform laser medium with high excitation efficiency. The influence of the cathode geometry is demonstrated experimentally by the behaviour of the 780.8 nm Cu ion line. The dependence of the laser power and gain as a function of the cathode length segments is measured. The PLASIMO modelling platform is used to construct a model allowing more profound studies of the plasma processes and plasma behaviour at different conditions. Calculations at different cathode lengths are made and typical results such as spatial potential and plasma density distributions are presented and discussed. It is demonstrated that when increasing the cathode length the plasma density in the centre of the cathode decreases and the discharge tends to separate into two independent parts causing axial inhomogeneity and reducing the discharge efficiency. The results also suggest that lower limit of the cathode length exist. Below this limit an inversion of the axial electric field occurs, which can be regarded as a transition between conventional and high-voltage HCD at the conditions under study.
Chapter 6.

6.1 Introduction

Hollow cathode discharges (HCDs) are widely used as active media for a large variety of metal vapour ion lasers with wavelengths extending from the infrared (IR) to as short as 224 nm [1, 2]. The metal vapours in the discharge are commonly produced via sputtering of the cathode material and the population inversion is realised typically through charge exchange collisions between the sputtered metal atoms and rare-gas ions such as He\textsuperscript{+} and Ne\textsuperscript{+}.

The advantage to use a HCD plasma as active medium is mainly based on its properties and its special geometry. The closed shape of the cathode leads to a merging or overlapping of the negative glow from opposing cathode surfaces and consequently the intensity of emitted light from the common glow is greatly increased. This makes the HCD an excellent spectroscopic source. The efficient use of charged particles enables the charge carrier production by secondary emission and an efficient sputtering of the cathode surface. These properties make the HCD a convenient medium for laser operation.

The HCDs, as no other discharges, are known by a great diversity of geometrical configurations. It is known that the geometry of the electrodes and their mutual position has a strong effect on the operating characteristics and hence the discharge stability and discharge uniformity. Various HC laser geometry designs are developed and studied with the aim to improve the excitation efficiency and performance of the sputtering metal vapour ion lasers: transverse, longitudinal, high-voltage, etc. [1, 2, 3, 4]. By experimental studies and numerical modelling of different electrode configurations, information for various plasma discharge parameters has been obtained [5, 6, 7, 8, 9, 10]. The stability of the longitudinal HCD — a setup, consisting of successive hollow cathodes and anodes — is much better compared to the transverse HCD, but the disadvantage is that an axial inhomogeneity may exist. This strongly affects the excitation efficiency of the discharge [1, 2, 8, 4]. Therefore it is advised to make use of a geometry that consist of sequence anode-cathode patterns. In such an elementary pattern, or segment, the discharge must be as uniform as possible in terms of plasma density and species density distributions.

The aim of this work is to study and to determine the optimal length of the hollow cathode segment in order to obtain a stable and uniform active medium with a high excitation efficiency which will contribute greatly to increase the laser gain and the laser power.

We have studied experimentally a copper ion laser excited in He-Ar longitudinal HCD with variable length of the cathode. The influence of the cathode geometry is demonstrated by the behaviour of the 780.8 nm copper ion line, which has the highest gain while laser oscillation can be observed at comparatively low input lower. Because of the similar mechanism of laser lines excitation, the oscillation in the infrared may be regarded as a template for lasing in the UV spectral range [11].

The PLASIMO modelling platform [12] has been used to construct a model that enables more profound studies and to find the optimum in parameter space of these type of discharges. PLASIMO facilitates the construction of various geometries and evaluation of the influence of the geometry on the plasma behaviour. As a result it is easy to test various discharge designs and this makes the presented model a convenient engineering tool in the
optimisation studies of HCDs. In a previous paper [13](chapter 5) the fluid model MD2D was presented, with a sputtering module extension in order to describe the behaviour of HCDs. The modelling results were validated by comparisons with the experimental data at different conditions. In the present paper the model (with some improvements) will be applied to optimise the hollow cathode geometry.

This paper is organised as follows: in section 6.2, the experimental setup will be described. In section 6.3, a short general description of the model will be given and we will outline the improvements of the model that have been realised. The experimental and modelling results as well as a qualitative comparison will be presented and discussed in section 6.4. Finally, conclusions will be given in section 6.5.

6.2 Experimental

The experimental investigations are performed using a specially designed discharge tube which is described in details in [14]. Figure 6.1 represent its schematic view. It comprises

![Discharge tube design](image)

Figure 6.1: Discharge tube design comprising of 31 electrode segments in total. Cathodes with different lengths can be made by connecting together several electrodes as cathode. On the bottom a zoom picture is shown, which sample one single A-C-A configuration with cathode length of 20 mm.
a series of 31 cylindrical electrodes made of electrolytic oxygen-free copper (99.99% purity) with an inner diameter of 4 mm and an outer diameter of 15 mm; 16 electrodes of 10 mm length and 15 electrodes of 20 mm length with total length 460 mm. All electrodes are isolated from each other by 0.5 mm thick quartz rings. The electrode segments are mounted in a quartz tube with 15 mm inner diameter, hence the discharge can burn only inside the copper rings.

Each electrode can be connected independently to an electric power supply, allowing individual ballasting of the electrodes for better discharge stability. By connecting together several electrode segments, it is possible to configure longitudinal HCDs with different cathode lengths and different anode-cathode configurations. The anodes are grounded, the cathodes are connected in parallel and set to a negative voltage. In table 6.1 the investigated electrode configurations are shown.

Table 6.1: The investigated electrode configurations $L_n$. Each configuration is of the form $A(CA)_n$ and thus consists of repetition of $n$ number of cathode segments. The length of the cathode segments $l$ is varied. $L$ stands for the full length of the active medium, that is the plasma in the hollow cathodes.

<table>
<thead>
<tr>
<th>Laser tube configuration, $L_n$</th>
<th>$L_1$</th>
<th>$L_2$</th>
<th>$L_3$</th>
<th>$L_4$</th>
<th>$L_5$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Single cathode length $l$, mm</td>
<td>10</td>
<td>20</td>
<td>30</td>
<td>50</td>
<td>80</td>
</tr>
<tr>
<td>Number of cathodes $n$</td>
<td>14</td>
<td>15</td>
<td>10</td>
<td>7</td>
<td>5</td>
</tr>
<tr>
<td>Full active length $L = l \times n$, mm</td>
<td>140</td>
<td>300</td>
<td>300</td>
<td>350</td>
<td>400</td>
</tr>
</tbody>
</table>

6.3 Modelling

6.3.1 Numerical description

To describe numerically the HCD behaviour we use the model described in detail in [13] and references therein. For completeness, we will briefly outline here the model and the
Influence of the cathode length

The present model differs somewhat from the model discussed in [13]. The difference lies in the set of species and reactions.

The model constructed with PLASIMO is a time-dependent two dimensional fluid model, based on balance equations derived from the Boltzmann equation. The spatial and temporal evolution of the particle densities are described by the particle density balance equations:

\[
\frac{\partial n_p}{\partial t} + \nabla \cdot \Gamma_p = S_p, \tag{6.1}
\]

where \( S \) is the net source term due to the various reactions by which the species are created or destroyed. The flux density \( \Gamma_p \) is solved in the drift-diffusion approximation by:

\[
\Gamma_p = \mu_p E n_p - D_p \nabla n_p, \tag{6.2}
\]

where \( \mu_p \) is the mobility of species \( p \), \( E \) the electric field and \( D_p \) the diffusion coefficient. The mobility is taken to be negative for negatively charged species and zero for neutral particles. The first term at the right hand side gives the flux due to the electric field (drift) and the second term represents the flux due to density gradients (diffusion). We assume that the Einstein relation can be used to calculate the diffusion coefficients from the mobility:

\[
D_p = \frac{k_B T_p \mu_p}{q_p}, \tag{6.3}
\]

where \( k_B \) is the Boltzmann constant, \( q_p \) the charge, and \( T_p \) the particle temperature, corresponding to the energy of the random particle motion.

The electron energy is determined by solving the electron energy balance equation. It has a form similar to the continuity equation:

\[
\frac{\partial n_e}{\partial t} + \nabla \cdot \Gamma_e = S_e \tag{6.4}
\]

with

\[
\Gamma_e = \frac{5}{3} \mu_e E n_e - \frac{5}{3} D_e \nabla n_e. \tag{6.5}
\]

Here \( n_e = n_e \varepsilon \) is the electron energy density and \( \varepsilon \) is the mean electron energy. The effective source term \( S_e \) includes Ohmic heating and energy losses due to elastic and inelastic collisions.

Poisson equation is solved in the plasma region and the dielectric regions, in order to account for the influence of charged species on the electric field

\[
\nabla \cdot (\varepsilon \mathbf{E}) = -\nabla \cdot (\varepsilon \nabla V) = \rho, \tag{6.6}
\]

where \( \varepsilon \) is the dielectric permittivity, \( V \) the electrostatic potential, and \( \rho \) the space charge density:

\[
\rho = \sum_p q_p n_p. \tag{6.7}
\]
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Figure 6.2: Computational domain. C and A are the cathode and anodes while D refers to the dielectricum. The dimensions of A and D are fixed while the length of C changes from 10 mm to 80 mm. For the plasma region a rectangular control volume grid is constructed having 120 grid points in radial direction while in axial the number of grid points is varied. Note that the radial extend of the non-plasma regions (A, C and D) is not important.

The layout of the computational grid used for the simulations has practically the same geometry as that of a single cathode segment $l$ (see figure 6.1) with two anode rings at both sides. The schematic picture is presented in figure 6.2. The computational domain is bound by the symmetry axis of the device, the planes $z_0$ and $z_1$ and the physical boundaries: the electrodes and the dielectricum in between the electrodes. For symmetry consideration, the plane $z_0$ and $z_1$ lie physically in the middle of the cathode and anode respectively. So, practically we simulate half of the cathode region, half of the anode region and the dielectric in between. The length of the half anode region is fixed to 5 mm and does not change for all constructions; the same applies to the thickness of the dielectric, which is fixed to 0.5 mm. The half cathode length region is varied from 5 mm to 40 mm corresponding to cathode segments from 10 mm to 80 mm.

At the symmetry axis and the planes $z_0$ and $z_1$, homogeneous Neumann boundary conditions (BC) are employed, meaning that the derivatives of the quantities in the directions perpendicular to these boundaries are set to zero. For the potential distribution, the boundary conditions are the voltages at the cathode and anode, which are set at fixed values. The BC for $n_p$ and $n_e$ at the physical boundary are given by expressions for the perpendicular components of the flux densities $\Gamma_p$ and $\Gamma_e$. For the heavy particles (Cu excluded) we follow [16], BC are determined by the reflection coefficients and the secondary emission coefficients. We use constant secondary electron emission coefficients, i.e. independent of the bombarding energy, equal to 0.2 for He$^+$, 0.1 for Ar$^+$ [8] and 0.1 for the Cu$^+$. The $\Gamma$-value for the sputtered metal atoms at the metallic boundaries is obtained from the summation of the product of the flux density of the bombarding ions and the corresponding sputtering yield. We use constant values for the sputtering yield coefficients equal to 0.01 for He$^+$, 0.2 for Ar$^+$ and 0.5 for Cu$^+$. 
6.3.2 Species and reactions

The density and temperature of the background gas are assumed to be constant in time and uniform in space; the main gas is helium with an admixture of 5% argon. The total gas pressure is set to 2.3 kPa, which was experimentally found to be the optimal gas pressure for lasing. Table 6.2 lists the set of species for which the balance equations were solved, together with the transport coefficients used in the model and the respective references. The mobility of the electron is specified as a lookup table as a function of the mean electron energy \( f(\varepsilon) \). The data are obtained from the free-ware Boltzmann equation solver BOLSIG+ [17].

For the mobility of the ions the local field approximation is used, which assumes a direct relation between the particle energy distribution and the electric field, hence the mobility coefficients are regarded as a function of the reduced electric field \( f(E/N) \). Then the diffusion coefficients for the charged particles are obtained using Einstein relation (equation (6.3)). He* is an effective state that represents the triplet and singlet metastable states of He, and similarly Ar* represents the 4s metastable states of Ar. We assume that the resonance states decay instantaneously and do not contribute to stepwise ionization. Other states are not included in the model, as with this set of species we expect the model to capture the essential phenomena and the discharge behaviour.

Table 6.3 gives the complete set of reactions together with the reaction rate coefficients used in the model. The gas atoms (He and Ar) are ionized mainly by electron collisions: direct ionization (reactions 3 and 6) and stepwise ionization (reactions 2 and 5). The ionization through the metastable levels (reaction 13 and 14) also contributes to the ionisation degree. The ground state copper atoms are brought into the discharge by sputtering solely and the ionization mechanisms of Cu atoms include electron impact ionization (reaction 7), Penning ionization (reaction 10 and 11) or charge transfer with buffer gas atoms (reaction 8 and 9).

The process most responsible for the population inversion of Cu excited ion states is...
Table 6.3: Reaction rate coefficients used in the model. The electron impact reactions are specified as a function of the mean electron energy $k_r(\varepsilon)$.

