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HYBRID MODELLING OF ELEMENTARY PROCESSES OF COLD CATHODE GLOW DISCHARGES

Ph.D. thesis

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1 Introduction

Glow discharges have a wide range of applications, including gas and metal vapor lasers, spectral lamps, analytical analysis, layer deposition, surface treatment. Besides the scientific interest the optimization of discharges also required and still require the investigation of discharges for a better understanding of the processes which take place in the discharges. At the beginnings the role of the processes in the discharge operation were investigated by means of analytical models. The appearance of fast computers led to the appearance and development of numerical models, which made possible the quantitative description of the discharges. The early numerical models discussed separately the discharge regions and it did not give a self-consistent description of the whole discharge. With the recently developed hybrid models [1] each discharge region can be described at the same time. The hybrid models consist of a fluid model [2] and a Monte Carlo model [3]. The fluid model makes possible to describe the motion of charges which are in hydrodynamic equilibrium with the field in the low electric field region (negative glow, positive column). With the Monte Carlo model can be traced the electrons which move in the high electric field present in the cathode vicinity.

In order to get a clear image about the discharges and to understand their operation it is need to know:

- their current-voltage-pressure characteristics, which determine their operation conditions,
- the processes which take place in the discharge and their importance in the discharge operation,
- the sources, losses and density of charged and excited particles present in the discharge, and their role played in different processes.

The aim of the work is the development of self-consistent hybrid models which help in the determination of the discharge characteristics mentioned above. Further on to get a more detailed image about some interesting phenomena observed in some noble gas discharges.

2 Aim of the work

In the thesis three problems are investigated:

1. axial light intensity distributions and electron emission from the cathode surface in abnormal argon glow discharges,

2. development of hollow cathode effect in a plane-parallel hollow cathode discharge,

3. the presence of molecular ions and their role in the self-sustainment of helium discharges.

The background of these problems are presented below.

1. The elementary processes which take place in gas discharges has been investigated for a long period. The processes which are responsible for the self-sustainment of the cold cathode discharges have been in the central of the attention from the beginnings. The first theory for the low current dark discharges and for the low pressure breakdown was developed by Townsend [4]. The latter theories revised this theory by including the space charge effects, and the role of the fast atoms, metastable atoms and photons in the electron production at the cathode and in the gas phase. Recently, it has been established that a large number of processes participate in secondary electron production at the cathode [5], the ions dominate only in a very narrow range of conditions, besides them an important role have the fast atoms, metastable atoms and ultraviolate photons.

   In the last few years significant advances in discharge modelling have been made with the development of hybrid codes, which provide important information about the phenomena taking place in a wide variety of discharges [1,6–8]. The secondary electron emissions coefficient (γ) values are important for modelling because with the knowledge of γ the electrical characteristics of the discharge can be calculated. Hybrid codes, usually assume that every electron is emitted due to the ions, so they use an apparent electron emission coefficient, and besides this the value of γ is taken constant for different discharge conditions. In order to improve the accuracy of these models it was justified to address the question whether application of γ obtained for low-current discharge conditions (i.e. homogeneous field) [5] would yield satisfactory results in the region of the normal (constricted) glow and abnormal (high current diffuse) glow. Making this test it has been found that the electrical characteristics obtained with the model do not fit the experimental ones.

In order to further improve the accuracy of the models using energy-dependent γ values for argon ions and fast atoms, the apparent γ has recently been derived for cathode-fall conditions, from a heavy-particle hybrid model [10]. The γ values obtained in this study have been found to be significantly lower compared to the case of the homogeneous field and to change remarkably with changing discharge conditions. This implies that it is generally difficult to prescribe a certain value of γ which describes the discharge correctly for a wide range of operating conditions.

In the literature there is a lack of reliable experimental γ values, furthermore the most of the values are determined under high vacuum conditions in ion beam experiments, which can not be used directly for glow discharges. *Therefore the aim of the investigations is the determination of the apparent electron emission coefficient with a fitting method for the abnormal glow region.*

Choosing this alternative solution, γ is used as a fitting parameter and in the iterative solution of the model it is adjusted at the different discharge conditions to match the calculated electrical characteristics with the experimental ones. In order to prove the correctness of the model the experimental and the calculated axial profiles of emission are compared. These intensity distributions provide further information about the discharge: e.g. the length of cathode dark space, and in this way helps in the better understanding of the discharge.

2. An important issue in the gas discharge physics is the effect of the electrodes' geometry on the discharge operation and characteristics. An important group of discharges with special electrode configuration which are important even in some applications is that of hollow cathode discharges.

   Hollow cathode discharges are formed by confining the negative glow into a cathode cavity, which may be one of many different forms, e.g. it can be inside a cylindrical cathode or between two flat cathodes. The current flowing in a hollow cathode discharge may be orders of magnitude higher compared to that in a single plane cathode discharge operating at the same voltage. This advantageous property of the hollow cathode discharges originate from the increased ionization rate in the plasma (so-called hollow cathode effect). The most important reasons of the appearance of the hollow cathode effect are:

the oscillatory motion of fast electrons between oppositely situated cathode surfaces, the effect of enhanced photoelectron-emission from the cathode (due to geometrical effects) and the appearance of stepwise processes (due to the high plasma density). The existence of oscillating electrons – originally proposed by Güntherschulze [11] – was first proved experimentally by Helm [12].

