Dynamics in Micro-scaled Atmospheric Pressure Plasma Arrays

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1. Introduction

1.1. Preface

The aim of this study is to contribute to the understanding of the interaction and dynamics of single microplasmas in a silicon-based micro-structured electrode array arrangement.

Silicon-based micro-structured electrode array devices have huge application potential. Typically operated in rare gases, these devices allow simultaneous homogeneous large-scale surface treatment. Moreover, they are excellent sources of ultraviolet radiation due to operation at high pressure and associated production of excimers through three-body collisions. The homogeneity of emission intensity is exploited in lighting applications. Despite their huge potential in technological applications, the underlying physics of such non-equilibrium dielectric barrier discharge arrays at atmospheric pressure are not well understood yet.

Their small confining structures represent a challenge for diagnostic access which is almost restricted to optical diagnostics. Thus, experimental evidence is rare. Due to the high surface-to-volume ratio of microplasma arrays, dominant processes can significantly deviate from those in discharges of comparably bigger confining structures. At the same time, the array geometry requires complex computational domains which exceed available computer power necessary for detailed simulation: While every single cavity in an array arrangement acts as an autonomous microplasma, adjacent cavities may interact and promote collective behaviour. If computer power was not an issue, the large number of open parameters such as simplifying assumptions, considered species, reactions, and cross sections would still require experimental verification.

This study focuses on single microplasma dynamics and the interaction between adjacent microplasmas in an array structure as these processes deter-
mine device performance. Only if such processes are known, the specific tailoring and modification of new devices to application demands is possible. The discharge mode of the microplasma arrays as well as their dynamics are investigated by means of (phase and space resolved) optical emission spectroscopy as well as voltage-current characteristics. The experimental observations on microplasma interaction are compared to numerical simulations.

1.2. Thesis outline

In the first chapter, plasmas, their basic properties, and main fields of industrial application are briefly introduced before low-temperature atmospheric pressure microplasmas are motivated and the issue of scalability and microplasma array devices as a possible solution are discussed. Advantages and challenges of the micro-structured atmospheric pressure plasma devices investigated in this study are presented together with state-of-the-art knowledge. Here, the focus is on characterisation of such devices as well as on diagnostic access. Finally, the manufacturing process and configuration of the investigated devices are introduced.

In chapter two, the principles of the applied optical diagnostics, that is (phase resolved) optical emission spectroscopy, are summarised. With regard to characterisation of the investigated devices, the various discharge modes of parallel plate atmospheric pressure dielectric barrier discharges are discussed with particular focus on their distinction.

The experimental setup including technical components and respective specifications is introduced in chapter three. Moreover, the analytic software developed in the frame of this study and underlying data preparation schemes are described.

Chapter four comprises the results obtained in the frame of this study. A characterisation of the discharge mode of the investigated devices with respect to self-pulsing of emission and current under variation of operation parameters is given. The dynamics of single microplasma cavities in the MSE array structures as well as the coupling of adjacent cavities are investigated by means of phase and space resolved optical emission spectroscopy using an intensified charge-coupled device camera with attached long-distance microscope. Propagation velocities of observed ionisation waves are determined, observations
are compared to two-dimensional fluid simulation results and underlying processes identified on this basis. The individual microplasma emission behaviour is compared to that of the integral array devices. Finally, dynamics of single cavities and asymmetry of the latter with respect to electrode polarity are examined by means of temporally and spatially resolved emission profiles.

The last chapter summarises the obtained results and gives an outlook on possible further investigations as well as improvements.

1.3. Plasma

Plasma as fully or partly ionised gas gained increasing importance in technological applications and attention in research since the middle of the twentieth century. The ability to control species densities and energies makes it a very versatile tool in processing.

Plasma in or close to thermal equilibrium is exploited as energy converter in fusion experiments and as carrier of thermal energy in welding, cutting as well as in drilling and plasma spray coating applications. In gas-blast switches, plasma is used as buffer gas to prevent sparks while it exhibits high electric conductivity.

The named applications mostly rely on the thermal character of plasma. However, it is often desired to take advantage of single plasma species’ properties at simultaneous absence of heat. While radiation of excited species is most important in illumination and display industries, separate control of ion momentum and ion flux is crucial for deep-trench etching or ion implantation in semiconductor industries as well as for space propulsion. Applications mainly relying on plasma chemistry include, for example, synthesis of ozone, acetylene, nano-particles or destruction and reduction of nitric and sulphur oxides in exhaust fumes respectively.

It is obvious that applications often demand well-directed supply of energy only to specific plasma species. Thus, most technological plasmas have a strong non-equilibrium character. Their description is therefore often very complex.

Energy to create, sustain, and confine a plasma is usually supplied by externally applied electric or magnetic fields. These external fields can be used to
accelerate and control propagation of charged species. Sufficient kinetic energy gain provided, these charged species can collide with other species and transfer momentum or lead to dissociation, excitation and ionisation. As the energy gain between subsequent collisions is strongly dependent on the mean free path of particles, many technological plasmas are realised at low gas pressures and low particle densities respectively. This allows to sustain a discharge at common voltages of a few hundred volts up to some kilovolts.

As many technological applications of plasmas involve treatment of heat-sensitive substrates, most technological plasmas are generated using alternating electric fields at high frequencies. This way, the light electrons are accelerated and used to sustain the plasma while ions cannot instantaneously follow the alternating field due to their comparably high inertia. As the ions do not gain and therefore cannot transfer much kinetic energy to neutral species while momentum transfer through electrons is ineffective, the gas temperature remains close to room temperature. Thus, these discharges are often referred to as ‘low-temperature’ or ‘non-equilibrium’ plasmas.

Technological low-pressure plasmas are particularly well suited for substrate treatment. As the sheath in such plasmas is collisionless, ions are effectively accelerated towards substrates and high-energetic ion bombardment is achieved. This effect can be enhanced in capacitively coupled plasmas by choosing asymmetric electrode configurations which lead to build-up of a self-bias. However, in some cases high-energetic ion bombardment of substrates may also be unwanted (thermolabile substrates).

However, operation at low pressure requires potential substrates to be treated inside expensive vacuum chambers which involves the use of load locks and remote assembly techniques. Apart from inconvenience, such setups require high maintenance and limit the use of plasma treatment to vacuum-compatible substrates. Moreover, the size of treated substrates is limited by vacuum chamber dimensions.

1.4. Atmospheric pressure plasmas

A concept to avoid these downsides of low-pressure discharges is operation close to or even at atmospheric pressure. However, the comparably high pressure and particle densities respectively lead to small mean free paths of particles. Therefore, electric fields to accelerate charged particles must be higher
than at low pressure to ensure sufficient energy gain for ionisation before subsequent collisions. This can either be realised by higher applied voltages or a reduction of the electrode gap size. This correlation of applied voltage, pressure, and discharge dimension has first been stated in 1889 by Friedrich Paschen [1] and is commonly known as ‘Paschen’s law’.

In this collision-dominated operation regime, ions are practically not accelerated in the sheath. If high-energetic ion bombardment of targets is desired, this has, for example, to be achieved by pulsing the discharge and explicit biasing of the substrate.

Due to high collision rates, atmospheric pressure plasmas are susceptible to instabilities. Apart from surface impact of heavy species, effective stepwise ionisation and Penning ionisation [2] involving meta-stable species can lead to avalanche-like secondary electron generation. This can result in a transition of the discharge from stable glow mode to thermal arc.

A promising concept to avoid instabilities and thus generate stable low-temperature atmospheric pressure discharges is the design of plasma sources with small confining structures [3]. Due to their small dimensions, this kind of discharges - although often measuring several millimetres - are referred to as ‘microplasmas’ or ‘microdischarges’. Over the last decades, many different microplasma sources have been developed for a variety of applications. In particular, the ability to effectively generate ultraviolet (UV) radiation and to treat vacuum incompatible substrates such as polymers and biological materials make this kind of plasma sources attractive. Apart from biomedical applications, such as disinfection of wounds [4], treatment of tissue or skin [5, 6] or dental caries [7], this kind of plasmas is also tested for consumer market purposes such as pre-treatment of fingernails [8] or biological deactivation in the food industry, just to name a few. However, most knowledge of the interaction and behaviour of such microplasmas is of phenomenological nature. The complex chemistry and actual causalities are not yet fully understood. Thus, despite their enormous application potential, most of the specified sources remain prototypes.

A drawback of microplasmas is their inherent limited individual size. Many applications such as treatment of large or complex surfaces as well as illumination demand scale-up of single microplasmas without simultaneous renunciation of the benefits of cold atmospheric pressure discharges.
1.5. Micro-structured atmospheric pressure plasma devices

During the last decade, the scale-up of microplasma discharges has been realised by arrangement of multiple microdischarges in an array geometry. Many different concepts have been introduced. One well-established scheme is represented by so-called plasma display panels (PDPs) with separately addressable dielectric barrier discharges which enjoy wide spread in the display technology (see below). Atmospheric pressure plasma jets have the advantage of remote species generation and a directed particle flow towards substrates. Arrays of such jet devices have been realised ranging from linear configurations of ten devices [9] to two-dimensional setups with 5 by 5 capillary devices [10] or 37 devices in a honeycomb configuration [11]. However, the packing density and area of coverage of such plasma jet arrays is still very limited.