<table>
<thead>
<tr>
<th>Nr.</th>
<th>Reaction</th>
<th>$k_r$ (cm$^3$ s$^{-1}$)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$e + He \rightarrow He^* + e$</td>
<td>$k_1(\varepsilon)$</td>
<td>[17]</td>
</tr>
<tr>
<td>2</td>
<td>$e + He^* \rightarrow He^+ + 2e$</td>
<td>$k_2(\varepsilon)$</td>
<td>[17]</td>
</tr>
<tr>
<td>3</td>
<td>$e + He \rightarrow He^+ + 2e$</td>
<td>$k_3(\varepsilon)$</td>
<td>[17]</td>
</tr>
<tr>
<td>4</td>
<td>$e + Ar \rightarrow Ar^* + e$</td>
<td>$k_4(\varepsilon)$</td>
<td>[17]</td>
</tr>
<tr>
<td>5</td>
<td>$e + Ar^* \rightarrow Ar^+ + 2e$</td>
<td>$k_5(\varepsilon)$</td>
<td>[17]</td>
</tr>
<tr>
<td>6</td>
<td>$e + Ar \rightarrow Ar^+ + 2e$</td>
<td>$k_6(\varepsilon)$</td>
<td>[17]</td>
</tr>
<tr>
<td>7</td>
<td>$e + Cu \rightarrow Cu^+ + 2e$</td>
<td>$k_7(\varepsilon)$</td>
<td>[17]</td>
</tr>
<tr>
<td>8</td>
<td>$Ar^+ + Cu \rightarrow Ar + Cu^+$</td>
<td>$3.16 \times 10^{-10}$</td>
<td>[9]</td>
</tr>
<tr>
<td>9</td>
<td>$He^+ + Cu \rightarrow He + Cu^+$</td>
<td>$1.0 \times 10^{-9}$</td>
<td>[9]</td>
</tr>
<tr>
<td>10</td>
<td>$He^* + Cu \rightarrow He + Cu^+ + e$</td>
<td>$9.48 \times 10^{-10}$</td>
<td>[9]</td>
</tr>
<tr>
<td>11</td>
<td>$Ar^* + Cu \rightarrow Ar + Cu^+ + e$</td>
<td>$4.4 \times 10^{-10}$</td>
<td>[9]</td>
</tr>
<tr>
<td>12</td>
<td>$He^* + Ar \rightarrow He + Ar^+ + e$</td>
<td>$1.68 \times 10^{-10}$</td>
<td>[9]</td>
</tr>
<tr>
<td>13</td>
<td>$He^* + He^* \rightarrow He^+ + He + e$</td>
<td>$2.9 \times 10^{-9}$</td>
<td>[22]</td>
</tr>
<tr>
<td>14</td>
<td>$Ar^* + Ar^* \rightarrow Ar^+ + Ar + e$</td>
<td>$1.2 \times 10^{-9}$</td>
<td>[23]</td>
</tr>
</tbody>
</table>

Asymmetric charge transfer between the Cu atoms and He ions (reaction 9). Since the process requires good energy overlap between the excited levels of the Cu ion and the rare gas ion ground state, the process populates selectively the $6s^3D_3$ level of Cu$^+$. Figure 6.3 compares the partial energy level diagram of the CuII system with the energy levels of He$^+$ and Ar$^+$. The laser transition $(6s^3D_3 - 5p^3F_4)$ gives rise to a number of transitions in the IR spectral range with the strongest transition at 780.0 nm. This transition is of special interest because the upper laser level $(6s^3D_3)$ is also the upper level for a number of potential transitions in the 150 – 170 nm (VUV) spectral range. Although UV laser action may be of major interest, we have performed the experiments in the IR spectral range using He-Ar mixture, because the IR operation offers a number of advantages: low laser thresholds, allowing a number of parameters to be measured and the possibility of considerably shorter laser tubes, making it more convenient to study a variety of different HCD geometries.

6.4 Results and discussion

All experiments and calculations are made for a cylindrical copper hollow cathode with 4 mm inner diameter and a variable length of the single cathode segments and two ring anodes at both ends of the cathode (see figure 6.1). The experimentally determined optimal conditions for laser oscillation of the 780.0 nm copper ion line, are used as an input for the model: a mixture of He-Ar with 5% Ar concentration and a total constant pressure of 2.3 kPa.
6.4.1 V-I characteristics

The measured voltage-current characteristics are presented in figure 6.4. The measurements are done for different configurations $L_n$ (see table 6.1). Experimentally it was observed that the discharge stability is influenced by the variation of $l$ and that it is more stable at smaller $l$. For $l = 80\, \text{mm}$ despite of the low voltage the discharge is limited due to arcing. For $l = 10\, \text{mm}$ the voltage-current characteristic is completely different from the other cases: the voltage grows approximately from 400 V at 0.17 A/cm$^2$ (3 A) up to about 500 V at 0.6 A/cm$^2$ (10 A). Such a V-I characteristic is more typical for the high-voltage HCD construction [2, 9, 24]. For the longer cathode segments $l = 20 - 80\, \text{mm}$ the V-I characteristics are very similar: almost flat thus having a low slope resistance. This is typical for the conventional hollow cathode discharge: a large increase in current density occurs at nearly constant voltage [25]. At constant current density and pressure, the discharge voltage is lower for longer cathodes.

Figures 6.5 and 6.6 show the simulated potential distribution and plasma density distribution for four lengths of the cathode: $l = 10\, \text{mm}$, 15 mm, 20 mm and 80 mm. The $l = 15\, \text{mm}$ case is simulated in order to get insight into the transition between the high-voltage operation (at $l = 10\, \text{mm}$) and conventional HC operation (at $l = 20\, \text{mm}$). Although the computations are based on cylindrical symmetry we show, for clarity, the complete A-
Figure 6.4: Measured voltage-current characteristics for the studied configurations $L_n$ (see table 6.1) with single cathode segment lengths $l = 10 - 80$ mm.

C-A cavity: the cathode segment and two anode rings at both sides. As shown recently [13], the potential distribution is inhomogeneous in both radial and axial directions. The non-uniform potential distribution, especially found for longer cathodes, is associated with a radial and axial electric field component. The axial electric field in the negative glow is responsible for the longitudinal current. The field distribution for $l = 20 - 80$ mm cases, has maxima close to the anodes, and in this regions the potential is slightly positive. As a whole the potential in the plasma is negative. The axial extension of the cathode segment enforces the axial electric field component. The axial electric field pulls the electrons to the anode, but then the axial field also leads to decreasing of the cathode fall regions. This results in a decrease of the plasma density in the centre of the cathode (figure 6.6) and leads to back-diffusion of the electrons inside the cathode. Obviously, the axial electric field (slope of potential) has to be strong enough to overcome the back-diffusion. When decreasing the cathode length down to 15 mm the regions with positive potential merge at the centre of the cathode and there is practically no axial component of the electric field. Further decreasing the cathode length down to $l = 10$ mm, due to the overlap of positive potential regions, the maximum potential is in the centre of the discharge cavity. Such a potential distribution and V-I characteristics show an operating regime of the plasma different from the conventional hollow cathode operation. For $l = 10$ mm the diffusion is directed to the anode (note that this is the case even for longer cathodes in the regions very close to the anode) and the diffusion is fast enough to evacuate all created electrons. To slow down the diffusion, an inversion of the axial electric field occurs to compensate the enhanced diffusion losses. Consequently there is a limit of the cathode segment length, below which it is not any longer a conventional hollow cathode operation.

It is found that for increasing $l$ the non-uniformity increases and that the central $n_e$-
value decreases. The maxima of the electron density are close to the anode ends of the cathode, leading to an efficient electron impact ionization in these regions. Consequently in the same regions the ionic species will have maximum density values, which results in higher ionic fluxes to the wall and hence a higher sputtering rate. When the cathode is longer there are two distinct discharges at both edges inside the cathode. Decreasing the cathode length these two discharges starts to merge into one. At $l = 10$ mm the maximum of the plasma density is in the middle of the cathode cavity with much higher maximum electron density values of $\sim 1 \times 10^{20} \text{ m}^{-3}$, while for the longest cathode the maximum value is $\sim 4 \times 10^{19} \text{ m}^{-3}$. The optimum cathode length is found to be in the case where the peaks of the plasma densities from both edges just start to merge, i.e. $l \approx 20$ mm, where the highest laser power is observed (see below).

6.4.2 Axial profiles

The non-uniform potential and plasma density results in a non-uniform distribution of the charged particles and sputtered atoms [13]. The modelling results are confirmed experimentally by measuring the axial spontaneous emission intensity of Cu atom line (510.6 nm)
through the cathode slit, along the tube length. Typical longitudinal distributions of the optical emission intensity of the copper atom line are shown in figure 6.7 together with the calculated profiles for 4 different $l$: 50 mm, 30 mm, 20 mm and 10 mm. So, a qualitative comparison can be made between the measured axial spontaneous emission intensity of CuI 510.6 nm atom line with the calculated Cu atom densities profiles. This gives information for the nonuniform sputtering of the cathode material. A good agreement between the calculated and measured profiles is found.

The axial profiles appear to have almost the same non-uniform behaviour. There is a maximum at $2 - 3$ mm from the edges of the cathode and a minimum in the centre of the cathode region. Such a non-uniform behaviour is also observed in the density profiles of all the other active species [13]. In the centre of $l = 50$ mm case (figure 6.7(d)) the Cu atom density profile drops to less than 20% of its maximum value. Decreasing the cathode length results in decreasing the concavity in the centre of the cathode and at $l = 20$ mm the Cu atom density at the cathode centre is nearly 75% of its maximum value, hence the plasma is regarded more or less uniformly distributed inside the cathode cavity. Therefore the requested long and uniform laser active volume can not be achieved by increasing the cathode length; it can be achieved by subsequent HCDs, each with length not more than 20 mm.
Figure 6.7: Comparison of the calculated Cu atom density profiles and the measured emission line intensity profile of CuI 510.6 nm line at different length of the cathode segment \( l \). Note that although we simulate only half of the cathode segment, the figures present the axial profile for the whole cathode segment for clarity.

### 6.4.3 Laser gain and laser power

Laser oscillation on the 780.8 nm Cu ion line is observed experimentally in all five discharge tube configurations: \( L_1 - L_5 \) (see table 6.1). Figure 6.8 illustrates the measured laser gain and laser power per unit active length. The highest laser power is obtained when the laser tube comprises a series of cathodes with 20 mm length. This is in agreement with the spectroscopic measurements (figure 6.7), demonstrating best axial homogeneity at \( l = 20 \) mm. It also confirms that a good uniformity of the discharge is needed for efficient laser operation.

As mentioned before, in the model the laser levels are not treated separately, but only the total metal ion density is calculated. It is beyond the scope of this paper to analyse in detail the kinetics of the laser levels and to calculate exactly the laser power and laser
gain is a subject of a future work. Here we present the quantities from the model related to the output power and laser gain. One may expect that the laser power is proportional to the metal ion density in the cavity or, more precisely to the rate of the charge transfer reaction. These quantities can be obtained from the model for qualitative comparison. Figure 6.9 presents the calculated Cu ion density and the charge transfer reaction rate, averaged over the volume. The calculations show the same trend of the Cu$^+$ density as the measured laser power. The maximum density is obtained at a cathode length of 20 mm (figure 6.9(a)). Further increasing the cathode length leads to decreasing the Cu$^+$ density.
nearly in half at \( l = 80 \) mm. Also at cathode lengths shorter than 20 mm the density of the \( \text{Cu}^+ \) decreases.

The charge transfer reaction rate is highest at \( l = 10 \) mm and decreases with increasing cathode length (figure 6.9(b)): approximately with 75\% when changing \( l \) from 10 mm to 80 mm. Despite the higher production at \( l = 10 \) mm, the metal ions are efficiently lost by transport. In this case the field is reversed and the ions have additional losses by axial transport towards the anodes in which case they do not contribute to the secondary emission. From the modelling results we estimate that the ion transport losses are \( \sim 50\% \) faster at \( l = 10 \) mm than at \( l = 20 \) mm. This is the reason of the observed maximum of \( \text{Cu}^+ \) density at \( l = 20 \) mm.

### 6.4.4 Measurements on the laser efficiency

In figure 6.10 the laser output power dependence on the input power is presented. As mentioned earlier the discharge is more stable for shorter cathodes, while at \( l = 80 \) mm, in spite of the lower discharge voltage, the input power is limited by arcing. As the total discharge active length is different for different cathode lengths, we consider the laser power obtained per unit cm of cathode length as more reliable measure of the corresponding discharge configuration efficiency. The highest laser power is obtained when the discharge tube consist of alternating anodes and cathodes, with single cathode length not longer than 20 mm. The laser power obtained from longer cathode configurations (\( l = 30 \) mm, 50 mm and 80 mm) is lower, which is an indication that the excitation efficiency is lower, probably because a part of the cathode volume does not participate in the laser excitation process. The considerably lower laser power obtained from configuration with \( l = 80 \) mm cathodes

![Figure 6.10: The measured laser power as a function of the input power at the studied \( L_n \) configurations with single cathode segments \( l = 10 \) – 80 mm.](image)
is presumably also due to additional losses as a result of optical absorption in the longer inactive volume of the cathode region. It also confirms that a good uniformity is needed for high excitation efficiency.

### 6.5 Conclusions

We have studied experimentally and by numerical modelling HCDs with cylindrical copper electrodes, in He-Ar mixtures for laser applications. In this experimental and theoretical study special attention was paid to the influence of the cathode length in order to optimise the HC geometry for obtaining a stable and uniform active medium with high excitation efficiency. The influence of the cathode geometry is demonstrated experimentally by the behaviour of the intensity of 780.8 nm copper ion line. The PLASIMO modelling platform [12] has been used to construct a model that enables more profound studies and to find the optimum in parameter space of these type of discharges. Calculations at different cathode lengths have been presented and a qualitative comparison with the experimental data have been made.