Due to the enhanced light intensity from the negative glow, hollow cathode discharges have a wide range of applications, including gas lasers [13–15] and spectral lamps [16]. At high pressures the so-called micro-hollow cathode discharges are also used as high-intensity light sources [17, 18], while other hollow cathode configurations (at low pressures) have switching applications [19, 20].

In the thesis the development of the hollow cathode effect in a plane-parallel hollow cathode argon glow discharge is investigated experimentally and by means of a two-dimensional hybrid model. The results of the hybrid model are compared with the experimental ones, in this way the correctness of the model is checked. The aim of the work is to follow experimentally and theoretically the development of hollow cathode discharge with changing the discharge conditions at constant electrode separation. A further aim is to prove the existence of oscillating electrons in the discharge.

3. In the low pressure noble gas discharges presented above due to the low pressure and low ionization rate it is considered that the positive charges are the atomic ions. However at higher pressures (10-100 mbar) in noble gas (e.g. helium) discharges used for different applications : e.g. spectral and illumination light sources, as well as for different types of lasers the molecular ions play an important role. In the above mentioned pressure range it has been found that the helium molecular ions influence the discharge conditions and participate in the pumping mechanism of the He-Cd^+ laser, and the He-Zn^+ laser. He_2^+ molecular ions have also been found to be responsible for the excitation in the He-Ar^+ laser. Similarly, they play an important role in the discharges formed in plasma addressed liquid crystal displays (operated in H_2+He mixtures at $p \sim 100$ mbar pressures) [21].

In helium discharges at higher pressures UV and VUV lasers can be realized, e.g. the He-N_2^+ laser which is pumped by a very efficient reaction between helium molecular ions and nitrogen molecules [22, 23]. The UV and VUV radiation of helium discharge lamps originates from the excited He_2 and He_2^+

molecules [24]. As the ground state of He_2 molecule is unstable the excited He_2 molecules result mainly from the recombination of He_2^+.

Furthermore He_2^+ ions are expected to be present in discharges even at relatively low pressures, as indicated by the calculations for positive column discharges, where molecular helium ions were found to become the dominant ionic species at pressures exceeding ≈ 15 mbar [25].

The aim of the studies is to observe how the molecular helium ions become important in the discharge with increasing pressure and to identify the important source and loss processes of atomic and molecular ions as a function of pressure. Therefore in the thesis a helium discharge in a wide pressure range is investigated experimentally and by means of a one-dimensional hybrid model.

3 Investigation methods

The discharges are investigated experimentally and by means of hybrid models.

The hybrid model consist of a fluid model and a Monte Carlo model. In the fluid model the motion of the positive ions and slow electrons are described (by using the fluid equations), the fast electrons (whose kinetic+potential energy is higher than the excitation energy of gas atoms) are traced in the Monte Carlo (MC) model (with particle simulation). The potential distribution in the discharge is determined in the fluid model with the Poisson equation. The input parameters of the fluid model are the gas pressure, discharge voltage, the diffusion coefficients, the mobility and the electron temperature. The input parameters of the Monte Carlo model beside the gas pressure are the electron impact cross sections.

In the hybrid model the fluid and the MC models are solved in an iterative way until the stationary state of the discharge is reached. In the first step of the iterations the fluid model is solved without charge sources to obtain an initial electric field distribution, in which field the primary electrons can be traced by the MC simulation. In the MC model the slow electron and ion sources are determined, which are normalized by the actual value of the current I (calculated in the previous fluid cycle). The next fluid cycle is solved with the sources calculated in the MC model.

In the experiments the electrical characteristics, the axial distribution of the emission light intensity and the spectra of the discharges have been determined. The emission light intensity distribution gives important information about the structure of the discharge and besides this the accuracy of the models can be verified by comparing this experimental distribution with the calculated one. The electrical characteristics of the discharges are input parameters of the hybrid models, the spectrum of the discharges makes possible to determine the electron temperature which is also an input parameter of the models.

4 New scientific results

1. *A one-dimensional hybrid model for abnormal argon glow discharges has been developed. The model made it possible to determine the apparent secondary electron electron emission coefficient (γ) for argon glow discharges operating in the abnormal region.* [f3,k3].

   A discharge in argon gas, with $d = 1.1$ cm electrode separation has been investigated in the $pd = 45$ - 150 Pa cm pressure and $I = 300$ - 2000 μA current range, corresponding to the abnormal glow conditions. In the iterative solution of the model the apparent secondary electron emission coefficient has been adjusted to obtain voltage-current characteristics fitting the experimental values. The model made it possible to calculate several discharge characteristics: spatial distributions of the charge densities, charged particle fluxes, and the electric field. The results have shown that the value of γ changes considerably with discharge conditions; the values obtained for homogeneous field calculations and from ion beam experiments cannot be directly applied in hybrid models for the calculation of voltage-current curves.