A concept to achieve high packing densities and large areas of coverage is introduced by arrays of so-called micro-structured electrode (MSE) devices. Their name derives from confining structures in the range of 100 µm or even less. These devices are typically fabricated using micromachining technologies such as, for example, photolithography, plasma activated chemical vapour deposition (PCVD), chemical and plasma etching or galvanic techniques. A multitude of different MSE array designs has been introduced over the recent years, ranging from comb-like alternating electrode structures on an insulating substrate [12, 13] over arrays of micro hollow cathode (MHC) discharges in silicon devices [14, 15, 16], arrays of quarter-wave linear microwave resonators [17], and plasma stamps based on porous sinter metal [18] to arrays of cavities in doped silicon [19], alumina [20] or glass substrates [21]. Transparent arrays of microplasma cavities fabricated by plastic-based replica molding with indium tin oxide (ITO) electrodes [22] as well as flexible array devices [23] have also been reported on. This study investigates silicon-based MSE devices which exhibit high flexibility in design and features as discussed in the following.

Apart from high packing densities, MSE array devices exhibit several features which make them valuable tools in atmospheric pressure plasma generation. The manufacturing of such devices offers high flexibility in designing the sources with respect to substrate material, possible electrode configurations, and stack structures. Moreover, reproducibility of photolithographically de-
fined structures is very high. In particular, doped silicon-based devices have established over the recent years, as silicon is a well-known material and its conducting and isolating properties can be influenced by the chosen doping. Silicon wafers are widely spread in industrial applications. Thus, the raw material for such devices is easily available, where processing equipment is well-established in the microchip and microelectromechanical system (MEMS) as well as in photonics industries. Typical feature sizes smaller than 100 nm are technically realisable with comparably inexpensive standard equipment. The size of single array devices is mainly limited by photolithographic components, mask and wafer sizes as well as substrate stability.

Typically, such silicon based micro-structured atmospheric pressure plasma devices consist of a doped silicon wafer as one electrode with cavities etched into the silicon. A dielectric around the aperture of the cavities serves as separation from a second, mostly metallic, electrode which is usually coated on top of the former. While some devices are operated with bare electrodes and DC voltage, others are equipped with dielectric coatings on the electrodes and are thus limited to AC operation. Typical voltage amplitudes for both kinds of excitation are in the order of some hundred volts, where in AC operation usually driving frequencies in the range of a few up to some ten kilohertz are applied.

Devices based on arrays with typical cavity sizes in the order of several ten micrometres and cavity spacings in the same range have huge application potential. Their semiconductor-based nature allows their integration into electronic devices and subsystems. For example, their small feature sizes promote lab-on-a-chip applications, where the microplasma (array) can be used to dissociate, excite or modify species or targets for subsequent or in-situ analysis. They may also be used as an optical source in MEMS which those systems lack to date.

When operated in DC with bare electrodes in neon, silicon-based microplasma arrays have shown to be effective photodetectors in the visible, ultraviolet, and near-infrared spectral range [24, 25]. Photosensitivities in the order of 1 A W$^{-1}$ have been realised, depending on cavity size as well as process gas, pressure, and irradiating wavelength. The incident emission is registered by a voltage excursion caused by photocurrent. Compared to bulk silicon, the photosensitivity of the hybrid detector is typically one order of magnitude higher, exhibiting the same wavelength dependence. This indicates, that primarily the bulk silicon determines the spectral response. However, the plasma enables
and enhances electron multiplication, leading to the observed increased photosensitivity. Due to the strong electric fields in the cavity (~ 200 kV cm$^{-1}$ [26]), band bending at the plasma/silicon interface is sufficient to produce a thin n-type region at the surface further enhancing photosensitivity, comparable to the inversion layer produced in metal-oxide-semiconductor (MOS) capacitors. This can enhance tunnelling of electrons originating from radiatively produced electron-hole pairs from the silicon surface into the plasma. Electron multiplication factors of at least 4 have been achieved, where no strong influence of incident power on photosensitivity has been observed. Although realised photosensitivities cannot compete with that of commercially available semiconductor-based avalanche photodiodes (APD, ~ 40 A W$^{-1}$ for Hamamatsu S2381), the ability to produce large scale detectors at comparably low costs may be attractive. The coupling of low-temperature plasmas with materials such as semiconductors at an interface also allows tailoring of novel electrooptical devices.

Furthermore, such microplasma arrays can efficiently generate highly uniform radiation over large areas. Emission intensity has been shown to be reproducible and constant over arrays of 40000 and more single cavities within ±10% for operation with various rare gases [27] and small molecular additives at atmospheric pressure. The collisional character of such discharges abets three-body collisions. These are in combination with typical power loadings of tens of kW cm$^{-3}$ [28] essential for efficient generation of short-lived atomic or molecular species such as rare gas-halide excimers. The latter are an important source of UV radiation. Thus, effective generation of UV radiation in such microplasma arrays allows to uniformly treat large surfaces and samples, for example in sterilisation or surface modification processes.

Moreover, these arrays are suitable as plasma display panels, where single cavities can be separately addressed by using slightly modified electrode structures [29]. They were shown to exhibit luminous efficacies of up to 7.2 ± 0.6 lm W$^{-1}$ [30], depending on operation parameters and employed phosphors. This is a very good value compared to typically 2 lm W$^{-1}$ for commercially available PDPs. To date, arrays with cavity sizes as small as 10 µm [30] and 5 µm have already been realised. As typical dimensions of PDP pixels are in the range of ~ 300 µm, the silicon-based microplasma arrays do not only exceed commercial PDPs in luminous efficacy, but also in possible spatial screen resolutions.

Apart from using the combination of arrays and phosphors for PDPs, it can
also be used as a component of homogeneous light sources. For a gas mixture of xenon (Xe) and iodine (I₂) vapour in a ratio of 50 to 1 operated at a rather low pressure of 68 mbar in a silicon micro-hollow cathode of 400 μm diameter, strong UV emission has been observed in the interval of 200 – 300 nm. Moreover, the XeI molecule shows peaked emission at 254 nm originating from the B → X transition [28], coinciding with the mercury (Hg) resonance line which is exploited in most energy-saving light bulbs. Thus, microplasma array devices operated with this rare-gas halide vapour mixture in combination with a phosphor have the potential to become a non-toxic replacement of energy-saving light bulbs containing Hg. Commercially available light sources based on such arrays are envisaged to exhibit edge lengths in the range of metres [31] while edge lengths of several 10 cm have already been realised.

First tests show that linear microplasma arrays or microchannel devices are suitable to achieve gain of certain rare gas excited states and emission lines respectively [30]. In combination with adequate optical resonators, they may be exploited in lasing applications.

One challenge in the field of MSE devices is their lifetime. While many devices are driven in DC with bare electrodes, their lifetime is rather limited to a few hours of operation [27]. Ion bombardment at the cathode as well as electron sputtering at the anode lead to deterioration of the comparably thin electrode structures of typically some micrometres thickness. Here, dielectric coatings of the electrodes are a promising approach to enhance device lifetimes. The coatings offer resistance to chemical and physical erosion. More than 100 hours of continuous operation without noteworthy reduction of the radiative output have been achieved to date [27]. However, dielectric coatings imply restriction to AC operation and totally different discharge physics compared to bare electrode DC operation. In this case, single microplasma discharges in the array arrangement function as dielectric barrier discharges (DBDs).

It is obvious that arrays of MSE devices as atmospheric pressure plasma sources have huge application potential due to the flexibility in both, their design tailored to specific application standards as well as purpose of use. Thus, development of new microplasma array sources is rapidly advancing. However, the small confining structures of only a few Debye lengths (~ μm) may entail new physics where no quasi-neutral plasma can exist and ignition processes may deviate from well-known Paschen behaviour. The understanding of basic discharge properties and plasma dynamics of such devices is still rudimentary. In particular the coupling of adjacent microplasmas in an array arrangement...
has neither been deeply investigated nor understood to date. In order to tailor arrays of MSE devices to specific application demands and to optimise their design, the understanding of such dynamics and underlying processes is indispensable.

While manufacturing processes and electrical characterisation as well as integral optical emission of whole microplasma arrays have been extensively studied [19, 30, 32], little is known about single microplasma behaviour. The electrode structure does not allow to directly access electrical parameters of single cavities.

Due to the homogeneity of observed integral emission features of such microplasma arrays, they are often referred to as atmospheric pressure glow discharges [28]. However, distinction of discharge modes of dielectric barrier discharges is rather complex investigating microplasma arrays. A glow discharge is typically characterised by the build-up of a positive column and cathode fall respectively in the plasma volume which in this case is intricate to observe by optical diagnostics due to the small confining structures of the single microplasmas. As the gap voltage is not easily accessible due to the rather complex layer structure and cavity geometry, a characterisation of the discharge mode by means of voltage-current characteristics in combination with an equivalent circuit model is challenging. Nevertheless, a characterisation of the discharge mode is necessary in order to optimise MSE array devices in dependence of envisaged applications as it determines dominating processes, for example, domination of secondary electron multiplication at the dielectric surfaces compared to electron multiplication in the bulk or vice versa.