The properties of the longitudinal hollow cathode discharge for which only part of the cathode surface is covered by the discharge differ essentially from those of the normal glow discharge between flat or convex electrodes. The longitudinal discharge in a HC having dimensions typical for laser applications is more stable than in the other HCD geometries in the sense that it less predisposed to arc formations. However, a strong axial non-uniformities exist. The axial potential and plasma density distributions are non-uniform with maxima close to the anodes. Decreasing the cathode segment length \( l \) leads to a more uniform axial potential distribution and a lower axial electric field. At \( l = 10 \) mm the potential distribution is completely different, with a maximum in the center of the cathode associated with an inverse axial electric field. These results, together with the measured V-I characteristics, exhibit an operation typical for high-voltage constructions [2, 9, 24]. It was found that a lower limit of the cathode length exists, beyond which the HC operation differs from the conventional HCDs. This lower limit is at \( l \sim 15 \) mm where an inversion of the axial electric field occurs to compensate the enhanced diffusion losses. This case marks a transition between the conventional HC and high-voltage operation at the conditions under study.

The non-uniform distribution of the potential and the electron density have an influence on the distribution of all species in the plasma. The calculations show a strong axial non-uniformity of the sputtered metal atoms in reasonable agreement with experimental data. The results show that the densities of the active species are highest close to the anode sides. The real active length is about 10 – 15 mm inside the cathodes at the anode sides. So, some part of the HCD remains inactive, reducing the real active length of the laser tube. As a result, part of the cathode remains inactive and cannot contribute to efficient laser oscillation. This also can introduce additional optical losses in the resonator and can strongly affect the laser operation. Decreasing the length of the cathode segment results in a more uniform distributions of the plasma inside the single cathode segment. It
was shown that the optimum cathode segment length \( l \) for lasing is at \( l \sim 20 \text{ mm} \), where the maxima of the plasma density at the two cathode edges just start to merge and the discharge tends towards uniformity. Thus, the possibility to obtain uniform and stable long discharge volume for lasing, is to use several subsequent cathodes each segment not longer than \( 20 \text{ mm} \). The above results are confirmed by the measured laser power dependence on the cathode segment length. We have demonstrated that the most efficient laser oscillation is achieved when the laser active volume comprises a series of anodes and cathodes, each cathode with a length of around \( 20 \text{ mm} \).

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References


Modelling of sputtering hollow cathode discharges for laser applications: influence of the cathode diameter

Abstract. The PLASIMO modelling platform has been used to construct a model of a sputtering hollow cathode discharge (HCD). The model allows in-depth studies of the plasma mechanisms and enables to find the optimum in the working conditions of the hollow cathode discharges. In this work special attention is devoted to the influence of the size of the cathode diameter. The spatial distribution of the potential and plasma density distribution are presented and the observations are discussed. Strong axial non-uniformities are observed at small diameters.

7.1 Introduction

Hollow cathode discharges (HCDs) are attractive radiation sources and employed in a wide variety of applications. The intensive light emission from the *negative glow* of the HCD, makes them excellent spectroscopic sources and convenient medium for laser operation. A large variety of metal vapour ion lasers is developed with wavelengths extending from the infrared (IR) to the deep ultra-violet (DUV) (as short as 224 nm) \[1, 2\]. Most of the devices use conventional HC discharges, that operate at voltages between 300 – 500 V. The two main types of discharge geometries used for laser applications are the *transversal* and *longitudinal* hollow cathode discharge. Both types, as well as various intermediate types of HCD laser geometry designs, are developed and studied with the aim to improve the excitation efficiency and performance of the sputtering metal vapour ion lasers \[3, 4, 5, 6, 7, 8\]. It is known that the dimensions of the HC geometry have strong effects on the operating characteristics and the spatial structure of the discharge, and hence on its stability and uniformity. In \[1\] it was found that the stability of the longitudinal HCD — a setup, consisting of successive hollow cathodes and anodes — is much better than the transverse HCD. However a considerable axial inhomogeneity may exist in the longitudinal setup. This can strongly affect the excitation efficiency of the discharge \[1, 2, 6, 9\].

The aim of this work is to study and to determine the optimal hollow cathode geometry in order to obtain a stable and uniform active medium with a high excitation efficiency. It is expected that such a study will give insight and thus contribute greatly to increase the laser gain and laser power.

In a previous paper \[10\] (chapter 6 of this thesis) we discussed the influence of the *cathode length* on the discharge efficiency. A time-dependent fluid model was constructed to enable in-depth studies in the optimisation of the HCD geometry. The modelling results were validated by comparisons with experimental data obtained for different cathode lengths. In the present contribution we investigate the influence of the hollow *cathode diameter* by means of modelling studies.

The paper is organised as follows: in section 7.2 the experimental setup will be outlined as well as the model. The model results will be presented in section 7.3 and followed by discussion in section 7.4. Finally, conclusions will be given in section 7.5.

7.2 Experimental and theoretical setup

Since the experimental setup and the model setup are described in details in \[10\] (chapter 6 of this thesis) we will only present a brief outline here.

The experimental setup consists of a metal cylinder with two anode rings at both sides, separated from each other with quartz rings. The length of the cathode is fixed to 30 mm and the inner diameter can be changed from 1 mm to 12 mm. The geometry is symmetric around the centre of the cathode (both axially and azimuthally). Hence, the simulation domain consists of a half of the cathode length, the anode region and the dielectric in between.
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The applied voltage is in the order of few hundreds volt. In the presented results the input power is fixed to 300 W. The operating gas is a mixture of He and Ar. The latter (Ar) is added to the main gas (He) to produce more efficient sputtering. The optimal concentration of Ar is determined to be about 5% [11] using modelling results, which were confirmed experimentally. The parameter $pd$ is kept constant at a value of 9.2 kPa mm, meaning that the increase of cathode diameter $d$ goes along with a decrease in pressure $p$.

The model used for describing the plasma behaviour is a time-dependent fluid model solving three types of equations: the continuity equation for the evolution of the relevant species, the energy balance equation for the electrons and the Poisson equation for computation of the electric field.

The gas atom densities are assumed to be uniformly distributed in time and space. The considered active species are He$^+$, He$^+$, Ar$^+$, Ar$^+$, Cu, Cu$^+$ and the electrons. The buffer gas atoms (He and Ar) are ionized by electron impact ionization, stepwise ionization and Penning processes. The metal atoms (Cu) are introduced into the discharge by sputtering due to ion bombardment of the cathode surface. The ionization processes of Cu include electron impact ionization, Penning processes and charge transfer processes with gas ions. More details about the species and reactions can be found in reference [10] (chapter 6).

7.3 Results

Figure 7.1 shows the voltage-current characteristics for HCD as predicted by the model at different inner diameters. The V-I characteristics of the different diameters are similar and have a low slope-resistance, meaning that a large increase of current can take place at nearly constant voltage. The voltage being lower at smaller diameters increases as a function of

![Figure 7.1: V-I characteristics at different diameters as predicted by the model for cathode length 30 mm and constant pd = 9.2 kPa mm.](image)
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Figure 7.2: Measured V-I characteristics at different diameters. Constant parameters are the cathode length of 70 mm and the $pd$ value of 9 kPa mm. Although the values are different from those in the modelling, the same trend is observed: the voltage is lower at smaller diameters and increases as a function of the diameter.

The same trend is observed in the experimental studies (figure 7.2) where the same parameters are kept constant: cathode length of 70 mm and $pd$ of 9 kPa mm. Although the values of these parameters differ from those in the model, the experimental results show the same trend: less voltage is needed at small diameters. The voltage increases from 300 V to 490 V with increasing the diameter from 2 mm to 6 mm.

Figure 7.3 presenting the plasma potential distribution for different cathode diameters, shows that the axial structure of the potential distribution changes drastically as a function of $d$. A strong axial non-uniformity is observed at $d = 1$ mm which decreases for increasing diameters.

Figure 7.4 presents the plasma density distribution. The plasma non-uniformity is more pronounced at small $d$ and decreases with increasing diameter. The plasma is more intense close to the anode edges inside the cathode cylinder. At small $d$ two separated plasma regions are present (close to the anode edges), which do not affect each other. Increasing $d$ the two separated discharge regions approach each other and start to merge, reducing the non-active region in the middle of the cathode.

7.4 Discussion

Due to space charge as offered by the plasma, most of the applied voltage falls in the radial direction across a thin sheath in front of the cathode. In this so-called cathode fall (CF) region the electron and ion transport is predominantly radial. The electrons emitted from the cathode are accelerated away from the cathode and launched toward the negative glow, where most of the energy is dissipated. Thus, the plasma is sustained by volume ionization
Figure 7.3: Potential distribution for different diameters. For clarity the figures represent the whole cavity: the hollow cathode cylinder (C), two anode rings (A) at both sides and the dielectric in between. The simulated region is marked with dashed line on 7.3(g). Note that for the latter three cases 7.3(e)–7.3(g) the potential variation along the axes is small and ranges from −50 V to 20 V. Moreover the radial scales are different.
Figure 7.4: Electron density distribution at different diameter of the cathode. For clarity, the figures represent the whole cavity: the hollow cathode cylinder, the anodes and the dielectric in between.
using the energy gained from the CF. The ions created in the plasma are all transported to the cathode and realize an important contribution to the secondary electron emission, which controls the current and plasma density and supports the sustaining of the plasma. Therefore the CF can be very thin and the ion flux density in the CF is high, producing high enough secondary emission and the discharge operates at high current density. For a given \( d \) a small increase of voltage results in a thinner CF increasing the field and hence the ion flux density in the CF. Consequently the secondary emission rate is greatly increased and the current density rises rapidly. This explains why the V-I characteristics presented in figure 7.1 are almost horizontal.

From the existing strong axial electric field at small \( d \)-values the electrons absorb additional energy. This could explain the shift in the V-I characteristics: when the axial non-uniformity is higher, the electrons absorb more energy and consequently less voltage is needed for obtaining the same current.

To get an in-depth understanding of the phenomena and trends revealed by the model, we will explore the model results using a quasi analytical approach. This approach is guided by one of the basic equations of the model namely the continuity equation:

\[
\frac{\partial n_p}{\partial t} + \nabla \cdot \Gamma_p = S_p, \quad (7.1)
\]

where the subscript \( p \) stands for one of active particle species. In steady state solution we have \( \frac{\partial n_p}{\partial t} = 0 \). If we apply this to the collection of charged particles we get, realizing that the creation of each electron is associated with that of an ion, \( S_e = S_i \). In steady state this leads to:

\[
\nabla \cdot \Gamma_e = \nabla \cdot \Gamma_i. \quad (7.2)
\]

After volume integration and employing Gauß theorem we get:

\[
\varphi_e = \iiint \Gamma_e dA = \iiint \Gamma_i dA = \varphi_i, \quad (7.3)
\]

where \( \varphi \) stands for the outward directed net flux. Taking for the integration volume a cylinder that is co-axial with the discharge we can write

\[
\varphi_i^{rad} + \varphi_i^{ax} = \varphi_e^{rad} + \varphi_e^{ax}, \quad (7.4)
\]

where \( \varphi \) is written as a sum of a radial and axial contributions.

For a proper understanding of the essential geometrical features of a HCD we first apply the above equation to a positive column (PC) of a cylindrical discharge. The PC is homogeneous in axial direction \( \frac{\partial}{\partial z} = 0 \), which leads to \( \varphi_i^{ax} = \varphi_e^{ax} = 0 \), so that \( \varphi_i^{rad} = \varphi_e^{rad} \). Taking an arbitrary radius the integral over the cylinder surface results in \( 2\pi rl \Gamma(r) = \pi r^2 l \langle S \rangle \) or

\[
\Gamma(r) = \frac{1}{2} r \langle S \rangle. \quad (7.5)
\]
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Figure 7.5: Schematic diagram of the electron and ion fluxes in PC-like discharge (top) and in a HCD (bottom). In PC-like discharges the ions (+) and electrons (−) move together radially to the mantle of the cylinder (top figure), while in HCD apart from the radial flux of ions, the electrons have a radial (due to secondary emission) and an axial component (to the anode).

So, in the mechanism known as ambipolar diffusion, the electrons and ions flux together to the mantle of the plasma cylinder; the flux density is equal for electrons and ions and scales with the radial position.

An essential difference between the HCD and a PC is that in the HCD-case the electron and ion flux densities differ in both direction and magnitude. The difference concerning the direction of the electron and ion fluxes is shown on figure 7.5, giving a schematic view of the fluxes in PC and in HCD.

The ion-flux $\varphi_i$ in the plasma volume is mainly directed radially to the cathode, while the flux of electrons $\varphi_e$ has, apart for the radial (due to secondary emission), also an axial component (to the anode). Due to the radial nature of $\varphi_i$, we can still apply $\Gamma_i(r) = \frac{1}{2} r \langle S \rangle$. However to find the flux density components of the electrons we have to apply Gauß law to a cylindrical volume bound by a radial cross section of the cylinder close to the anode and the mantle of the cylinder close to the cathode. In steady state this leads, using equation (7.4), to

$$
\varphi_{\text{rad}}^i = \varphi_{\text{ax}}^e - \gamma \varphi_{\text{rad}}^i
$$

$$
\varphi_{\text{ax}}^e = (1 + \gamma) \varphi_{\text{rad}}^i,
$$

(7.6)

where $\varphi_{\text{ax}}^i = 0$ and $\varphi_{\text{rad}}^e = -\gamma \varphi_{\text{rad}}^i$ are used. The latter is the essential HCD aspect of secondary electron generation: each ion colliding with the cathode surface has the probability...
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\begin{align*}
\varphi_e^{ax} &= \pi r^2 \langle \Gamma_e^{ax}(z = \frac{l}{2}) \rangle \\
\varphi_i^{rad} &= \frac{l}{2} \pi r \langle \Gamma_i^{rad}(r = R) \rangle.
\end{align*}

Substituting equations (7.8) into equation (7.6) results in the following relation between the radial and axial fluxes:

\[ \langle \Gamma_e^{ax}(\frac{l}{2}) \rangle = \frac{l}{r} (1 + \gamma) \langle \Gamma_i^{rad}(R) \rangle. \]

Note that \( \langle \Gamma_e^{ax} \rangle \) is averaged in \( r \) direction, whereas \( \langle \Gamma_i^{rad} \rangle \) is averaged in \( z \) direction.