2. *Through a detailed comparison of the experimental light intensity distributions with those obtained from the hybrid model of the discharge the correctness of the model has been demonstrated. By determining the length of the cathode dark space (cathode sheath) from the calculated light emission profile and from the electric field distribution it has been proven that the position of the intensity distribution peak corresponds to the position of the cathode sheath - negative glow boundary.* [f3,k3,k4].

   During the comparison a good agreement has been found between the calculated and measured light intensity distributions both in terms of shape and relative magnitude of the intensity of the negative glow. This good agreement has proven the correctness of the model. The length of the cathode sheath – as determined from the light intensity distributions, assuming that the position of the cathode sheath - negative glow boundary corresponds to the position of the emission peak – showed a good agreement with the data obtained from the electric field distributions (by obtaining the position of the cathode sheath - negative glow boundary from a linear extrapolation to zero value of the calculated electric field in the sheath region). This study has confirmed that the position of the cathode sheath - negative glow boundary coincides with the position of the maximum of the light intensity distribution.

3. *A plane-parallel hollow cathode discharge in argon gas with a 2 cm cathode-cathode distance has been investigated experimentally and by means of a two-dimensional hybrid model. By determining the ionization source functions and the emission light intensity distributions the presence of the hollow cathode effect in the discharge at low pressures has been shown. By visualizing the electron avalanches in the simulation the presence of the oscillating electrons in the plane-parallel hollow cathode discharge has been proven*[f1].

   This study has focussed on the effect of changing gas pressure on the characteristics (e.g. ionization source functions) of a hollow cathode discharge. At high pressures the source of ions is concentrated near the cathodes. With decreasing pressure more ions are created further away from the cathode and finally at low pressures ($p \leq 0.4$ mbar) the ionization source becomes uniform in the central part of the discharge. By visualizing the electron avalanches in the simulation it has been found that at the lowest pressure studied ($p = 0.2$ mbar) 90% of the electrons (primaries + their secondaries) enter the sheath of the opposite cathode, and 80% of these electrons make an oscillations between the two cathode surface. At the highest pressure studied ($p = 1$ mbar) only 1% of the electrons cross the central part of the discharge and none of them oscillates between the two sheath regions. The model has been verified through a qualitative comparison of the calculated and the measured light intensity distributions.

4. *The emission spectra of helium molecules has been recorded in a wide pressure range in a plane electrode helium discharge. These spectra have indicated the presence of molecular ions in the discharge even at low gas pressures ($p \approx 20$ mbar). The electron temperature has been also determined experimen-*

tally by means of a spectroscopic technique[f2,k1].

As the ground state of the He_2 molecule is not stable, the excited He_2 molecules cannot be created by electron impact excitation; they result mainly from the recombination of the He_2^+ molecules. By recording the emission spectra of He_2 molecules, it has been found that even at low pressures ($p \geq 20$ mbar) a high density of He_2^+ is present and efficient recombination channels exist in the discharge. As the rates of the recombination processes depend strongly on the electron temperature ($\mathrm{d}n/\mathrm{d}t \sim T_\mathrm{e}^{-4}$), in order to have an accurate model, the electron temperature has been determined experimentally with a spectroscopical method. It has been found that in the 6-60 mbar pressure range the value of the electron temperature is in the range of 0.1-0.12 eV.

5. *For theoretical studies of the helium discharges a one-dimensional hybrid model has been developed. With the model the relative contributions of different processes to the charged particle (electrons, atomic and molecular ions) sources and losses have been determined. The calculated charge density distributions have indicated that even at low pressures ($p = 6$ mbar) the density of He_2^+ is significant. With the calculated particle fluxes the importance of helium molecular ions in the self-sustainment of the helium discharges has been shown.* [f2,k1].

In the pressure range investigated 99% of the atomic ions are created via the electron impact ionization process. At low pressures the ions are mainly lost at the electrodes (at $p = 6$ mbar 85%), with increasing pressure the recombination and ion conversion processes also become important (at $p = 60$ mbar 30% and 12%, respectively).

The molecular ions are created mainly through ion conversion and associative ionization processes (at $p = 6$ mbar 49% and 47%, respectively) and the contributions of these processes vary slightly with pressure. At low pressures the molecular ions are mainly lost at the electrodes (at $p = 6$ mbar 92%), with increasing pressure the importance of the recombination processes increases, becoming the dominant loss processes at $p = 60$ mbar (collisional radiative recombination (44%) and three-body recombination (9%)).

The ratio of the maximal atomic to the maximal molecular ion density at $p = 6$ mbar is ≈ 3.4, this ratio decreases with increasing pressure and at $p = 60$ mbar becomes ≈ 2.2. This result demonstrates that at low pressures the absence of the bands of helium molecules in the discharge spectra is due to the low recombination rate of the molecular ions at these pressures.

Taking into account the electron emission coefficients of different particles and the fluxes of particles on the cathode surface it has been shown that $\approx 10\%$ (this value varies slightly with pressure) of the primary electrons are released by the molecular ions.

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