Simulations for a single microplasma cavity exist [26] but are restricted to DC operation with bare electrodes and neglect the array arrangement and resulting influence of adjacent cavities. Thus, neither can existing theoretical investigations comprehend dynamics in a single dielectrically coated cavity in an array arrangement, nor is the periodicity of AC excitation allowed for.

Access to plasma parameters using probes is very limited due to the small confining structures. Even if microprobes were available, their intrusive character would not allow deduction of undisturbed plasma parameters. A promising approach to investigate such devices lies in optical diagnostics. The enclosed character of single MSE array cavities restricts the use of active optical diagnostics such as absorption spectroscopy and \textit{(laser-induced)} fluorescence spectroscopy. Thus, passive optical emission spectroscopy \textit{(OES)} which exploits
1. Introduction

Radiative emission of excited energetic states of plasma species in combination with collisional-radiative models (CRM) to deduce plasma parameters, is an appropriate approach to investigate plasma dynamics of the microplasma array devices.

This thesis comprises investigations of plasma dynamics of single microplasmas arranged in an array structure as well as of microplasma arrays as a whole by means of phase and two-dimensionally space resolved optical emission spectroscopy in combination with integral voltage and current measurements. In particular, the interaction of adjacent microplasmas and underlying processes and the resulting influence of the array arrangement on discharge behaviour are analysed. Moreover, the transient and periodical character of microscale DBDs and its impact on discharge stability and modes are reported.

In the following, the two different kinds of micro-structured electrode array devices as well as one discharge channel device investigated in this work are described in detail. The array arrangements investigated in this study are specified by typical designs used in the research groups of collaborating partners.

1.5.1. Pyramidal cavity arrays

These kinds of devices exhibit arrays of inverted, square pyramids. They are based on (100) oriented p-doped silicon (Si) wafers of typically 6-8 Ωcm resistivity and 300 µm thickness [19] which serve as electrode. The pyramidal cavities of 50 or 100 µm square bases are produced by anisotropic wet etching of the Si wafer. Here, potassium hydroxide (KOH) in a 33% wt/wt solution in water is used as etching reactant. As KOH etches (100) planes of Si much faster than (111) planes [33], the characteristic inverse pyramidal pits result with sides of the pyramid under an angle of 54.7° with respect to the wafer’s principal plane. Afterwards, the whole wafer is coated with an approximately 0.1 µm thick layer of silicon nitride (Si$_3$N$_4$, relative permittivity $\varepsilon_r = 7.5$) using plasma-enhanced chemical vapour deposition. In the next step, a dry etchable polyimide (Dupont 2611, $\varepsilon_r = 2.9$) of 8 µm thickness is deposited on the unetched part of the wafer surface by spin coating. This process is followed by e-beam evaporation of a 0.12-0.2 µm thick nickel (Ni) film on top of the polyimide. While the resulting Ni grid serves as second electrode, the polyimide separates it from the silicon wafer. Finally, the device surface is again coated with a second 0.1 µm thick layer of Si$_3$N$_4$. 
1. Introduction

Figure 1.1.: Sketch of the cross section of a pyramidal cavity array device.

Figure 1.2.: Scanning electron microscope images of the cross section of (a) an inverted pyramidal cavity of 50 µm base length and (b) the magnification of the area indicated by the white circle in image (a) as published by Eden et al. [34].

Desired patterns such as pyramidal bases as well as dielectric films on the wafer are defined photolithographically with a chromium (Cr) mask and shaped by wet and reactive ion etching respectively. Further details on the production of these kinds of devices can be found in the literature [19].

A sketch of the cross section of such pyramidal cavity array devices as well as scanning electron microscope (SEM) images of the cross section of a single inverted pyramidal cavity of 50 µm base length are shown in figure 1.1 and figure 1.2 respectively. The SEM images illustrate the high precision in manufacturing such devices allowing excellent reproducibility.

These microcavity devices are typically arranged in a square array geometry comprising 2500 single discharge cavities. The cavity spacing complies with
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the lateral length of the square cavity base. However, various array geometries and configurations are possible and arrays of up to 500x500 single cavities have already been realised [32]. Devices investigated in this thesis typically consist of 50x50 cavities with 50 or 100 µm edge length and cavity spacing respectively.

The array arrangement of the pyramidal cavity devices is shown in figure 1.3.

All pyramidal cavity arrays investigated in this study are provided by the Laboratory for Optical Physics and Engineering, University of Illinois at Urbana Champaign, Illinois, USA.

1.5.2. Straight cavity arrays

These devices are as well based on doped (100) oriented Si wafers of 500 µm thickness as electrodes. For these devices, the doping is negative and typical resistivities are 2-5 Ωcm. While the general configuration of such devices is very similar to that of the arrays of inverted pyramidal cavities described above, the fabrication process is different. The wafers are first coated with a 6 µm thick layer of silicon dioxide (SiO$_2$) before a 10 nm thick titanium (Ti) and a 100 nm thick copper (Cu) layer are consecutively deposited as adhesive
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agents. Subsequently, cavity structures are defined photolithographically using a photoresist. After that, a 1 µm thick Ni layer is coated on those parts of the wafer where no cavities are defined to form the second electrode. In the next step, the cavities are shaped. First, Cu and Ti are etched using selective wet etchants BTP and BHF respectively. Afterwards, the SiO$_2$ is etched using a trifluoromethane and argon ($CHF_3/Ar$) discharge. The Si is then etched using the STiGer process, which involves cycling passivation steps in a silicon tetrafluoride and oxygen ($SiF_4/O_2$) plasma and anisotropic etching steps using a sulfur hexafluoride ($SF_6$) discharge [35]. Thus, cavities with sides under an angle of 90° with respect to the wafer principal plane are shaped 2 µm into the Si surface. Finally, an Si$_3$N$_4$ layer of 2 µm thickness is coated on the whole wafer surface to form a dielectric barrier.

The backside of the Si wafer is finally coated with a 200 nm thick gold (Au) layer using a 10 nm thick Ti layer as adhesive agent. This layer serves enhanced contacting of the Si wafer.

All dielectrics are deposited using plasma enhanced chemical vapour deposition (PECVD). While a sputtering metal deposition is realised for Ti, Cu and Au, the Ni coating is deposited electrochemically.

Figure 1.4 and figure 1.5 show a sketch of the cross section of such straight cavity array devices and a SEM image of the cross section of a single cylindrical straight cavity of 150 µm base diameter.

These kinds of devices are more versatile as to cavity shapes compared to the
1. Introduction

pyramidal cavity devices as there are no geometrical restrictions for the etching reactant. This method of fabrication would also allow spherical cavities shaped by isotropic etching using an $\text{O}_2/\text{SF}_6$ plasma. However, devices investigated in this thesis are fabricated using anisotropic etching.

Several different cavity configurations and two kinds of array geometries are available. Cavity configurations in this thesis comprise cavities of rectangular shape with edge lengths of 500 $\mu$m times 25, 50, 100 and 150 $\mu$m respectively as well as cavities with circular bases of 25, 50, 100 or 150 $\mu$m diameter. The cylindrical cavities are either arranged in square arrays of 32x32 cavities of the same size or in four different arrays of 16x16 cavities each, with one array per cavity size. The rectangular cavities are arranged in the configuration of four arrays of 5x16 cavities, one array per cavity size only. Cavity spacings are in each case 150 $\mu$m.

The different array arrangements for straight cavity arrays are illustrated in figure 1.6.

All straight cavity devices investigated in this study are provided by the Groupe de Recherches sur l’Energétique des Millieux Ionisés, Université d’Orléans, Orléans, France.
1. Introduction

Figure 1.6.: Straight cavity array arrangements: 4x5x16 rectangular cavities (a), 4x16x16 cylindrical cavities (b), 32x32 cylindrical cavities (c).
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A third kind of devices investigated in this work is a discharge channel device. From its geometry and fabrication, it is similar to the pyramidal cavity arrays. It offers a more simple symmetry compared to the array devices. While it exhibits the same wafer processing and layer structure, it consists of one single extended groove cavity of 50 µm width, 35 µm depth, and 3.5 cm length. The groove cross section is an inverted equilateral triangular structure resulting from the fabrication processing. As the cross section of the groove resembles that of an inverted pyramidal cavity of the array devices, this channel device is suitable to investigate single cavity behaviour. It is not as sophisticated to model this discharge due to its linearity in the third spatial dimension. This kind of device is only briefly discussed in the frame of this study in order to justify assumptions made in a simulation of the array devices (see section 4.2.1). A schematic sketch of the discharge channel device is shown in figure 1.7.

Discharge channel devices investigated in this study are provided by the Laboratory for Optical Physics and Engineering, Illinois, USA.

Figure 1.7: Schematic sketch of the discharge channel device.