The considerations given above provide means to interpret the model results. That will be consecutively done below:

- Equation (7.9) predicts that the flux density \( \Gamma_e^{ax} \) must be larger than \( \Gamma_i^{rad} \) by a factor of \( \frac{l}{r}(1 + \gamma) \). It is not a big surprise because the collecting area for ions (the mantle of the cylinder) is much larger than that for the electrons (the tube cross-sectional area); Numerically this equality is validated using figure 7.6, which shows the model results of the electron and ion flux densities at different diameters. Taking \( d = 6 \text{ mm} \) as an example it can be seen that indeed \( \Gamma_i^{rad} \) at \( r = 2.5 \text{ mm} \) differs from \( \Gamma_e^{ax} \) at \( z = 14 \text{ mm} \) by a factor of \( \sim (l/r)(1 + \gamma) \). The same applies for the other \( d \)-values. The \( \gamma \)-value is 0.1 for the Ar\(^+\), which are the main ionic species in the discharge.

Figure 7.6: Axial electron and radial ion flux-densities for different \( d \)-values. Due to the symmetry of the geometry, the flux densities are presented from the centre of the discharge (0,0).
• Figure 7.6 shows also that the radial profile of the ion density increases along the radius according to $\Gamma_i(r) = \frac{1}{2}r\langle S \rangle$ (equation (7.5)).

• The decrease of the maximum of the ion flux $\Gamma_i(R)$ with increasing diameter (attained at $r = R$ and constant power) can be explained by the fact that, due to the slight change in voltage, the constant power almost implies constant current. For the same (ion) current the current density can be small if the collecting area is large. The collecting area scales with the radius; so that $\Gamma_i(R)$ scales with $R^{-1}$.

• Moreover, it is clear that $\Gamma_i$ decreases with increasing $d$, while $\Gamma_e$ decreases as well, in agreement with equation (7.9).

Figure 7.7 shows how the on-axes potential changes at different cathode diameter. In general the current density, has two contributions due to drift and diffusion $\Gamma_p = \mu_p E n_p - D_p \nabla n_p$. For the ions only the radial drift is of importance; for the electrons the model shows a competition between drift and diffusion in axial direction. At low $d$-values when the pressure is high, most of the voltage drop is found over a small sheath gap generating a large ion drift. To compete with this, the $\Gamma_e$ close to the anode has to be large as well in order to transport the electrons to the anode. So that means that a strong axial $E$-field is needed and that an axial non-uniformity appears. By increasing $d$ (and thus decreasing $p$) the $E$-field driving the ions will be smaller meaning that $\Gamma_{e\text{ax}}$ will be smaller as well. This leads to a decrease in the axial voltage drop, meaning that electrons (and ions) can get relatively large densities in the plasma centre. Eventually the $d$ is so large that practically there is no axial $E$-field and the electron diffusion will be the driving force (see figure 7.7).

At $d = 8\text{ mm}$ the potential becomes uniform and there practically is no axial field component. The electrons are lost predominantly by diffusion to the anodes. Further
increasing $d$, the diffusion losses become high enough to evacuate all the created electrons in the plasma. To control the diffusion losses an inversion of the electric field can occur – the maximum potential is in the centre on the cathode cavity.

As shown above, the plasma non-uniformity at small inner diameter is more pronounced and decreases with increasing diameter. At small diameters the plasma is most intense close to the anode edges inside the cathode cylinder. Consequently in the same regions the electron impact ionization is more efficient and hence the ion density is higher exhibiting the same non-uniform spatial distribution as the plasma density distribution. This results in higher ion fluxes to the cathode wall in the region close to the anodes. This leads to non-uniform sputtering of the surface and non-uniform metal vapour distribution. Increasing the cathode diameter, the two separated intensive plasma regions from both edges approach each other reducing the concave in the middle of the cathode. At $d = 6$ mm they just start to merge and the plasma density distribution is more or less uniform. Further increasing $d$ turns the density distribution from a concave to convex shape. Figure 7.8 shows the on-axes changes in the plasma density profile at different diameters. The decreased radial ion flux with increasing $d$ reduces surface processes as secondary electron emission and sputtering. When $d$ becomes so large that a uniform potential distribution is reached, the diffusion losses become much more important. In order to avoid that the axial electron transport becomes too high, an inverse $E$-field is built up. In our previous paper [10] the same effect was found as the cathode-lengths: in the case of short cathode lengths a field inversion occurs in order to moderate the enhanced losses of electrons. Thus, increasing the length has the same effect as decreasing the diameter namely an increased non-uniformity; and an inverse electric field with decreasing the length. The limit aspect ratio $l/d$ is that for which the potential distribution is uniform so that practically no axial electric field exists. Approaching this limit at increasing $d$, a decrease of the electron density is observed. Contrary to this, approaching the limit at decreasing $l$ an increase of the electron density is observed. The reason for the observed decreased density is related to the fact that in
this study the parameter $pd$ is kept constant, meaning that with increasing $d$, the pressure $p$ decreases which results in a decrease of the plasma density. Obviously, the limit and the ratio of the HC dimensions is pressure-dependent and for each particular geometry an optimal pressure exists.

In view of the laser application, a good uniformity is needed, which is the case at bigger diameter. Another demand for laser application is a high enough plasma density and hence concentration of Cu, which is fulfilled at smaller diameters. This suggests that there is a maximum of the laser power as a function of diameter. In [10] the maximum laser power was obtained when the discharges from the both edges of the cathode just start to merge, which is just before approaching the limit, i.e. when the discharge is more or less uniform, but an axial electric field still exists to transport the electrons to the anodes from which the electrons gain additional energy. Following the same considerations, as a function of the diameter this happens when the cathode diameter is $\sim 6\, \text{mm}$. The aspect ratio for which both discharge regions just start to merge, before the limit, are when the length is about 5 times larger than the diameter at the operation conditions under study. If this ratio $l/d$ is bigger than 5, a strong non-uniformity starts to occur, while for smaller values an inversion of the electric field occurs.

### 7.5 Conclusions

In a search of the optimum in hollow cathode geometry for laser application, calculations are made for different values of the inner diameter of the cathode while keeping the parameter $pd$ constant. Strong axial non-uniformities of the potential and plasma distribution are observed at small diameters, which decreases for increasing diameters. In comparison with the PC of a classical discharge, where the discharge is axially uniform, the HCD geometry is essentially different in a sense that the flux densities of ions and electrons differ from each other in both direction and magnitude. The ion transport is predominantly radial to the cathode wall, while the transport of electrons has an axial and radial component: radially launched from the cathode due to secondary emission and in axial direction attracted by the anode. The axial non-uniformity appears in order to generate an axial electron current to the anodes. With increasing $d$, the ion flux-density decreases and the electron flux-density decreases as well, hence the axial non-uniformity decreases. This implies that the axial non-uniformity is linked to the size of the radius.

The collecting area of ions (the mantle of the cylinder) is much larger than that for the electrons (the tube cross sectional area) and hence the flux density differs by a factor, scaling with the length and radius.

The axial non-uniformity involves an axial electric field from which the electrons absorb additional energy, which could explains the shift in the current-voltage characteristic as a function of the diameter: when the axial non-uniformity is higher, the electrons absorb more energy and consequently less voltage is needed, in order to obtain the same current.

The plasma is more intense close to the anode edges and the discharge can be regarded as two separate discharge regions that not affect each other. Increasing the diameter
leads to more uniform plasma distribution. The two separated intensive plasma regions from both edges approach each other reducing the concave in the middle of the cathode. Further increasing $d$ results in completely uniform potential distribution for which almost no axial electric field is present. The diffusion losses become high enough to evacuate all the created electrons in the plasma. To compensate the diffusion losses an inversion of the electric field occurs and this turns the density distribution from a concave to convex shape.

In a previous paper [10] the same effect of decreasing the diameter was observed when increasing the cathode length (enhanced non-uniformity) and opposite — the same effect at increasing the diameter and decreasing the length (inverse electric field). From the point of view of laser action the non-uniformity is undesirable because the middle part of the cathode that remains inactive can not contribute to efficient excitation of the laser levels. Moreover it can introduce additional optical losses in the resonator. The maximum laser power was obtained when the discharges from the both edges of the cathode just start to merge, i.e. when the discharge is more or less uniform, while an axial electric field still exist to transport the electrons to the anodes and from which the electrons gain additional energy.

The results from the simulations predict a link between the diameter and length of the hollow cathode. According to the modelling results, the diameter should be around 5 times smaller than the length of the cathode at the operation conditions under study. In such configuration the discharge from both sides of the cathode just start to merge, so that the plasma is more or less uniform while an axial field still exist. If the ratio $l/d$ is larger than 5, a strong non-uniformity starts to occur, while smaller $l/d$ values invoke an inversion of the electric field.

Acknowledgments

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References

Chapter 7.


Abstract. This chapter presents an analytical model of a longitudinal hollow cathode discharge used for metal vapor lasers. The model describes the principle relations between the voltage, current, plasma density, and the axial structure of the discharge. Contrary to standard DC discharges, electron multiplication in the cathode fall is not necessary to produce ions, but rather to satisfy the electron energy balance. A self-sustainment condition for hollow cathode discharges is obtained from the electron energy balance. From this, it follows that there is a maximum voltage at which the cathode fall thickness tends to zero and the current density tends asymptotically to infinity. The discharge develops axial non-uniformity and an axial electric field in order to evacuate the created electrons to the anode. The axial profile of the plasma density is obtained from the electron continuity equation. It is shown that additional energy absorption from the axial field, similar to electron heating in DC positive columns, modifies the self-sustainment condition, which leads to a shift in the voltage-current characteristic, that depends on the cathode length.
Chapter 8.

8.1 Introduction

Hollow cathode discharges (HCDs) are widely used in applications in different fields: laser technology, atomic spectroscopy, UV generators, vacuum microelectronics, materials processing, etc. Although these discharges have a large variety of geometrical configurations, they are all characterised by the so-called hollow cathode effect: an exceptionally high discharge current, compared to conventional DC discharges at the same voltage, due to the cathode surface surrounding a large part of the plasma. The hollow cathode effect is accompanied by an exceptionally high plasma density, intensive light emission, and cathode sputtering, which are paramount for HCD applications.

The general physical principles of HCDs have been studied for decades and are discussed in many papers, on the basis of experiments, analytical models, and numerical simulations [1, 2, 3, 4, 5, 6, 7, 8, 9]. Several phenomena are considered responsible for the hollow cathode effect, in particular the electrostatic trapping of fast electrons in an oscillating motion inside the cathode, known as the pendulum effect [10, 11], and the enhanced secondary-electron emission by UV photons [1] and ions [5] created in the negative glow plasma inside the cathode. A number of papers is devoted to the role of metal vapour atoms due to cathode sputtering [9, 4]. However, the importance of each of these phenomena depends on the HCD geometry [5].

In several previous papers [12, 13, 14] (chapters 5, 6, 7 of this thesis) we investigated the spatial structure of longitudinal HCDs by comprehensive two-dimensional numerical simulations, which we compared with experimental results. These simulations are based on the self-consistent solution of the continuity and drift-diffusion equations for different plasma particle species, the electron energy equation, and Poisson equation. The models and their results are described in detail in chapters 3–7. In [14] (chapter 7) a quasi analytical approach was presented, guided by the structure of the main equations solved in the numerical model. Exploring the modelling results gives a proper insight in the trends revealed by the experimental work exposed in chapter 2.

In this chapter we present an analytical model of HCDs, with the aim to interpret the numerical results in order to get a more in-depth understanding of the phenomena and plasma behaviour. To that end, we revisit and combine some elementary theories and extend them to account for the axial non-uniformity of longitudinal HCDs. Comparison of the trends from the numerical model with the analytical solution can increase the confidence in the validity of the numerical model.

The chapter is organised as follows: In section 8.2 we present the geometrical configuration and summarise the observations from the numerical modelling results. In section 8.3 and 8.4 we discuss the self-sustainment conditions and the cathode fall thickness in HCDs. Section 8.5 analyses the radial transport, and it is followed by an analysis of the axial transport in section 8.6. Then in section 8.7 we discuss how the axial transport influences the self-sustainment condition. Finally, conclusions are given in section 8.8.
8.2 Configuration and physical principles

The configuration that is considered is shown schematically in figure 8.1. It consists of a copper cylindrical hollow cathode bounded on either side by a thin dielectric ring and an anode ring of the same inner radius. The geometry is symmetric around the center of the cathode (both axially and azimuthally). A DC voltage of a few hundred volts is applied between the cathode and anodes to sustain the discharge. The values of the control parameters are given in table 8.1.

Experimentally, the following features have been observed:

- The discharge in this configuration develops non-uniformities in axial direction in such a way that intense plasma regions are formed close to the anodes, while the central part of the cathode remains inactive.