1.5.3. Discharge channel devices
2. Fundamentals

To investigate the discharge behaviour of single microplasma cavities as well as the interaction between adjacent cavities in an array structure, an optical setup is installed, allowing phase, two-dimensional space as well as spectral resolution of the emission of the examined device. As this study mainly focuses on spectroscopic analysis, the basic principles of (phase resolved) optical emission spectroscopy are discussed in the following.

Subsequently, atmospheric pressure dielectric barrier discharges and inherent discharge modes and regimes are presented. As all of the MSE array devices investigated in this thesis exhibit dielectric coatings of the electrodes, such knowledge about dielectric barrier discharges is indispensable for the characterisation of their discharge mode.

2.1. Optical emission spectroscopy

Optical emission spectroscopy is a valuable tool for the investigation of excited electronic states in plasmas. The optical emission and absorption of a discharge implicitly mirrors the population density distribution of excited states and thus allows deduction of plasma parameters. However, processes involved can be numerous and complex and sophisticated collisional radiative models are needed in order to relate emission intensities to densities of excited states.

In particular at atmospheric pressure, heavy particle collisions play an important role so that electron impact excitation out of the ground state and spontaneous emission cannot be considered the only relevant processes determining the population density of excited states, as assumed in the so-called ‘Corona model’. In the Corona model, the population density $n_i$ of an excited state $i$ can be described considering ground state density $n_0$, electron impact excitation function out of the ground state $E_{0i}^e$ and the sum of all transition rates
2. Fundamentals

\(A_{ij}\) of spontaneous emission from excited state \(i\) to lower states \(j\) (also called ‘Einstein coefficients’ for spontaneous emission):

\[
\frac{dn_i(t)}{dt} = n_0 E_{ei}^c(t) - n_i(t) \sum_j A_{ij}
\] (2.1)

As an excited state \(i\) can also be populated due to electron impact excitation out of lower excited states \(j\) of population density \(n_j\) and spontaneous emission competes against induced emission, the Corona model has to be extended by induced emission transition rates \(R_{ij}\) and a generalised electron impact excitation function. Due to their rather high population densities, in particular metastable states often have to be considered as an important source for electron impact excitation of higher excited states.

\[
\frac{dn_i(t)}{dt} = \sum_j n_j(t) E_{ej}^c(t) - n_i(t) (R_{ij}(t) + A_{ij})
\] (2.2)

Apart from electron impact excitation, collisions with heavy particles (excitation function \(E_{ji}^h(t)\)) as well as absorption of radiation originating from a higher state \(j\) (rate \(R_{ji}\)) can lead to further population of an excited state \(i\).

\[
\frac{dn_i(t)}{dt} = \sum_j n_j(t) \left( E_{ji}^c(t) + E_{ji}^h(t) + R_{ji}(t) \right) - n_i(t) (R_{ij}(t) + A_{ij})
\] (2.3)

The electron impact excitation function \(E_{ji}^c\) is determined by electron density \(n_e\), the energy dependent electron impact excitation cross section \(\sigma_{ji}(E)\) and the integral of the normalised electron energy distribution function (EEDF) \(f(E)\) with the electron mass \(m_e\):

\[
E_{ji}^c = n_e \int_0^\infty \sigma_{ji}(E) \sqrt{\frac{2E}{m_e}} f(E) dE
\] (2.4)

The equation for heavy particle impact excitation is similar and thus omitted here.
2. Fundamentals

Heavy particle impact cannot only contribute to the population density of excited states, but also lead to collisional de-excitation, so-called ‘quenching’. This collisional loss channel competes against spontaneous and induced emission and can be considered by introducing an effective de-excitation rate $A_i$:

$$A_i = \sum_j A_{ij} + \sum_q k_q^i \rho_q$$  \hspace{1cm} (2.5)

The additional de-excitation rate introduced here is composed of heavy particle quantum state densities $\rho_q$ and the corresponding quenching coefficients $k_q^i$. The $k_q^i$ respect all de-excitation and excitation processes due to inelastic collisions of species $q$ with particles in the excited state $i$. This kind of collision is usually characterised by domination of de-excitation and thus energy transfer of the excited state $i$ to internal energy of the species $q$ and translational energy of both species.

The same scheme applies for multi-body collisions with an effective collisional de-excitation rate $Q_i$:

$$Q_i = \sum_{q,s} k_q^i \prod_s \rho_{qs}$$  \hspace{1cm} (2.6)

Quenching coefficients are dependent on the mean velocity $\langle v \rangle$ of species $q$ and collisional de-excitation cross sections $\sigma_q^i$. As the $\sigma_q^i$ show only a weak temperature dependence [37, 38], the following correlation derives for a thermal velocity distribution:

$$k_q^i(T) = \sigma_q^i \langle v \rangle = \sigma_q^i \sqrt{\frac{8k_B T}{\pi \mu}}$$  \hspace{1cm} (2.7)

where $\mu$ is the effective mass of both colliding species, $T$ is the temperature of species $q$, and $k_B$ is the Boltzmann constant.

It is obvious from equation 2.7 that OES considering quenching does not only require knowledge of possible quenching partners as well as their densities
and corresponding quenching coefficients, but also of respective species’ temperatures. Here, each quantum state of each colliding species must be considered separately. However, due to the low ionisation degree of low-temperature plasmas, it is for the latter in most cases sufficient to consider only species in the ground state. The comparably low densities of excited species in such plasmas and the consequential low collision probability can allow negligence of their contributions to quenching. Still, data for quenching coefficients is very limited. Thus, it is often not possible to consider contributions of individual species. Nevertheless, the total effective collisional de-excitation rate $Q_i$ of an excited state $i$ can be determined by measuring its reduced lifetime $\tau_i^*$ via the temporal decay of the fluorescence signal of the excited state:

$$Q_i = \frac{1}{\tau_i^*} - \frac{1}{\tau_i} \quad (2.8)$$

where $\tau_i$ is the natural lifetime of excited state $i$.

Respective natural lifetimes can, in the case of atomic energy levels, be calculated from Einstein coefficients for spontaneous emission found in the literature [39]:

$$\tau_i = \left( \sum_j A_{ij} \right)^{-1} \quad (2.9)$$

Quenching coefficients can be experimentally determined from electron beam and laser experiments or low pressure phase resolved optical emission spectroscopy. After peaked excitation, the decay of emission intensity of the investigated state is determined for a variation of partial pressure of the quenching partner [40, 41].

Further discussions and investigations covering quenching and involved processes can be found in the literature [42, 43, 44, 45, 46, 47, 48].

A similar principle applies for inelastic collisions of excited species with electrons which can also lead to de-excitation (rate $C_{ij}$). However, in particular for low-temperature atmospheric pressure plasmas, this kind of collision is rather improbable. The high number of neutral species compared to excited species
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Illustration of possible transitions between two energetic states \( i \) and \( j \). as well as the comparably low cross section values for such reactions make them negligible compared to electron impact excitation.

Possible transitions between two energetic states \( i \) and \( j \) as discussed in the section above are illustrated in figure 2.1.

Population densities of excited states may also be influenced by radiation trapping, that is absorption of radiation emitted during the transition from an excited state \( i \) to a lower state \( j \) by an other particle in state \( j \). This can be considered by introducing so-called ‘escape factors’ \( g_{ij} \) which give the probability for one photon to be absorbed as described above:

\[
A_i = \sum_j A_{ij} g_{ij} + \sum_q k_q^i n_q
\]

(2.10)

Radiation trapping can thus decrease the effective de-excitation rate \( A_i \) and therefore increase the effective lifetime of an excited state. The extreme cases of total (\( g_{ij} = 0 \)) and of no re-absorption (\( g_{ij} = 1 \)) are called ‘optically thick’ and ‘optically thin’ respectively.

For the case of Doppler broadening dominated emission line profiles, corresponding escape factors can be determined using the optical thickness \( \tau_{ij} \) [49]:

\[
\tau_{ij} = \frac{1}{8\sqrt{\pi}^3} \lambda_0^3 A_{ij} \frac{g_i}{g_j} \sqrt{\frac{m}{2k_B T}}, \quad \forall \tau_{ij} \geq 3
\]

(2.11)

\[
g_{ij} = \frac{1}{\tau_{ij} \sqrt{\pi \ln (\tau_{ij})}}, \quad \forall \tau_{ij} \geq 3
\]

(2.12)
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where $\lambda_0$ is the central wavelength, $l$ is a characteristic length of the plasma, $g_i$ and $g_j$ are statistical weights, $m$ is the mass of the excited particle, and $T_g$ is the gas temperature.

As $g_{ij}$ scales reciprocally with $n_j$, re-absorption requires a noteworthy population density $n_j$ of the absorbing state $j$ in order to occur. This usually only applies for the overpopulated ground state or metastable states. Excited states that only allow resonant transitions to the ground state may also build up high population densities due to radiation trapping and thus may as well have to be treated like metastable states at low pressure. However, at atmospheric pressure quenching becomes effective and short lifetimes of resonant states result. Moreover, line broadening for non-equilibrium atmospheric pressure discharges is dominated by pressure broadening rather than Doppler broadening, so that the above relation may not apply. The optical thickness of a plasma can, for example, be examined by backscattering of the plasma emission into the plasma volume and determination of absorption of respective emission lines. Another approach is the examination of emission line profiles. The optical thickness can be deduced from line broadening, as the former leads to increased emission line wing intensity. However, at atmospheric pressure it is sophisticated to distinguish between the influence of pressure broadening and optical thickness.

Another source contributing to the population of excited state densities are cascade processes. First order transition cascades from higher excited states $c$ to an excited state $i$ can be included by introducing accordant effective excitation rates $A_{ci}$:

\[
\frac{dn_i(t)}{dt} = \sum_j n_j(t) \left( E_{ji}(t) + E_{ji}^h(t) + R_{ji}(t) \right) - n_i(t) \left( A_i + R_{ij}(t) + C_{ij}(t) \right) + \sum_c n_c(t) A_{ci} \tag{2.13}
\]

Higher order cascades can also contribute to population of excited state densities and can be considered in the same manner as shown above for first order cascades.

Furthermore, recombination of differently charged particles as well as formation and dissociation of molecules such as electron-ion recombination or dissociative excitation may contribute to population of excited state densities.
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Illustration of possible transitions between energetic states in a multi-state system, where 0, 1, \ldots, j, i, \ldots, n-1, n denote the energetic states of successively higher order with the ground state 0. Arrows indicate excitation and de-excitation mechanisms and associated transitions between respective energetic states as shown in figure 2.1. Thick arrows represent several transitions in summary.

For a multi-state system including vibrational, rotational, and ionisation states as described by equation 2.13, a multitude of possible transitions between energetic states results. Figure 2.2 illustrates the complexity of such systems and possible transitions.

However, in case of non-thermal atmospheric pressure plasmas, assumptions on involved processes and species allow to simplify this system.

While for this kind of plasmas electron impact excitation mostly dominates radiative excitation, de-excitation is mainly borne by fluorescence and inelastic collisions with heavy particles.

As electron impact excitation depends on the population density of the lower state, it mainly occurs out of the overpopulated ground state. High particle densities lead to enhanced quenching and thus cause short effective lifetimes of resonant states $r$. Thus, electron impact excitation out of resonant states is negligible as it has to occur within the lifetime of respective states.

However, metastable states $m$ with significantly longer lifetimes may be notably populated and therefore have to be taken into account as source of step-
2. Fundamentals

Illustration of main excitation and de-excitation mechanisms for low-temperature atmospheric pressure plasmas, where resonant, metastable energetic states, and the ground state. Thick arrows indicate excitation, thin arrows denote de-excitation mechanisms.

Figure 2.3 illustrates main excitation and de-excitation mechanisms for low-temperature atmospheric pressure plasmas.

The observed emission intensity $I_{ij}$ at a certain wavelength $\lambda_{ij}$ depends on the number of photons per volume and time $\dot{N}_{ij}$ emitted by the observed energetic state $i$ which is defined by the spontaneous fluorescence rate $A_{ij}$ and the population density of $i$:

$$I_{ij} \propto \dot{N}_{ij} = A_{ij} g_{ij} n_i$$

(2.14)

In order to relate the fluorescence yield to plasma parameters such as species densities, electron density or EEDF, one has to solve the system of rate equa-
2. Fundamentals

tions of quantum states involved defined by equation 2.13. Here, optical dipole selection rules limit the number of allowed radiative transitions.

The resulting system of coupled differential equations is completed by two free parameters: Electron density and electron energy distribution function. Both determine, together with electron impact excitation cross-sections, the electron impact excitation function (see equation 2.4).

In most cases, one has to take into account the transient spatio-temporal character of the EEDF and electron density. Both can be calculated ab initio using appropriate plasma models. Another approach of access is the derivation from absolutely calibrated emission profiles of transitions originating from excited states with considerably different excitation thresholds. Sufficient electron density and spatio-temporal as well as spectral resolution provided, the former can also be calculated from Stark broadening, for example, of the hydrogen Balmer emission lines for small hydrogen admixtures [50]. Yet, experimental determination based on OES as well as computation of electron density and EEDF always involve underlying models and tabulated cross-sections which limit the accuracy of such methods.

Some diagnostic techniques such as actinometry allow to deduce plasma parameters without specific knowledge of electron properties. If a small admixture of an actinometry rare gas of well-defined density is added to a discharge and exhibits an excited state with a similar electron impact excitation cross section as the excited state under investigation, determination of line-ratios of transitions originating from both excited states can yield species densities. Such line-ratios are often recorded spatio-temporally averaged and evaluated under the assumption of an averaged EEDF in the detection volume. This can lead to distinct deviations of derived quantities depending on the similarity of involved electron impact excitation cross sections. Accordingly, consideration of the transient spatio-temporal character of the EEDF is necessary in order to minimise systematic errors [51, 52, 53].

In any case, the deduction of plasma parameters from OES requires assumptions on discharge properties that allow to reduce the complexity of underlying collisional-radiative models. Moreover, the use of such models requires the knowledge of cross section data for involved processes. However, the database of experimentally and theoretically derived cross sections is very limited. Apart from that, values for cross sections of the same processes can deviate by orders of magnitude comparing results published by different research
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groups. Despite this complexity, OES often is the only applicable diagnostics for the investigation of microplasmas as their small dimensions do not allow to use invasive probes.

**Phase resolved optical emission spectroscopy**

In order to investigate emission dynamics of AC driven plasmas within one excitation period, it is necessary to observe respective fluorescence signals on a very short timescale. In particular, for microplasmas and the accompanying small detection volume, the low fluorescence yield on short time scales can remain below or close to the detection limit even of intensified sensors such as photomultiplier tubes, diodes, and cameras. The higher the desired temporal, spectral or spatial resolution, the less fluorescence yield can be used for detection. Nevertheless, in some cases high resolutions are indispensable.

Here, a feature of homogeneous AC driven plasmas can be exploited: After a short settling time of typically a few excitation periods, this kind of plasmas reach a dynamic equilibrium. Hence, resulting emission shows a periodic behaviour synchronised to the driving frequency. In particular, the fluorescence signal is the same for same phasing within the excitation period.

Gating of the fluorescence detector with a fixed gate width and a variable delay, phase-locked and synchronised to the driving frequency, allows a high temporal resolution within the excitation period at high photon yields when the fluorescence signal for each phasing is integrated over several excitation periods. This diagnostics is called ‘phase resolved optical emission spectroscopy’ (PROES). However, non-periodic processes such as streamers or other statistical effects cannot be observed by means of PROES. Figure 2.4 shows a schematic diagram of the basic PROES principle.

To achieve high temporal and spatial resolution, a charge-coupled device chip combined with a fast repetitively gateable intensifier in form of a microchannel plate and an optical system can be utilised [41]. Additional spectral discrimination can be realised using filters inserted into the optical path.
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![Schematic diagram of the realisation of phase resolved optical emission spectroscopy. A variable delay \( d \) determines the point within the excitation period when the camera is repetitively gated with a set gate width.]

2.2. Atmospheric pressure dielectric barrier discharges

Dielectric barrier discharges, also referred to simply as barrier discharges or silent discharges, have been first reported on by W. Siemens in 1857 [54]. He designed a glass discharge tube with exterior electrodes for the purpose of ozone generation. While ozone generation remained the major large-scale application of DBDs for several decades, many different fields of application and research of such discharges emerged until now, taking advantage of the low-temperature character of these discharges. Today’s application of atmospheric and elevated pressure DBDs ranges from ozone generation for disinfection of drinking water [55] over decomposition of gaseous pollutants such as oxides of nitrogen [56] or sulphur [57] as well as volatile organic compounds [58], abatement of greenhouse gases [59], surface treatment and modification (e.g. increase of surface energy) [60], CO\(_2\) laser sources [61], ultraviolet excimer and fluorescent lamps [62, 63] to display panels [64]. Further applications and details can be found in the literature [65].

Typical parallel plate DBD configurations are sketched in figure 2.5. These discharges are characterised by the insertion of at least one dielectric barrier (grey) in the discharge gap between the electrodes. Thus, they have to be op-
2. Fundamentals

Typical parallel plate DBD configurations: AC voltage is applied over a discharge gap, where one or both electrodes are covered with a dielectric (grey) and a dielectric barrier inhibits direct current between the electrodes respectively.

erated at alternating voltages as the insulating character of the dielectric does not allow direct currents. While parallel plate geometry is most simple, many different kinds of geometries have been realised so far, comprising, for example, cylindrical setups. However, most designs can be reduced to parallel plate geometry for general description of DBDs. If the employed dielectrics are not symmetrically aligned in the discharge gap or do not exhibit the same characteristics, the discharge will show asymmetric behaviour in dependence of the applied voltage polarity. The advantage of the dielectric barrier design results from the current limitation and protection of the electrode surface.