- The plasma density attains a maximum value at small distance from the anodes and gradually decreases as a function of the axial position away from the anode.

- The non-uniformities are not influenced by the gas composition, the pressure of the metal vapours and the input current. They are mainly dependent on the dimensions of the hollow cathode.

These experimental observations were confirmed by means of simulation models. In a search for a lean chemistry model we have constructed and compared various models. It was found that the values and distributions of the main plasma parameters do not change much if a complex chemistry model is applied. Moreover, there is no substantial difference of the plasma density profiles comparing models with and without metal vapours. In the latter case the density distribution of the species that are essential for the laser action can be found by post-processing.

In view of the above, we concentrate in this analytical approach on the main plasma properties such as plasma density, potential and current, rather than plasma chemistry or the behaviour of the metal vapour atoms or other effects in the plasma. We derive an
Chapter 8.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Notation</th>
<th>Value</th>
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</thead>
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</tr>
<tr>
<td>cathode length</td>
<td>$2L$</td>
<td>50 mm</td>
</tr>
<tr>
<td>applied voltage</td>
<td>$V$</td>
<td>380 V</td>
</tr>
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<td>discharge current</td>
<td>$I$</td>
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<td>gas composition</td>
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</tr>
<tr>
<td>ion mobility</td>
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</tr>
<tr>
<td>Townsend coefficient</td>
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</tr>
<tr>
<td>secondary emission coeff.</td>
<td>$\gamma$</td>
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</tr>
<tr>
<td>energy per e-i pair</td>
<td>$W$</td>
<td>50 eV</td>
</tr>
</tbody>
</table>

Table 8.1: Parameters of the HCD configuration under study. The top part of the table gives the settings of the control parameters. In the bottom part, some main values of the averaged internal plasma quantities are given as found by the numerical model.

analytical model giving insights in the main plasma properties as a function of the control parameters such as voltage and geometrical dimensions.

The discharge behaviour in this configuration as found by numerical simulations, is illustrated in figure 8.2. The figures present only the simulated region of the geometry: half of the cathode with length of 25 mm, the anode and the dielectric in between. The species taken into account are the electrons and different ionic and excited neutral species of helium, argon, and copper (see table 6.3 in chapter 6). Particle source terms due to ionization, excitation, and other plasma chemistry are calculated using rate coefficients as a function of the mean electron energy. Wall recombination, secondary electron emission, and copper sputtering are accounted for by wall flux boundary conditions.

Figure 8.2 shows that, owing to the presence of a plasma, the anode potential propagates along the cylinder axis inside the cathode, such that most of the applied voltage falls in the radial direction across a thin space-charge sheath in front of the cathode, the cathode fall (CF) region (figure 8.2(a)). Outside the CF, the potential is close to the anode potential, with small radial variations of the order of the electron temperature to ensure quasi-neutrality of the plasma, and an axial gradient to ensure continuity of the plasma current.

The plasma is sustained by volume ionization (figure 8.2(e)) using the energy absorbed by electron acceleration in the CF (figure 8.2(f)) and these two regions feed to each other to sustain the discharge. What distinguishes the HCDs from conventional DC glow discharges is that the ions created in the plasma are all transported to the cathode where they give...
an important contribution to the secondary electron emission, so that less electron multiplication in the CF is needed to sustain the discharge. As a result, the CF is thinner, the ion density in the CF higher, and the discharge operates at higher current density for the same applied voltage. So that the transport of ions and electrons in the CF controls the current and plasma density. To study this more in detail, the cathode fall thickness will be analysed in the next section of this chapter (section 8.3).

In the CF the electron and ion transport is predominantly radial, while in the plasma apart from the radial transport the electron transport also has an axial component along the axis to the anode. The radial and axial transport will be analysed in section 8.5 and 8.6. Then, we will explore the influence of this axial non-uniformity on the trends of the plasma density and voltage, and on the self-sustainment condition.
8.3 Self-sustainment condition

The thickness $d$ of the CF in conventional DC discharges, adjusts to satisfy the self-sustainment condition: an electron emitted from the cathode by ion impact must cause, during its life time, the creation of enough ions to ensure the emission of a new electron. According to the Townsend model the flux of electrons $\Gamma_e$ in the CF results from the flux $\Gamma_i(0)$ of ions impacting the cathode and grows exponentially with the distance $x$ from the cathode as:

$$\Gamma_e(x) = \Gamma_e(0)e^{\alpha x},$$

where $\alpha$ is the Townsend coefficient characterising the ionization probability per unit of travelled length [15]. The Townsend coefficient $\alpha$ is determined mainly by the discharge gas: it is proportional to the gas particle density and not very dependent on the electron energy if this is sufficiently high (higher than the ionization potential). The effective emission probability per ion impinging the cathode surface, expressed as secondary emission coefficient $\gamma$, is determined mainly by the combination of ion species and cathode material and is relatively independent of the ion impact energy.

This analysis also applies to discharges ‘with a sheath’ or CF region, if we identify $d$ to be the CF thickness, the flux densities of electrons at the boundaries of the cathode fall are:

$$\Gamma_e(0) = -\gamma \Gamma_i(0), \quad \Gamma_e(d) = -\gamma \Gamma_i(0) \exp(\alpha d).$$

The ion flux entering the CF follows from current conservation:

$$\Gamma_i(d) - \Gamma_e(d) = \Gamma_i(0) - \Gamma_e(0),$$

yielding the self-sustainment condition [15]:

$$\Gamma_i(d) = \Gamma_i(0) \left[ 1 + \gamma(1 - \exp(\alpha d)) \right]. \quad \text{(8.3)}$$

In conventional DC discharges, most of the ions impacting the cathode are created in the CF, so that $\Gamma_i(d) \approx 0$, meaning that the factor given in the brackets must also be zero. This means that an electron leaving the cathode, crossing the CF generates $\exp(\alpha d) - 1$ positive ions that are transported to the cathode. Then in conventional DC discharges the cathode fall thickness $d$ is:

$$d = \frac{1}{\alpha} \ln \left( 1 + \frac{1}{\gamma} \right). \quad \text{(8.4)}$$

However, in HCDs the ions created in the plasma beyond the CF are nearly all transported to the cathode and give an important contribution to the secondary electron emission as well. So, from equation (8.3), we observe that if a non-zero ion flux enters the CF from the plasma $\Gamma_i(d) < 0$, then the term in the brackets also must be positive $[1 + \gamma(1 - \exp(\alpha d))] > 0$. Then we find that

$$d < \frac{1}{\alpha} \ln \left( 1 + \frac{1}{\gamma} \right). \quad \text{(8.5)}$$
We observe that in HCD the thickness of the CF can be smaller than the conventional DC discharges, since all the ions created in the plasma arrive at the cathode producing high enough secondary emission to sustain the plasma, the self-sustainment condition is satisfied with smaller $d$. Like in the conventional discharge, the cathode fall thickness adjusts to satisfy the self-sustainment condition, to ensure that enough ionization occurs to sustain the plasma.

### 8.4 Cathode fall thickness in HCDs

In this section we will calculate the thickness $d$ of the CF in HCDs. In the analysis that follows we will assume that $d$ is much smaller than the cathode inner radius $R$, so that we can neglect the curvature of the CF meaning that the radial position coordinate $r$ behaves as a Cartesian coordinate for $R - d < r < R$ (see figure 8.3). In this coordinate system we assume that the potential $U$ in the CF is parabolic and thus the electric field $E$ linear:

\[
U_{CF}(r) = -V \left(1 - \frac{(R - r)}{d}\right)^2, \quad (8.6)
\]

\[
E_{CF}(r) = -\frac{\partial U_{CF}}{\partial r} = \frac{2V}{d} \left(1 - \frac{(R - r)}{d}\right), \quad (8.7)
\]

where $V$ is the total voltage difference across the CF.

The value of the CF thickness $d$ can be estimated from the electron energy balance, e.g. by balancing the average energy absorption $H$ and the energy losses $W$, associated with the creation and loss of a single electron-ion pair. The loss term $W$ also has two contributions: energy losses due to collisions in the plasma and energy losses associated with electron transport to the anode. The average energy absorption $H$ in principle has two contributions, one associated with the energy gain in the CF and second, energy gain due to the axial electric field, $H_{CF}$ and $H_z$. The addition of the latter term, would not be necessary if the plasma is axially uniform, which can be the case depending on the conditions. We will come back later (in section 8.7) to the influence of this term. In this section we will consider that the electrons predominantly gain energy in the CF and quickly
dissipate lose their energy after leaving the CF by collisions in the negative glow, so that the energy balance is \( H_{CF} = W \).

The electron energy losses can be conveniently represented by the energy per electron-ion pair \( W \), which is a rather constant parameter, typically a few times the ionization potential, determined mainly by the discharge gas and weakly dependent on the electron energy (for the energy range of interest here). This parameter \( W \) is often used and given in the literature for high-energy electron beams [16] but it can be defined for gas discharges in general [17, 18]. For our HCD, the total energy per electron-ion pair consists of different contributions due to collisions and transport [17]:

\[
W = \frac{\text{lost power}}{\text{ionization rate}} = \sum_r \varepsilon_r n_r k_r + W_{tr},
\]

(8.8)

where \( \varepsilon_r \) is the mean energy lost in a collision, \( k_r \) the rate coefficient, \( n_r \) the fractional density of the target species, while the sums run over all collision processes \( r \) and over the ionization processes \( i \) only. The last term \( W_{tr} \) is the energy loss associated with electron transport to the anode. From the numerical simulation results, for the geometry given in figure 8.2, the two terms of equation (8.8) can be estimated to be approximately 37 eV and 13 eV, adding up to \( W \approx 50 \text{ eV} \).

The electric energy absorbed in the CF by the electrons can be calculated from the flux \( \Gamma_e(r) \) and the \( E \)-field as given by (8.7). The variation of the electron flux with the distance to the cathode has been derived in the previous section and is given by equation (8.2). In the present coordinate system it reads

\[
\Gamma_e(r) = -\gamma \Gamma_i(R) \exp(\alpha (R - r)).
\]

(8.9)

Then the absorbed energy on average per ion impacting the cathode is:

\[
H_{CF} = -\frac{e}{\Gamma_i} \int_{R-r}^{R} \Gamma_e(r) E(r) dr = \frac{2}{(\alpha d)^2} \exp(\alpha d) - 1 - \alpha d \gamma eV \equiv f(\alpha d) \gamma eV.
\]

(8.10)

This absorbed energy depends on the electron multiplication because also new electrons created in the CF absorb energy, leading to “multiplication” of the absorbed energy. So the thicker the CF, the higher the absorbed energy per electron-ion pair. The lower limit for the absorbed energy is \( \gamma eV \), corresponding to the case \( d = 0 \) when the emitted secondary electrons do not multiply at all in the CF.

Equating the absorbed energy \( H_{CF} \) (equation (8.10)) to the energy lost \( W \) (equation (8.8)), we find:

\[
f(\alpha d) \gamma eV = W.
\]

(8.11)

This energy balance can be seen as a self-sustainment condition for HCDs and controls the CF thickness \( d \), meaning that \( d \) adjusts such that the right amount of energy is absorbed to create the ions necessary to sustain the discharge by secondary emission.
The function $f(\alpha d)$ is a monotonically rising function of $\alpha d$, and it can be observed that $f(0) = 1$. This implies that $V$ cannot exceed a maximum voltage, which is given by:

$$V_{\text{max}} = \frac{W}{e\gamma}. \quad (8.12)$$

This maximum voltage (8.12) corresponds to the point, where the energy balance is satisfied without electron multiplication in the CF. Beyond this voltage the absorbed energy always exceeds the energy loss, regardless of $d$, so the energy balance cannot be satisfied. Substituting some typical values $\gamma = 0.1$ and $W = 50\,\text{eV}$ from table (8.1), we find a maximum voltage of $500\,\text{V}$.

The function $f(\alpha d)$ can be well approximated by Taylor expansion of its logarithm, $\ln(f(\alpha d))$ around zero, so that $d$ can be solved. Together with equation (8.12) this yields:

$$d \approx \frac{6}{\alpha} \left( \sqrt{1 + \ln \left( \frac{V_{\text{max}}}{V} \right)} - 1 \right) \quad (8.13a)$$

$$d \approx \frac{3}{\alpha} \left( \frac{V_{\text{max}}}{V} - 1 \right). \quad (8.13b)$$

The first approximation, equation (8.13a), is accurate to within a few percent over the entire range of $V$. The second simpler approximation (equation (8.13b)), also obtained by R. Arslanbekov et al [5], is reasonably accurate for $\alpha d < 0.3 - 0.4$, but in practice $\alpha d$ is often found to be in this range [5]. Both approximations in (8.13), together with the exact solution of (8.11) are shown in figure 8.4.

![Figure 8.4: Relation between the CF voltage and the CF thickness $d$ as given by the energy balance: exact solution of equation (8.11), first approximation equation given by (8.13a) and second approximation given by equation (8.13b).](image-url)
Summarising, the CF thickness in HCD can be smaller than the conventional DC discharge, since the ions created in the plasma are all transported to the cathode and cause secondary emission. Thus electron multiplication in the CF is not directly necessary to sustain the discharge. However, it is indirectly necessary to satisfy the energy balance: the absorbed energy increases as a function of the electron multiplication in the CF. The CF thickness adjusts such that the right amount of energy is absorbed to create the necessary number of ions to sustain the discharge by secondary emission of electrons. The cathode fall thickness $d$ found from equation (8.13a) will be used in the equations describing the ion transport in the CF. From this the electron density and electron temperature will be determined.