Favoured materials used as dielectric barriers include glass or quartz, ceramics as well as thin enamel and polymer layers. The dielectric constant and thickness of the barrier as well as the temporal derivative of the applied voltage waveform determine the displacement current in the barrier. As no DC current is passed through the barrier, current transport through the discharge gap is only possible if a breakdown occurs in the gas. At higher frequencies, the current limitation by the dielectric barriers becomes less effective due to the capacitive character of the latter. This natural limitation leads to typical operating frequencies between some hertz and a few megahertz. As most DBDs are operated at or close to atmospheric pressure, typical discharge gap sizes are in the range of a few micrometres up to several centimetres. These gap sizes require operating voltages in the order of some hundred volts up to kilovolts in order to ignite a discharge. In the following, typical discharge modes of atmospheric pressure DBDs are discussed for parallel plate geometry.
2. Fundamentals

2.2.1. Filamentary mode

Most atmospheric pressure DBDs are found to exhibit distinctive short-lived current filaments [66] as electrical breakdown occurs in form of these so-called microdischarges. This filamentary breakdown originates from electron avalanches launched in the discharge gap. Due to the highly collisional regime at atmospheric pressure, considerable charge densities can build up at the front of these avalanches. Sufficiently high pressure and gap size provided, high amounts of ions ($\sim 10^8$) [67] can be created without the need of secondary electron generation from surfaces or Penning ionisation through metastable species. Charge separation due to different drift velocities of electrons and ions can then lead to establishment of a space charge field that superposes the applied field and further accelerates electrons between cathode and the positive space charge. In this region, collisional ionisation further abets streamer formation and propagation of the avalanche and forms a thin and bright plasma channel. Additionally, pre-ionisation caused by ultraviolet photons from the plasma channel is supposed to contribute to the streamer propagation. Typical streamer radii are in the order of hundred micrometres up to some millimetres with propagation velocities of several ten thousand metres per second [66]. As soon as a streamer bridges the discharge gap, a microdischarge evolves and its conductivity leads to charge deposition on the dielectric surfaces, where an area of surface discharge considerably larger than the filament diameter forms. The resulting accumulating surface charge weakens the local electric field until the microdischarge eventually extinguishes, leaving behind residual surface charges. Thus, additional microdischarges are preferably initiated at new locations of comparably higher electric field during the rising part of the applied voltage. Upon voltage reversal, sufficiently high driving frequency provided, locations of prior microdischarges are preferred starting points for successive build-up of streamers of reverse polarity due to remanent surface charges which locally reduce the required voltage swing for a new breakdown. Typical lifetimes of such filaments in DBDs are below 100 ns [67].

Consequently, resulting filament pattern formation is mainly dependent on applied voltage and frequency, where high voltage and low frequency promote more statistical behaviour and vice versa. Higher frequencies may additionally influence filament formation in form of residual excited and charged species in the discharge gap.

Figure 2.6 shows a photograph of microdischarge filaments as observed in an
2. Fundamentals

While breakdown occurs in form of streamers, a single plasma filament can be described as a transient glow discharge which exhibits a distinct cathode fall and a positive column [69] and typical electron densities of $10^{14}$ to $10^{15}$ cm$^{-3}$ and current densities of 100 to 1000 A cm$^{-2}$ [65]. The streamer channel radii are mainly dependent on the gas density and ionisation properties of the gas [65]. Thus, under certain experimental conditions, filamentary discharges can exhibit a homogeneous emission intensity distribution caused by either overlapping streamers or instantaneous breakdown of the latter. However, in this study such discharges are referred to as filamentary to distinguish them from discharges in a second stable homogeneous and streamer-free mode of DBDs: The so-called ‘homogeneous mode’.

Figure 2.6.: Photograph of microdischarge filaments in an atmospheric pressure DBD as published by Kogelschatz et al. [68]. It was recorded through a transparent electrode of a parallel plate setup in a 1 mm air gap with an exposure time of 20 ms (original size: 6 cm x 6 cm).
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2.2.2. Homogeneous mode

While DBD breakdown in form of streamers, as previously discussed, leads to formation of filamentary discharges, it is also possible to achieve Townsend breakdown behaviour which leads to homogeneous discharges [67, 70]. A prerequisite for Townsend breakdown is a rather slow gas bulk ionisation and ionisation at low $E/N$ values respectively. This allows generated ions to reach the cathode in a space charge-free field before a streamer can evolve and to trigger the release of secondary electrons. The latter can then contribute to sustainment of the discharge. Here, the ratio of ionisation in the gas bulk to secondary electron emission at the cathode is determined by the ratio of the electron mean free path to the discharge gap size. Consequently, for small values of the product of gas pressure $p$ and discharge gap size $d$ Townsend breakdown is most probable. Typical $pd$ values are smaller than 30 Torr cm, while streamer breakdown usually occurs at values larger than 200 Torr cm [67].

Terminology concerning homogeneous DBDs in the literature is ambiguous. For convenience, this study coincides with definitions established by Massines et al. [67], Radu et al. [70] and Bartnikas et al. [71]. Two substantially different homogeneous DBD modes can be distinguished: Atmospheric pressure Townsend-like discharges (APTD) and atmospheric pressure glow discharges (APGD). Both are characterised by a strong periodicity of voltage and current as well as emission intensity and coupling between half periods of the excitation cycle. While ignition processes are similar for both types of discharges, their plasma parameters and behaviour deviate. In the following, the term ‘half periods’ always refers to phasing within the excitation period under the assumption that one electrode is powered and the second electrode is grounded. The typical discharge behaviour of homogeneous atmospheric pressure DBDs is described for equilibrium conditions. The first onset of the discharges can vary and transform from a filamentary discharge to a homogeneous DBD as will be discussed in section 2.2.2.5.

2.2.2.1. Atmospheric pressure Townsend-like discharge

The APTD is characterised by a low ionisation level which does not allow the formation of a quasi-neutral plasma bulk. Hence, the electric field in the discharge gap is virtually undisturbed and equates the vacuum electric field. This
2. Fundamentals

Voltage and current oscillograms as measured (a) and simulated (b) by Naudé et al. [72] for a nitrogen APTD in parallel plate geometry of 1 mm discharge gap operated at 6 kHz bipolar sinusoidal excitation and 15 kV peak-to-peak voltage, where both electrodes are covered with a dielectric. Displayed quantities comprise discharge current $I_{\text{discharge}}$, displacement current $I_{\text{Cgas}}$, their sum and measured current respectively $I_g$, applied voltage $V_{\text{PS}}$, gap voltage $V_{\text{gas}}$ and voltage across the dielectrics $V_{sd}$.

This kind of discharge characteristically exhibits one current pulse in the first half of each half period. An emission pulse coincides with each current pulse. Naudé et al. [72] extensively studied a nitrogen APTD and found typical current density amplitudes and pulse durations in the order of a few mA cm$^{-2}$ and some $10 \mu$s respectively for a parallel plate setup of 1 mm discharge gap operated at excitation frequencies of a few kHz and peak-to-peak voltages in the order of 10 kV, where both electrodes were covered with a dielectric. They used an equivalent circuit model to simulate the APTD and the employed power supply. In order to discuss the typical behaviour and dynamics of the APTD, measured (a) and simulated (b) voltage and current oscillograms of that discharge are illustrated in figure 2.7.

The discharge dynamics are divided into three phases as indicated: The discharge is off (I), ignition (II) and Townsend discharge (III). For a voltage swing of the applied voltage $V_{\text{PS}}$ and gap voltage $V_{\text{gas}}$ after discharge extinction (phase I), the registered current $I_g$ is dominated by displacement current $I_{\text{Cgas}}$. The phase shift of gap voltage and applied voltage is due to residual electrons and ions which originate from the antecedent discharge of reverse polarity and are adsorbed at the dielectric surfaces. The resulting potential superposes the applied voltage and leads to an increased voltage drop across the dielectric layers. This implies a reduced gap voltage. A comparably negligible
discharge current $I_{\text{discharge}}$ caused by electron emission triggered by impinging metastable species at the cathode is also observed (not shown here). Once the gap voltage passes the breakdown voltage (phase II), the discharge current strongly increases and reaches its peak value of approximately 6 mA within 10 µs. For increasing gap voltage, the secondary electron yield at the cathode and associated electron collection at the anode becomes more effective due to higher kinetic energies of ions. Thus, the discharge current increases exponentially with the gap voltage. The voltage across the dielectrics $V_{sd}$ decreases, leading to a comparably smaller electron density on the dielectric surface of the cathode. This implies a reduced secondary electron yield [73] and leads to a saturation behaviour of the discharge current which reaches its maximum when the voltage across the dielectrics vanishes (phase III). At this point, the gap voltage almost remains constant and further increase of the applied voltage mirrors in an increase of the voltage across the dielectrics. This originates from the increased conductivity and thus reduced resistance of the plasma as well as from accumulating charge carriers which adsorb at the dielectric surfaces as described above.

As the discharge current in this phase mainly depends on the derivative of the applied voltage [72], the former slowly decreases and the discharge eventually extinguishes. For the next half period this evolution repeats for reverse polarities.