### 8.5 Radial transport

#### Cathode fall region

To estimate the radial ion flux $\Gamma_i$ in the CF we can use $\Gamma_i = n_i u_i$, where $n_i$ is the ion density and $u_i$ the radial mean ion velocity in the cathode fall.

The ion density in the cathode fall $n_i$ can be found from Poisson equation:

$$-\epsilon \frac{\partial^2 U}{\partial r^2} = \rho = q_i n_i - e n_e.$$

Assuming that $n_e \ll n_i$ and substitution of equation (8.6) yields:

$$q_i n_i = \epsilon \frac{2V}{d^2}.$$

(8.14)

The ion velocity $u_i$ varies across the CF; for collisional noble gas ions in high field it can be assumed to be of the form $\beta \sqrt{E}$ [15]. For $r = R - d/2$ (in the centre of the cathode fall) we have $E = V/d$, resulting from equation (8.7). On the other hand, if the ion mean free path is larger than $d$ so that the ion velocity is determined by inertia rather than collisions, the ion velocity is $u_i(r) = \sqrt{-2q_i U(r)/m_i}$, where $m_i$ is the ion mass. For $r = R - d/2$, the ion velocity in this case results $u_i = \sqrt{q_i V/2m_i}$.

Then, combining the expressions for the ion velocity $u_i$ with the ion density (equation (8.14)) we find the ion flux to the cathode:

$$\Gamma_i = n_i \beta \sqrt{V/d} = \frac{2\epsilon \beta V^{3/2}}{q_i d^{5/2}}$$

(8.15)

or

$$\Gamma_i = n_i \sqrt{q_i V/2m_i} = \epsilon \sqrt{2 \frac{V^{3/2}}{q_i m_i d^2}}.$$

(8.16)

When substituting equation (8.13a) into equations (8.15) and (8.16), we can plot the dependence of the ion flux on $V/V_{max}$ for both cases. This is presented in figure 8.5.
Analytical model

Figure 8.5: Ion current density at the cathode as a function of the voltage obtained by substitution of equation (8.13a) into equations (8.15) and (8.16), using the parameters in table 8.1 and the Ar ion mass.

for the parameters in table 8.1. Figure 8.5 shows that the ion cathode flux $\Gamma_i$ increases rapidly as $V$ approaches the maximum voltage $V_{max}$ and in that case we see from figure 8.4 that the CF thickness approaches zero. This is an important advantage of HCDs: since the self-sustainment condition allows a very thin CF, the current density can become much higher than in standard DC discharges. In fact, HCDs are usually operated at high current densities close to the maximum voltage $V \approx V_{max}$; to within 10\% or so. This implies that $\alpha d < 0.3 - 0.4$, so the approximation of equation (8.13b) is indeed reasonable. Consequently, most ionization happens in the plasma rather than the CF, so that the ion flux is very nearly constant across the CF: $\Gamma_i(0) \approx \Gamma_i(d)$ as can be seen from equation (8.3). In the following we neglect that some ions are created in the CF and assume that the ion flux $\Gamma_i$ is totally created in the plasma.

Plasma region

Given the ion creation in the plasma, the plasma density $n$ is determined by the ion transport losses to the CF. Following the standard elementary theory for DC positive column and many other discharges [15, 18], we assume that the electrons in the plasma have a uniform temperature $T_e$ that controls the ionization frequency $\nu_i$. The ions are transported by the ambipolar electric field $E_{amb} \approx -\frac{k_B T_e}{e} \nabla n/n$, where $k_B$ is the Boltzmann constant. Neglecting axial ion transport, the ion transport equation is:

$$
\frac{1}{r} \frac{\partial}{\partial r} \left( r \mu_i E_{amb} n(r) \right) = -\mu_i k_B T_e \frac{1}{e} \frac{\partial}{\partial r} \left( r \frac{\partial n}{\partial r} \right) = \nu_i(T_e) n(r),
$$

(8.17)
where $\mu_i$ is the ion mobility. The bounded solution is a Bessel function:

$$n(r) = n_0 J_0(k_r r), \quad k_r = \sqrt{\frac{e \nu_i}{\mu_i k_B T_e}}.$$  \hfill (8.18)

Neglecting the CF thickness ($d \ll R$) and the ion density in the CF, we impose the boundary condition that the ion density is zero at the cathode, $J_0(k_r R) = 0$. Then $k_r R = x_0$, where $x_0 = 2.405 \ldots$ is the first zero of $J_0(x)$. This yields an implicit equation for the electron temperature:

$$\nu_i(T_e) = \frac{k_B T_e}{e} \left( \frac{x_0}{R} \right)^2.$$  \hfill (8.19)

Since $\nu_i(T_e)$ is a very steep function, $T_e$ is determined quite precisely by this equation and not very sensitive to changes in the gas density or radius $R$.

The radially averaged plasma density is defined as:

$$\bar{n} = \frac{\int_0^R n(r) r dr}{\int_0^R r dr} = n_0 \frac{2 J_1(x_0)}{x_0} = n_0 \times 0.4318 \ldots.$$  \hfill (8.20)

These steps have been worked out in more detail in appendix 8.A.

The amplitude of the plasma density profile can be found by equating the total ionization rate ($\nu_i \bar{n}$) integrated over the volume to the ion flux $\Gamma_i$ at the cathode integrated over the surface area:

$$\nu_i \pi R^2 \bar{n} = 2 \pi R \Gamma_i,$$

$$\bar{n} = \frac{2}{R \nu_i} \Gamma_i.$$  \hfill (8.21)

(8.22)

The last equation indeed is the same as derived in the previous quas-analytical model (equation (7.5) in chapter 7): $\Gamma_i = \frac{1}{2} R \langle S \rangle$, where the source is used as $\langle S \rangle = \bar{n} \nu_i$. Then, using equation (8.19) for $\nu_i$ and substitution in equation (8.22) we find the radially averaged plasma density:

$$\bar{n} = \frac{2 R}{x_0^2 \mu_i k_B T_e / e} \Gamma_i.$$  \hfill (8.23)

This can be directly combined with equation (8.15)/(8.16) to find the plasma density from the CF voltage.

### 8.6 Axial transport

While the ions are lost by radial transport to the cathode, the electrons are lost by transport in axial direction towards the anode. This requires an axial electric field $E_z$ in the plasma, corresponding to a decrease of the voltage as a function of the axial distance $z$ from the
analyse. This in turn leads to a decrease of the mean plasma density as a function of $z$ (see equations (8.23) with (8.15)/(8.16)). In this section we derive the axial profiles $V(z), \bar{n}(z), \Gamma_i(z)$, etc, from the equations for the axial electron transport.

The axial electron flux results from both drift due to the electric field and diffusion due to the density gradient. Integrated over the cross section of the plasma, the electron continuity equation with the drift-diffusion flux gives:

$$\pi R^2 \frac{\partial}{\partial z} \left( \mu_e E_z \bar{n} + \mu_e \frac{k_B T_e}{e} \frac{\partial \bar{n}}{\partial z} \right) = 2\pi R (1 + \gamma) \Gamma_i,$$

where $\mu_e$ is the electron mobility and the Einstein relation $D_e = -\mu_e k_B T_e / e$ has been used. This equation is the differential form of equation (7.6) as derived in section 7.

Note that in the plasma, neglecting the CF thickness, $E_z$ is nearly constant in the radial direction, $\frac{\partial E_z}{\partial r} = 0$, as can be seen from the modelling results in figure 8.2. We assume that $\mu_e$ and $T_e$ are independent of $z$ and substitute the flux density from equation (8.23):

$$\frac{\partial}{\partial z} \left( \frac{e E_z}{k_B T_e} \frac{\partial}{\partial z} \bar{n} + \frac{\partial \bar{n}}{\partial z} \right) - (1 + \gamma) \left( \frac{x_0}{R} \right)^2 \frac{\mu_i}{\mu_e} \bar{n} = 0.$$  

(8.25)

To calculate the axial profiles, we could now substitute $E_z = -\frac{\partial V}{\partial z}$ and $\Gamma_i(V)$ from the previous section and then try to solve for $V(z)$, but this is cumbersome. Rather, we linearise equation (8.25) by assuming that $E_z$ is uniform, in agreement with the numerical simulation results in figure 8.2. As a boundary condition we impose that $\bar{n}$ is zero at the anode at $z = 0$, since the plasma cannot be maintained inside the anode due to the absence of a CF. The numerical results in figure 8.2 show that the plasma density drops indeed to zero within a small distance inside the anode. In the center of the cathode at $z = L$ we impose the boundary condition that the axial electron flux is zero:

$$\frac{e E_z}{k_B T_e} \bar{n}(L) + \left[ \frac{\partial \bar{n}}{\partial z} \right]_{z=L} = 0.$$  

(8.26)

The appropriate solution of equation (8.25) is (see appendix 8.B for details):

$$\bar{n}(z) = \bar{n}_0 \exp(-k z) \sin(K z),$$  

(8.27)

where

$$k = \frac{e E_z}{2 k_B T_e},$$  

(8.28)

$$k_0 = \sqrt{1 + \gamma \frac{x_0}{R} \sqrt{\frac{\mu_i}{|\mu_e|}}},$$  

(8.29)

$$K = \sqrt{k_0^2 - k^2}.$$  

(8.30)

The zero-flux boundary condition (8.26) requires that:

$$\tan K L = -\frac{K}{k}.$$  

(8.31)
From this equation we can find explicit equations for \( k/k_0 \) and \( K/k_0 \) (see appendix 8.B for details). In a good approximation (within 1% over the whole range \( k_0 L > \pi/2 \)), the solution is given by Taylor expansions around \( \pi \):

\[
k \approx k_0 \left[ 1 - \frac{\pi^2}{8} \left( \sqrt{\frac{4}{k_0 L}} + 1 - 1 \right)^2 \right].
\]

(8.32)

The exact solution (8.31) and the approximate solution (8.32) are shown in figure 8.6. The figure shows that the longer the cathode, the larger the decay constant \( k \); at small value of \( L \), \( k \) is close to zero and the exponential part of equation (8.27) is close to 1. \( K \) is the parameter controlling the sine part of the solution; this dominates the shape of the profile at short cathode length.

Figure 8.6: Solution of equation (8.31). Solid line: exact solution. Dashed line: the approximate solution (8.32).

The axial profile (8.27) of the plasma density is presented in figure 8.7 using the approximation (8.32).

The axial electric field is determined by equation (8.28):

\[
E_z = \frac{2 k_B T_e}{e} k,
\]

(8.33)
i.e. the field adjusts to satisfy the boundary conditions. Note that the value of \( E_z \) selected by our boundary conditions is also the smallest \( E_z \)-value allowed by the general solution of equation (8.25), corresponding to the most uniform \( \bar{n}(z) \). Equations (8.32) and (8.33) show that the axial electric field depends on the cathode length. Figure 8.8 shows a comparison, for different cathode lengths, between the on-axis potential in the numerical simulations and the analytical potential \(-E_z z\).
The field is weaker as the cathode is shorter, decreasing from $2k_0T_e$ for very long cathodes, to zero for $k_0L = \pi/2$. For still shorter cathodes the electric field is reversed:

$$L < \frac{\pi}{2k_0} \Rightarrow E_z < 0. \quad (8.34)$$

Note that our assumption of a uniform $E_z$ is no longer reasonable in this case, so the axial profiles should not be taken literally, but the condition (8.34) for field reversion is physically meaningful and agrees with our numerical simulation results [13] (chapter 6).

### 8.7 Influence of the $E_z$-field on the self-sustainment

To normalise the profile $\Gamma_i(z)$ and calculate the discharge current, we use equation (8.15) obtained from Poisson equation, in combination with the CF thickness $d$ from the energy balance (equation (8.13)). In order to do this rigorously, we should establish the energy balance integrated (or averaged) over the axial position $z$, since the energy is not necessarily lost at the same position where it has been absorbed, but nearly the same results can be obtained in a much simpler way by applying the energy balance locally at the position $z = z_m$, where $\Gamma_i$ is highest. This peak occurs at some distance inside the cathode (for long enough cathodes with $k_0L > \pi/2$) and is given by

$$z_m = \frac{\pi}{K} - L \quad (8.35)$$

$$\Gamma_m = \Gamma_i(z_m) = \Gamma_0 K \frac{k_0}{k_0} \exp(-k z_m), \quad (8.36)$$

so the profile (8.27) can be written as

$$\Gamma_i(z) = \Gamma_m \frac{k_0}{K} \exp(-k(z - z_m)) \sin(Kz). \quad (8.37)$$
We now estimate $\Gamma_m$ by substitution of equation (8.13) in equation (8.15)/(8.16), as shown in figure 8.5. The CF voltage $V$ at $z = z_m$ differs from the applied discharge voltage by the anode sheath voltage and by the axial voltage drop in the plasma $E_z z_m < 2 k_B T_e / e$ but in total this voltage difference so small that it can be reasonably neglected. We simply use the applied voltage for $V$ in equation (8.13).