2.2.2.2. Pulsating Townsend mode

Another form of Townsend discharge in atmospheric pressure DBDs discussed is characterised by several small current peaks during the first half of each half period of the excitation frequency. Experimentally observed [74] (a) and simulated (b) voltage and current oscillograms as published by Golubovskii et al. [75] are exemplarily shown in figure 2.8. The data was recorded and simulated for a pulsating helium APTD in parallel plate geometry of 2 mm discharge gap operated at 1.5 kHz bipolar sinusoidal excitation and 2.5 kV peak-to-peak voltage under a helium gas flow of 4 sccm, where both electrodes are covered with a dielectric. Here, the gas flow was chosen to ensure a defined and reproducible gas atmosphere.

For increasing gap voltage, a breakdown event occurs for sufficient voltage drop across the gap causing a current peak and a drop of the gap voltage. When the current decreases, a further increase of the gap voltage leads to a
2. Fundamentals

Voltage and current oscillograms as measured (a) by Visentin et al. [74] and simulated (b) by Golubovskii et al. [75] for a pulsating helium APTD in parallel plate geometry of 2 mm discharge gap operated at 1.5 kHz bipolar sinusoidal excitation and 2.5 kV peak-to-peak voltage and with helium gas flow of 4 sccm, where both electrodes are covered with a dielectric. Displayed quantities comprise applied voltage (dash dotted line), gap voltage (solid line), and discharge current (dotted line).

Figure 2.8: Voltage and current oscillograms as measured (a) by Visentin et al. [74] and simulated (b) by Golubovskii et al. [75] for a pulsating helium APTD in parallel plate geometry of 2 mm discharge gap operated at 1.5 kHz bipolar sinusoidal excitation and 2.5 kV peak-to-peak voltage and with helium gas flow of 4 sccm, where both electrodes are covered with a dielectric. Displayed quantities comprise applied voltage (dash dotted line), gap voltage (solid line), and discharge current (dotted line).

second breakdown and current peak of smaller amplitude respectively. This behaviour repeats until the vertex of the applied voltage waveform.

Simulations [75] show that the discharge behaviour for a single current peak is the same as described for the APTD above while the pulsing of the discharge can be attributed to dielectric properties in combination with gap size as well as excitation frequency and voltage. The underlying numerical one-dimensional fluid model is based on the continuity equations for electrons, ions, excited atoms, and excimer molecules as well as Poisson’s equation. Upon initiation of the incipient current pulse, the only source of primary electrons is shown to be desorption of previously adsorbed electrons at the cathode surface. These electrons lead to an avalanche breakdown and a respective current pulse. During the current pulse, a noteworthy ion density builds up near the cathode, exceeding the electron density and disturbing the electric field in the gap. No quasi-neutral plasma is formed. While the electron distribution is assumed to form instantaneously, the comparably slow ions generated near the cathode can release secondary electrons upon striking at the anode. Here, the time lag between ion generation near the cathode and related subsequent secondary electron emission at the anode lead to the pulsation of the current
which is mainly carried by the electrons. While the damping factor of the oscillations grows with increasing electron desorption, a domination of the latter over ion-induced secondary electron emission leads to an APTD without oscillations. In the case of this simulation, the total absence of electron desorption no longer yields an APTD but transition to an APGD [76]. Moreover, the space charge of ions could influence the oscillations of the current. It distorts the electric field and therefore changes the multiplication coefficient for electrons, resulting in the case of the simulation in an arrangement of the electric field and particle densities in form of waves in space and time. In their study, Golubovskii et al. moreover found that the oscillation frequency of the discharge current in a pulse train of a pulsating Townsend discharge amongst others depends on the growth rate of the external voltage \(\frac{dU}{dt}\).

The oscillograms in figure 2.8 show a significant decrease of the gap voltage during current peaks which distinguishes the pulsating Townsend discharge from the APTD. This behaviour is observed experimentally as well as reproduced in the simulations. This would suggest comparably higher ionisation levels leading to noticeable charging of the dielectric surfaces which somehow contradicts a Townsend discharge. However, the simulations yield a non-quasi-neutral Townsend discharge.

### 2.2.2.3. Atmospheric pressure glow discharge

While the general ignition mechanism of an APGD is similar to that of an APTD and follows typical Townsend breakdown as described above, the voltage-current characteristics of the discharge after onset deviate significantly. Figure 2.9 exemplarily shows measured (a) and simulated (b) voltage and current oscillograms of a helium APGD as published by Massines et al. [77]. The discharge was operated in a parallel plate setup of 5 mm discharge gap and powered by a bipolar sinusoidal excitation of 10 kHz and 1.5 kV peak-to-peak voltage, where both electrodes were covered with a dielectric. In the following, the typical discharge behaviour of an APGD is discussed on the basis of these measurements.

Before discharge ignition, the gap voltage is higher than the applied voltage. Again, this is due to residual charges which are adsorbed at the dielectric surfaces like for the APTD. The voltage across the dielectrics is referred to as ‘memory voltage’ in figure 2.9 and equates to \(V_{sd}\) in terminology used beforehand for the APTD in figure 2.7. Thus, a rather small increase of the applied
2. Fundamentals

Voltage and current oscillograms as measured (a) and simulated (b) by Massines et al. [77] for a helium APGD in parallel plate geometry of 5 mm discharge gap operated at 10 kHz bipolar sinusoidal excitation and 1.5 kV peak-to-peak voltage, where both electrodes are covered with a dielectric.

Figure 2.9.: Voltage and current oscillograms as measured (a) and simulated (b) by Massines et al. [77] for a helium APGD in parallel plate geometry of 5 mm discharge gap operated at 10 kHz bipolar sinusoidal excitation and 1.5 kV peak-to-peak voltage, where both electrodes are covered with a dielectric.
2. Fundamentals

Figure 2.10.: Voltage and current oscillograms as measured and published by Radu et al. [70] for a helium pseudoglow discharge in parallel plate geometry of 0.5 mm discharge gap operated at 2 kHz bipolar sinusoidal excitation and 600 V peak-to-peak voltage, where one electrode is covered with a dielectric.

ignited. For each current pulse, a discontinuity of the otherwise sinusoidal applied voltage waveform is observed in figure 2.9. This voltage drop due to the discharge current depends on the difference of the residual voltage in the gap and the breakdown voltage as well as the power supply specifications. Upon reversal of the gap voltage, a comparably small current peak can be observed which is caused by electrons previously trapped in the positive column [77]. Prerequisites for this observation are a fully established positive column of sufficient charge carrier density as well as a sufficiently high excitation frequency. Thus, this cannot be observed in APTDs.

2.2.2.4. Pseudoglow discharge

Under certain conditions, another form of homogeneous atmospheric pressure DBD can be observed. It exhibits a multitude of short current and emission pulses respectively for each half period of the excitation frequency. They are referred to as ‘pseudoglow discharges’, as introduced by Bartnikas for pulsating helium discharges under AC excitation in a bare-electrode parallel plate gap [71], where similar phenomena could be observed. However, this term has established for atmospheric pressure DBDs exhibiting the described behaviour. Typical oscillograms of voltage and current of a helium pseudoglow discharge as measured by Radu et al. [70] in parallel plate geometry of 0.5 mm discharge gap for bipolar sinusoidal excitation of a frequency of 2 kHz and 600 V peak-to-peak voltage are exemplarily shown in figure 2.10.
It is obvious that the total current registered is composed of the displacement current which is superposed by the discharge current pulses. For an increase of the applied voltage, a discharge current pulse evolves as soon as the breakdown voltage drops across the discharge gap, just like it is the case for an APGD. While the same mechanisms lead to a discharge extinction, the following increase of the gap voltage is sufficient to initiate a second current pulse and discharge respectively. This behaviour repeats for subsequent voltage increments until the reversal of the slope of the applied voltage and several pulses can evolve. However, the initiation of the current pulses is significantly influenced by preceding current pulses of the same voltage polarity within the same half period. In an APGD, subsequent breakdowns evolve in the respective next half period and excited species and metastable concentrations built up during the preceding breakdown can considerably diminish until the ensuing breakdown. In contrast, the ionisation behaviour of subsequent breakdowns within the same half period is strongly coupled in a pseudoglow discharge. Thus, the increase of the gap voltage needed to lead to breakdown for follow-up pulses within a pulse train is successively reduced compared to the breakdown voltage of the preceding pulse in each half period. Higher metastable and excited species concentrations can increase the ionisation rate through Penning ionisation of present impurities and lead to breakdown at significantly lower $E/N$ values compared to neutral gas. Bartnikas et al. observed increasing rise times and pulse widths as well as reduced magnitudes for successive current peaks in a pulse train, indicating a regular increase of free electron supply, supporting the above statement [78, 79]. Every single breakdown in the pulse train leads to the formation of a glow discharge, so that the pseudoglow discharge can be essentially described as a pulsating glow discharge. The specificity of the formation of subsequent breakdowns implies that the number of pulses per pulse train increases with increasing applied peak-to-peak voltage [70].

As the first successive breakdown in the following half period requires a fixed change of the voltage across the gap, a comparably higher peak-to-peak voltage results in incipient breakdowns at earlier phasing within the excitation period. Thus, the following voltage increase until its vertex allows additional breakdowns. This is a distinctive feature of the pseudoglow discharge and distinguishes it from the APGD which only allows one current peak per half period. For an increasing number of pulses per pulse train, the formation of a quasi continuous glow discharge and its superposition upon the pseudoglow
2. Fundamentals

pulses can be observed [70]. Here, the glow discharge formation is a direct consequence of the increased ionisation rate within the gap due to the temporal proximity of the single pulses.