However, the energy balance from section 8.3 requires an important modification in order to account for the axial electric field $E_z$. The electrons can absorb energy from this field, in addition to the energy $H_{CF}$ absorbed in the CF. Although the axial field is relatively weak, it can have a significant influence because it acts upon all electrons in the plasma, whereas the CF field acts only upon part of the electrons in a small part of the volume. Per electron-ion pair, the energy absorbed from the axial electric field is:

$$H_z = -e\frac{\pi R^2}{2\pi R \Gamma_i} \left( \mu_e E_z \bar{n} + \mu_e \frac{k_B T_e}{e} \frac{\partial \bar{n}}{\partial z} \right) E_z$$  \hspace{1cm} (8.38)

$$= (1 + \gamma) 4 k_B T_e \frac{k^2}{k_0^2} \left( 1 + \frac{1}{2k \Gamma_i} \frac{\partial \Gamma_i}{\partial z} \right).$$  \hspace{1cm} (8.39)

The last factor in brackets, accounting for electron diffusion, is negligible near the point $z = z_m$ or on average over the cathode length. When including the additional absorbed energy in the energy balance, $H_{CF} + H_z = W$, it compensates for some of the energy losses, hence it leads to a decrease of the maximum voltage in equation (8.13) and further. For the point $z = z_m$ we find

$$V_{max} = \frac{1}{\gamma} (W - H_z) = \frac{1}{\gamma} \left( W - (1 + \gamma) 4 k_B T_e \frac{k^2}{k_0^2} \right)$$  \hspace{1cm} (8.40)

rather than equation (8.12). Substituting the ratio $k/k_0$ from equation (8.32) and figure 8.8,
we see the maximum voltage now depends on the cathode length, decreasing monotonically by approximately $4/\gamma \left( \frac{k_B T_e}{q} \right)$ as the cathode length is increased from very short to infinity.

The total discharge current $I$ is directly related to $\Gamma_m$ as

$$I = 2\pi R (1 + \gamma) e \int_0^L \Gamma_i(z) dz = 2\pi RL (1 + \gamma) e^{\frac{\exp(k z_m)}{k_0 L}} \Gamma_m.$$  \hspace{1cm} (8.41)

Calculating this for different voltages $V$, we find the voltage-current characteristic $V(I)$, which is shown in figure 8.9 for different cathode lengths and compared with numerical simulation results.

![Figure 8.9: Voltage-current characteristics for different cathode lengths from analytical model (8.9(a)) and form the numerical model (8.9(b)).](image)

The V-I curves for different cathode lengths are shifted in voltage due to different energy absorption $H_z$ from the axial field, following the trend of the maximum voltage, equation (8.40). The shifts between the analytical curves are somewhat larger than between the numerical curves, which is consistent with the axial potential profiles in figure 8.8, which vary more in the analytical model than in the numerical simulations. Similar voltage shifts are observed in experimental V-I curves, as shown in chapter 2.

It is interesting to remark that most of the results in this and the previous section actually do not depend directly on the cathode length $l$, but rather on the parameter

$$k_0 L = x_0 \sqrt{1 + \gamma} \sqrt{\frac{\mu_i}{|\mu_e|} \frac{L}{R}}$$  \hspace{1cm} (8.42)

i.e. on the aspect ratio length/radius, so decreasing the radius has the same effect as increasing the length.
8.8 Conclusions

The analytical model presented in this paper gives the following insights in the operation of longitudinal HCDs:

- Because the ions created in the plasma are all collected by the cathode and cause secondary electron emission, electron multiplication in the CF is not necessary directly to sustain the discharge. This allows the CF thickness to be smaller than that in a conventional DC discharge, as given by equation (8.5).

- The multiplication in the CF is indirectly necessary to satisfy the energy balance (equation (8.11)): the absorbed energy increases as a function of the electron multiplication in the CF. The CF thickness depending on the voltage, adjusts such that the right amount of energy is absorbed to create the ions necessary to sustain the discharge by secondary emission. The size of the CF thickness is then found by equation (8.13).

- There is a maximum CF voltage (equation (8.12)) beyond which the absorbed energy always exceeds the lost energy so that the energy balance cannot be satisfied. When increasing the voltage up to this maximum voltage, the CF thickness decreases down to zero, as shown in figure 8.4. According to Poisson equation, the ion density and current density in the CF then increase to infinity (figure 8.5). The typical operation of the HCD is close to this maximum voltage, with a thin CF where almost no electron multiplication takes place.

- The longitudinal HCD develops an axial non-uniformity and an axial electric field in order to generate an axial electron current to the anode. The axial electric field necessary to achieve this is stronger as the cathode is longer, or more precisely, as the ratio cathode length/cathode radius is larger. For very short cathodes the axial electric field is reversed to compensate the electron diffusion losses to the anode.

- Additional energy absorption from the axial electric field in the plasma modifies the energy balance and hence modifies the CF thickness. This leads to a shift in the maximum voltage, which depends on cathode length and diameter. This resulting in a voltage-shift of the voltage-current characteristic. This is also observed in numerical simulations and experiments (chapter 7).

References

Analytical model

Chapter 8.

8.A Plasma density

In this appendix we work out the details following equation (8.17) in the text, which result in expression (8.23) for the cross-section averaged electron density profile.

We start from equation (8.17),

\[
\frac{1}{r} \frac{\partial}{\partial r} \left( r \mu_i E_{amb} n(r) \right) = -\mu_i \frac{k_B T_e}{e} \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial n}{\partial r} \right) = \nu_i(T_e) n(r)
\]

The bounded solution is the Bessel function \( J_0(k_r r) \),

\[
n(r) = n_0 J_0(k_r r), \quad k_r = \sqrt{\frac{e \nu_i}{\mu_i k_B T_e}}
\]

As boundary condition we impose that \( n(R) = 0 \), or

\[
J_0(k_r R) = 0 \Rightarrow k_r R = x_0,
\]

where \( x_0 \approx 2.405 \) is the first zero of the bessel function \( J_0(x) \). Then we find that

\[
k_r = \sqrt{\frac{e \nu_i}{\mu_i k_B T_e}} = \frac{x_0}{R} \Rightarrow \nu_i(T_e) = \mu_i \frac{k_B T_e}{e} \left( \frac{x_0}{R} \right)^2.
\]

Then the density is given by

\[
\bar{n} = \frac{\int_0^R n(r) r \, dr}{\int_0^R r^2 \, dr} = \frac{\int_0^R n(r) r \, dr}{\frac{R^2}{2}}
\]

\[
= \frac{2}{R^2} \int_0^R n_0 J_0(k_r r) r \, dr
\]

\[
= \frac{2n_0}{R^2 k^2} \int_0^{kR} J_0(x) x \, dx = \frac{2n_0}{x_0^2} \int_0^{x_0} J_0(x) x \, dx.
\]

Using \( x J_0(x) = \frac{d}{dx} x J_1(x) \) [19, equation 9.1.30] we find that

\[
\bar{n} = \frac{2n_0}{x_0^2} x J_1(x) \bigg|_{x_0}^{x_0} = n_0 \frac{2J_1(x_0)}{x_0} = n_0 \times 0.4318 \ldots,
\]

where we have used \( J_1(x_0) = 0.52 \ldots \)

8.B Plasma density profile

We consider equation (8.25):

\[
\frac{\partial}{\partial z} \left( \frac{eE_z}{k_B T_e} \bar{n} + \frac{\partial \bar{n}}{\partial z} \right) - \left( 1 + \gamma \right) \left( \frac{x_0}{R} \right)^2 \frac{\mu_i}{\mu_e} \bar{n} = 0
\]
This equation can be written in the form
\[ n'' - 2k' n' + k_0^2 n = 0, \]
where the coefficients \( k \) and \( k_0 \) are given by
\[
\begin{align*}
    k &= -\frac{eE_z}{2k_B T_e}, \\
    k_0^2 &= (1 + \gamma) \left( \frac{x_0}{R} \right)^2 \frac{\mu_i}{\mu_e}.
\end{align*}
\] (8.B.1)
This is an ordinary differential equation with constant coefficients. The solutions are of the form \( \exp(sz) \), the coefficients \( s_1, s_2 \) are the solutions of the characteristic equation:
\[ s^2 - 2k s + k_0^2 = 0 \]
and are given by
\[ s_{1,2} = k \pm \sqrt{k^2 - k_0^2} = k \pm \sqrt{D}. \]
The only possible solution that satisfy the boundary conditions is if \( D < 0 \), so that \( -k_0 < k < k_0 \). We can write \( s_{1,2} = k \pm i\sqrt{-D} \equiv k \pm iK \), where \( K = \sqrt{k_0^2 - k^2} \). In that case the solution is given by:
\[
n(z) = C_1 \exp(kz) \sin(Kz) + C_2 \exp(kz) \cos(Kz). \quad \text{(8.B.2)}
\]
From the boundary condition \( n(0) = 0 \), we conclude that \( C_2 = 0 \). The solution 8.B.2 casts:
\[
n(z) = n_0 \exp(-kz) \sin Kz \quad \text{(8.B.3)}
\]
on the interval \( z \in [0, L] \).
The zero-flux boundary condition (8.26) at \( z = L \) yields:
\[
2k \exp(-kL) \sin(KL) + K \exp(-kL) \cos(KL) - k \exp(-kL) \sin(KL) = 0
\]
\[
\begin{align*}
    k \sin(KL) &= -K \cos(KL) \\
    \tan KL &= -\frac{K}{k}. \quad \text{(8.B.4)}
\end{align*}
\]
We substitute the expression for \( K \) and rewrite equation (8.B.4) as
\[
\tan \left( k_0 L \frac{K}{k_0} \right) = -\frac{k_0 K}{k k_0} \Leftrightarrow \tan \left( k_0 L \sqrt{1 - \left( \frac{k}{k_0} \right)^2} \right) = -\frac{k_0}{k} \sqrt{1 - \left( \frac{k}{k_0} \right)^2}
\]
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We introduce the auxiliary quantities \( x = k_0 L \) and \( y = k / k_0 \); substitution yields

\[
\tan \left( x \sqrt{1 - y^2} \right) = -\frac{\sqrt{1 - y^2}}{y}.
\]  

(8.B.5)

Since \( y \) is in the range \((-1, 1)\) we can define \( y = \cos \phi \), with \( \phi \in (0, \pi) \). Substitution in the previous equation yields (the tan function is odd):

\[
\tan (x \sin \phi) = -\tan \phi = \tan(-\phi).
\]

Since \( \tan \alpha = \tan \beta \Leftrightarrow \alpha = N\pi + \beta \), where \( N \) is an integer value, the solutions are given by

\[
x \sin \phi = N\pi - \phi \Leftrightarrow x = \frac{N\pi - \phi}{\sin \phi} = \frac{N\pi - \arccos y}{\sqrt{1 - y^2}}.
\]  

(8.B.6)

We also find that \( KL = k_0 L (K/k_0) = x \sqrt{1 - y^2} = x \sin \phi \), or

\[
KL = N\pi - \phi.
\]

Since \( \phi \in (0, \pi) \) we have \((N - 1)\pi < KL < N\pi\). Since \( n(z) \), as given by equation (8.B.3), must be positive we require that \( 0 < KL < \pi \) (otherwise \( \sin Kz \) takes negative values for \( x \leq L \)). Then we conclude that the only acceptable solution is the one with \( N = 1 \) and the final results are

\[
x = \frac{\pi - \phi}{\sin \phi} = \frac{\pi - \arccos y}{\sqrt{1 - y^2}},
\]

(8.B.7)

\[
KL = \pi - \phi = \pi - \arccos y.
\]

(8.B.8)

Relation (8.B.7) provides an explicit expression for \( x = k_0 L \) as a function of \( y = k / k_0 \). An analytical expression for \( y(x) \) does not exist, but this graph is shown in figure 8.6.
Recent developments
Chapter 9.

9.1 Novel design of HCD laser

Based on the results of the studies on the longitudinal sputtering HCD Cu ion laser we have been able to define the optimum parameters and to determine the best configuration for efficient laser operation. However, there are still some remaining issues.

The first one is the disadvantage that the discharge current and the metal vapour pressure cannot be controlled independently. The second issue is the lack of long term stability; due to the sputtering process and re-deposition of metal the cathode shape will change during operation. This reduces the lifetime of the laser tube.

Based on the observations and results, reported in this thesis we suggest a new version of the hollow cathode discharge, the so-called multiple hollow cathode discharge [1]. It is aimed to tackle the issues mentioned above.

One of the options of the novel design [1] is schematically presented in figure 9.1 showing a combination of one longitudinal and several transversal discharges. The main discharge is composed by the active laser volume as determined by a main cylindrical cathode \( C \) made of the desired refractory metal (Cu, Ag, Ag, etc.) terminated at both sides by two ring anodes \( A_i \). This axial component is the longitudinal part, meaning that the structure is parallel to the laser light. Perpendicular to the longitudinal part we make a number of short radial cathode holes (CHs) arranged along the length of the main cathode. These CHs bridge the gap from the central discharge to auxiliary anodes \( A_i \) that are located outside.

Thus, the discharge is a superposition of two independent discharges: 1) a longitudinal discharge formed by the cathode \( C \) and two ring anodes \( A_0 \), and 2) a flute-type discharge(s) formed by the cathode \( C \) and the auxiliary anodes \( A_i \). An important aspect of this design

\[ \text{Figure 9.1: Experimental discharge tube design. Main cathode cylinder } C \text{ with an inner diameter 5 mm, outer diameter 20 mm and length 100 mm; three cathode holes with an inner diameter 3 mm and length 7.5 mm. Rode anodes } A_i \text{ are placed above each CH.} \]
is that the discharges that occur in the cathodes holes can be used as additional sources of metal atoms, obtained by sputtering.

Moreover the two discharge components can function independently. Inside the main cathode the optimal conditions for laser oscillation are realised (pressure and current density), while in the side CHs the current density is higher, thus facilitating the production of metal vapours. Hence the CHs can also be regarded as additional controllable reservoirs of metal atoms. So that, by varying the potential between the cathode and the auxiliary anodes, it is possible to control the metal atom density and the excitation of various species in the laser active volume independently.

Preliminary experiments show a considerable increase of laser power due to the additional copper atom density, injected from the cathode holes. When only the longitudinal discharge is run between $C$ and the two anodes $A_0$ no lasing is obtained. By switching on one of the anodes ($A_2$) the lasing starts and the laser power increases with the number of the anodes switched on.

The new design resembles the longitudinal segmented hollow cathode design. So the insights obtained with the study on that discharge type can be used in the new design. For instance by choosing a proper distance between the CHs, it is possible to achieve a nearly homogeneous plasma distribution and realize a uniform sputtering in the main cathode. This will prolong the life time of the laser tube. It is also possible to make the main cathode $C$ from a metal with negligible sputtering and load the metal atoms only from the side cathodes. Moreover, metal sleeves of different metals can be inserted into the side cathodes, which gives the possibility for simultaneous laser oscillation on different metals in the same active volume.

In order to simulate the discharge in the main cathode we have chosen the geometry as shown in figure 9.2. The axis of symmetry in cylindrical coordinates coincide with the

![Figure 9.2](image.png)

**Figure 9.2:** Computational domain. The axes of symmetry coincide with the optical axis of the laser.
optical axis of the laser. In this case, the cathode hole is not really a hole, but rather a ring shaped slit in the main cathode. The distance between the holes is 40 mm, but due to symmetry in the discharge we only simulate half the distance between the hole’s centres (i.e. 20 mm). The gas mixture is He with 5% of Ar and the total gas pressure is set to 1.3 kPa.

The flexibility of the model allows the sputtering process to be applied on different metallic boundaries. Simulation are made first when the main cathode sputters, but no sputtering is applied on the cathode holes. The distribution of the potential and the densities of the Cu atom, Cu ion and the electrons are shown in figure 9.3.

Inside the entire CH the potential remains close to that of the anode. Even on the axis at z = 20 mm we find a potential that differs only few volt from the anode potential. This implies that this region on the central axes plays the role of an anode for the main cathode.
This means that the geometry is practically similar to the segmented construction and that the insights obtained in the previous chapters can be applied for the novel design.

It can be seen that similarly to the segmented construction the maximum in plasma density is at a distance of 5 mm from the anode region and that in the optimal configuration the length should be about 5 times larger than the diameter. So that the side cathodes should be at distance of around 25 mm for obtaining uniform plasma inside the main cathode.

The novel design can also be realised by connecting the auxiliary anodes together in one coaxial anode as shown in figure 9.4 [1]. The advantage of this variant is that in this way the number of electrodes is less and this technically simplifies the construction of the laser tube.

The work continues with simulations of the same geometry when also the side cathodes are sputtered and investigations of the main features and properties of the novel design.

Figure 9.4: The novel discharge tube design
9.2 MD2D developments

The version of MD2D that has been used for the simulations presented in this thesis has proven to be capable of simulating hollow cathode discharges. Yet, it has a number of shortcomings:

- it is not capable of calculating buffer gas heating;
- it does not allow grid refinement in the regions where large gradients are expected;
- simulations are restricted to two-dimensional geometries; this means that, for example, the new discharge construction (section 9.1) can not be modelled accurately.
- Calculations are restricted to rectangular domains.

The author is involved in a project that aims to develop of a new version of MD2D that overcomes these problems. The first sub-project aims at allowing the usage of ortho-curvilinear coordinate systems in one-, two- and three-dimensional systems. This task has recently been completed and the code is being extensively tested at the moment of writing.

Figure 9.5 shows two examples of two-dimensional *ortho-curvilinear coordinate systems* that have been generated by this code. The usage of curvilinear coordinate systems (figure 9.5(a)) allows simulations on domains with curved boundary shapes, while the *grid stretching* (figure 9.5(b)) allows a higher grid line density in regions where large gradients are to be expected, like in the cathode fall region. Both techniques can be combined. For details on PLASIMO’s grid generation capabilities, we refer to Ref. [2].

The result of this project is that MD2D uses the same internal representation of meshes as the rest of PLASIMO. This is a prerequisite for the realisation of the next phases of the projects, in which PLASIMO’s modules for (gas) temperature and buffer gas flow calculations will be made available for MD2D-simulations. For a description of these modules, we again refer to Ref. [2].
9.3 Kinetic Monte-Carlo development

The results of the numerical simulations presented in this thesis have been obtained with the MD2D model. They were found to agree well with the experimental and analytical observations on hollow cathode discharges. However, not all discharge phenomena can be simulated with a pure fluid model:

- In the cathode fall region, the positive ions are accelerated towards the cathode in the strong electric field. In the absence of sufficient collisions, the ion energy will, apart from the reduced field, also be determined by the ion inertia. (see section 8.5, equation 8.16).

- The sputtering yield depends mainly on the energy of the ion impinging the surface (section 1.6, figure 1.5). Since the MD2D model does not calculate the ion energy distribution function, we have calculated the sputtering yields on the basis of assumed constant values for the ion energies (section 5.3). In reality the energy is determined by the dynamics in the cathode fall region, as we have just discussed.

- Electrons in the CF of HCDs may be accelerated to such an extent that their mean free path exceeds the plasma size. As a result they make multiple passes through the plasma with high velocities and are reflected by the CF on the opposite side. This electrostatic trapping is known as the pendulum effect and has been studied by [3, 4, 5]. Some authors even believe that this effect is primarily responsible for the hollow cathode effect.

In order to investigate these issues, or to test their influence on the simulation results, a kinetic plasma description is required, for example based on a Monte Carlo model. An Monte-Carlo PLASIMO sub-module has been previously developed to describe the electrons in a drift-diffusive plasma [6].

This module can be used in two ways. Firstly, it can be employed in a stand-alone application, where the particles move and collide in an environment with prescribed properties. This can be used for example to simulate drift tube experiments. Secondly, it can be used in hybrid simulations. In this case, some aspects of the system under study are described with a fluid model, whereas others are delegated to the Monte Carlo module.

In order to use the Monte Carlo code for the simulation of HCDs, various modifications and extensions are necessary. Some of these have been implemented in the framework of the present project:

- The code can be used to follow other species than electrons;

- A cylindrical vessel geometry has been implemented;

- Wall processes (sputtering, deposition, secondary electron emission) have been implemented;

- The stand-alone Monte Carlo code can be run with non-uniform electric fields [7].
In the previous hybrid studies, the Monte Carlo code was used to track the fast electrons inside the entire plasma region. Spatial hybridisation, in which the Monte Carlo code is used only in part of the simulation domain, is not supported. This would be useful for tracking the ions (only) in the cathode fall.

References


General conclusions
The aim of this work was to study sputtering hollow cathode discharges. More specifically, this investigation was devoted to the application of HCDs in laser technology with the final goal to optimise the operating conditions and HC geometry in order to build a reliable and efficient sputtering metal vapour laser.

The presented work includes experimental and theoretical studies of sputtering HCDs for Cu ion lasers. The numerical modelling results were validated with experimental observations. This comparison was guided by an analytical model that was constructed for that purpose. The insights obtained by this validation have led to an improvement of the numerical model that can now successfully be employed as an optimisation tool for HCD lasers in particular, and more in general, for the improvement of various other HCD applications.

- Experiments:
  Several laser tubes were designed, built and studied experimentally. The discharge performance was studied in detail by investigating the axial current distribution, the spontaneous emission distribution and the laser performance and how these features are influenced by the operating parameters such as input current, gas composition and electrode geometry. The impact of the external parameters settings on the laser operation were investigated by monitoring the behaviour of the IR copper ion line (780.8 nm), the transition with the highest gain. In this way the features of the optimal conditions and the best geometry could be determined experimentally.

- Numerical model:
  The PLASIMO modelling platform was used to construct a model that facilitates in-depth studies of the plasma behaviour and that can be used to find the optimum location in parameter space for the operation of these discharge types. To that end the PLASIMO modelling platform has been extended. In particular, an implementation of a wall-sputtering module was added to the time-dependent fluid sub model MD2D. The versatility of the model makes it possible to simulate the plasma processes in different geometries, gas compositions and operation conditions. It is demonstrated that the present model can be used as a tool to optimise the discharge conditions and the hollow-cathode geometry.

- Analytical model:
  An analytical model was constructed with the aim to interpret numerical results. The model is based on the revision and combination of some elementary theories and extends these to account for the axial non-uniformity of longitudinal HCDs. The model is focused on the main plasma properties such as plasma density, potential, current, rather than on plasma chemistry or the behaviour of the metal vapour atoms.
Hereafter we present the most important conclusions on a per-chapter basis.

- Chapter 2:
  Strong axial inhomogeneities of the discharge current, the plasma distribution and the spontaneous emission are experimentally observed. These inhomogeneities are nearly independent on the input current and gas mixture, and are mainly influenced by the electrode configuration and dimensions.
  
  The optimal conditions — input current, pressure and Ar addition — for lasing of Cu 780.8 nm were determined. The existence of these optimal conditions limits the possibilities to increase the laser power by increasing the input power, or by increasing the sputtering efficiency (by increasing pressure or adding heavier atoms). The most effective way to increase the laser power is by increasing the active volume, but the disadvantage of this way is that an axial inhomogeneity may exists, which affects the excitation efficiency and laser performance strongly. Therefore it is advised to make use of a geometry that consists of a sequence of anode-cathode patterns. In such an elementary pattern, or segment, the discharge must be as uniform as possible in terms of plasma density and species density distributions. The dimensions of the elementary segment pattern was determined: we have demonstrated that the most efficient laser oscillation is achieved when the laser active volume comprises a series of anodes and cathodes, each cathode with a length not longer than 20 mm for a cathode diameter of 4 mm.

- Chapter 3:
  A multi-fluid approach to model the plasma created in a hollow cathode is described. The model used is PLASIMO’s fluid sub-model MD2D. The chapter gives a brief overview of the physics behind the model, describes the discretisation aspects of the code, and outlines the extensions and improvements that were done to the code.

- Chapter 4:
  A chemistry analysis was made with the aim to construct a lean and reliable model that can be used as part of a design tool for HCD lasers. Based on the modelling results and estimative calculations, the HCD plasma for the conditions under study is determined to be an ionising EEK plasma and the model is simplified by means of reducing the number of species and reactions. The reduction is justified by comparing the results of simplified chemistry models with those of the more complete one. With the reduced chemistry model it is much easier to explore the parameter space and find the best geometrical construction, anode-cathode pattern, current setting and fill-chemistry.

- Chapter 5:
  The strength of the constructed model is demonstrated by simulations of the sputtering HCD in a He-Ar gas mixture. The obtained quantities are compared with the experimental data. Both experimental and theoretical studies indicate that the addition of heavy gas atoms such as Ar is required to produce efficient sputtering. This
is needed to obtain a Cu atom concentration that is sufficiently high for lasing. Ar is the main ionic species in spite of the fact that it constitutes only a small fraction of the total gas pressure. However the addition of Ar might also increase the axial inhomogeneity and reduce the efficient excitation of the laser levels. An optimum of the Ar concentration exists, which limits the possibility to increase the laser power by increasing the Ar addition.

• Chapter 6:
The influence of the cathode segment length is investigated in order to optimise the HC geometry for obtaining a stable and uniform active medium with high excitation efficiency. The simulation results for different cathode lengths are compared with experimental data. It was found that a lower limit of the cathode length exists, beyond which an inversion of the axial electric field occurs; this marks a transition between the conventional HC and so-called high-voltage operation (at the conditions under study). It was shown that the optimum cathode segment length \( l \) for lasing is at \( l \sim 20 \text{ mm} \), where the plasma tends toward uniformity, but an axial electric field still exists. The above results are confirmed by the measured laser power dependence on the cathode segment length.

• Chapter 7:
In search for the optimal HC geometry, calculations are made for different values of the inner diameter of the cathode while keeping its length and parameter \( pd \) constant. The results from the simulations and quasi analytical calculations predict a link between the diameter and length of the hollow cathode. Accordingly, the diameter should be around 5 times smaller than the length of the cathode at the operation conditions under study. In such a configuration the plasma is more or less uniform while an axial field still exist. If the ratio \( l/d \) is larger than 5, a strong non-uniformity starts to occur, while smaller \( l/d \) values invoke an inversion of the electric field.

• Chapter 8:
Analytical modelling shows that due to the fact that the ions created in a HCD plasma are all collected by the cathode where they cause secondary electron emission, makes that the energy balance can be satisfied with a thinner CF than the classical glow discharge. There is a maximum CF voltage beyond which the energy balance cannot be satisfied. The typical operation of the HCD is close to this maximum voltage, with a thin CF where almost no electron multiplication takes place. In longitudinal HC an axial non-uniformity appears in order to generate an axial electron current to the anodes. The axial electric field necessary to achieve this is stronger as the ratio length/radius is larger. For very short cathodes the axial electric field is reversed to slow down electron diffusion losses to the anode. Additional energy absorption from the axial electric field in the plasma bulk modifies the energy balance and hence the CF thickness. This leads to a shift in the maximum voltage, depending on cathode length, and results in a voltage-shift of the voltage-current characteristics.
• Chapter 9:
  This chapter is devoted to the new challenges and recent developments. Based on the results and observations, reported in this thesis, a new design of the hollow cathode discharge construction was proposed with the aim to overcome the remaining issues. The extension and the improvements in the code are outlined and the plans for future work are presented.
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