Moreover, Radu et al. observed a decreasing number of pulses per pulse train for increasing driving frequencies [70], where also width and rise time of individual pulses diminish. They relate this to higher overvoltages upon breakdown and lower metastable species concentrations due to the comparably short pulse widths for higher frequencies. Hence, they explain the reduced number of pulses per pulse train by higher needed overvoltages for breakdown of incipient as well as higher voltage increments to initiate subsequent pulses culminating in the observed behaviour.

It was also found for pseudoglow discharges, that the interval between successive current pulses depends on the voltage rise waveform [70]. While each successive discharge in a pulse train exhibits higher numbers of free electrons as well as metastable and long-lived excited species compared to preceding discharges, subsequent discharges can be initiated at comparably lower voltage rises. Thus, the growth rate of the applied voltage has significant influence on current pulse intervals.

The role of impurities was investigated as well [70] and it was found that small amounts in the order of 0.1% vol. of nitrogen, argon and hydrogen allow to increase the number of pulses per pulse train. The impurities can be effectively ionised by Penning ionisation through metastable process gas species and thus increase the ionisation rate and lower breakdown voltages respectively. Higher amounts of impurities, however, lead to a transition into spark discharge onset. If they exceed a certain threshold, the quenching of metastable species by impurities outweighs Penning ionisation and prevents Townsend breakdown.

2.2.2.5. Memory effect

For all homogeneous atmospheric pressure DBD modes, a pulsed discharge which exhibits a strong periodicity of the discharge current with respect to the excitation frequency is observed. All discharge modes show a discharge in the first half of each half period of the excitation frequency only. Thus, each discharge matches a DC discharge, where species formed in the preceding
half period and discharge pulse respectively influence the breakdown behaviour. Whereas the gap voltage is influenced by charge carriers adsorbed at the dielectric surfaces, the ionisation rate in the volume strongly depends on metastable and excited species concentrations. This influence of prior half periods on successive half periods is called ‘memory effect’. The importance of this effect for discharge stability and Townsend breakdown respectively becomes obvious in simulations which show that the first discharges after ignition show filamentary discharge behaviour, before a stable homogeneous Townsend breakdown can occur in subsequent discharges [67] due to the successive build-up of metastable species which accumulate from half period to half period. This can be exploited to stabilise atmospheric pressure DBDs and to trigger Townsend breakdown. While the choice of the dielectric material can influence ionisation rates due to secondary electron emissivity, systematic admixture of impurities can enhance the bulk ionisation rate which leads to breakdown at lower $E/N$ values and thus enlarge the Townsend breakdown regime. The species prevailing until the next half period then significantly influence the subsequent discharge. Apart from exploiting the memory effect, a resistor in series with the discharge can help to limit discharge currents and prevent streamer formation.

2.2.2.6. Distinction of discharge modes

The distinction between APTD and APGD including their respective pulsating modes is essential for the interpretation of experimental observations.

Most easy to access are parameters such as current peak widths and phasing within the excitation period. While for APTDs typical current peak widths are in the range of several 10 µs, those for APGD are shorter (~ 1 µs). Both discharges exhibit a strong periodicity of the current with respect to the excitation period. The current density of APGDs (> 10 mA cm$^{-2}$) typically exceeds that of APTDs (< 10 mA cm$^{-2}$) by an order of magnitude. Moreover, the gap voltage in APGDs usually decreases significantly during a current peak, whereas in APTDs it reaches a plateau level [80]. This is due to the comparably higher ionisation level and charge densities respectively in APGDs which allow a noticeable reduction of the gap voltage due to adsorbed charge carriers at the dielectric surfaces. However, pulsating Townsend discharges are reported to exhibit a significant voltage drop upon current peaks so that the distinction from pseudoglow discharges solely on the basis of gap voltage behaviour may
2. Fundamentals

not be possible. For filamentary discharges, the current peaks are in the order of a few 10 ns and appear statistically in the first half of each half period of the excitation frequency.

If the gap voltage and discharge current are accessible, the evaluation of voltage-current characteristics for single current peaks allows to identify the formation of a cathode fall and the transition from Townsend to subnormal glow discharge in form of a decreasing gap voltage for increasing discharge current after passing the Townsend plateau [67, 80, 81]. As single current peaks and associated discharges can be treated like DC discharges with remanent species concentrations due to the memory effect and preceding discharges respectively, this scheme can be applied and transferred to AC driven atmospheric pressure DBDs.

Some authors employ voltage/charge Lissajous figures to distinguish discharge regimes [65, 82]. For homogeneous discharges with one or more current pulses, the respective Lissajous figure is a parallelogram with distinct steps at the vertical edges, mirroring the discrete current pulses and associated charge transfer. For filamentary discharges, the Lissajous figure is a regular parallelogram, due to the statistical occurrence of the filaments. However, this technique does not allow to definitely distinguish between APTD and APGD, although charge transfer during current peaks is inherently more pronounced for the latter.

If optical access allows to identify the formation of a cathode fall and positive column respectively during the current peak, fast intensified charge-coupled device camera (ICCD) imaging can be used to distinguish between APTD and APGD. This is only given for sufficient gap size or optically transparent electrodes. Even if no positive column is observed, ICCD camera imaging can help to distinguish between filamentary and homogeneous discharges. However, fast single shot cameras with gate times of some 10 ns are a prerequisite to observe and resolve filaments respectively.

Plasma parameters such as electron density, which is typically higher in APGDs compared to APTDs, can be used to identify discharge modes as well. Certainly, invasive probes can only be used in rather large discharge configurations, whereas non-invasive optical emission spectroscopy may in certain cases allow deduction of plasma parameters as well. The underlying collisional-radiative models may be restricted to and the observation of certain effects only possible for certain parameter ranges. For example, the electron density
can be deduced from Stark broadening of H$_\alpha$ or H$_\beta$ emission lines of small hydrogen admixtures, sufficient electron density provided [50]. In atmospheric pressure low-temperature argon discharges, the electron density can, within certain limits, be calculated from line-ratio measurements of the 2p levels [83]. Another approach for such argon discharges consists of the determination of the electron density from broadening of argon emission lines of high energetic argon levels which are susceptible to quadratic Stark broadening [84]. However, at atmospheric pressure, this effect requires sufficient electron densities of $\sim 10^{16}$ cm$^{-3}$ in order to be distinguished from pressure broadening.

For DBD discharges, typical electron densities are smaller (see table 2.1 and figure 4.18). Both, optical and probe measurements require time resolutions of at least 1 µs to be able to resolve the discharge evolution within one current pulse as for APGDs the actual subnormal glow is reached shortly before extinction of the discharge and abortion of the current pulse respectively. The time interval allowing to observe respective plasma parameters is thus very limited. Time-averaged measurements can only yield limited information on discharge regimes as homogeneous atmospheric pressure DBDs are by nature pulsating discharges almost never exceeding duty cycles of 50%.

For a better comparison of APTDs and APGDs, typical orders of magnitude for different plasma parameters as published by Massines et al. [67] are in summary opposed in table 2.1. These classifications can only give a rough idea of typical discharge regimes and are not strict definitions.
Table 2.1: Typical orders of magnitude of different plasma parameters for APTDs and APGDs as published by Massines et al. [67].

<table>
<thead>
<tr>
<th></th>
<th>APTD</th>
<th>APGD</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum electron density [cm$^{-3}$]</td>
<td>$10^7 – 10^8$</td>
<td>$10^{10} – 10^{11}$</td>
</tr>
<tr>
<td>Maximum ion density [cm$^{-3}$]</td>
<td>$10^{10}$</td>
<td>$10^{11}$</td>
</tr>
<tr>
<td>Neutral plasma formation</td>
<td>no</td>
<td>yes (positive column)</td>
</tr>
<tr>
<td>Metastable density [cm$^{-3}$]</td>
<td>$10^{13}$</td>
<td>$10^{11}$</td>
</tr>
<tr>
<td>Current density [mA cm$^{-2}$]</td>
<td>0.1 – 10</td>
<td>10 – 100</td>
</tr>
<tr>
<td>Gas voltage variation at the current maximum</td>
<td>constant (Townsend plateau)</td>
<td>decrease (cathode fall formation)</td>
</tr>
<tr>
<td>Process gas</td>
<td>N$_2$, air, N$_2$O</td>
<td>Penning mixtures in He, Ar, Ne</td>
</tr>
<tr>
<td>Discharge gap size</td>
<td>&lt; 2 mm</td>
<td>&gt; 2 mm</td>
</tr>
<tr>
<td>Driving frequency</td>
<td>&lt; 10 kHz</td>
<td>&gt; 1 kHz</td>
</tr>
<tr>
<td>Power density for excitation at 10 kHz</td>
<td>~ W cm$^{-3}$</td>
<td>~ 0.1 W cm$^{-3}$</td>
</tr>
<tr>
<td>Approach to resolve firmaments</td>
<td>current, optical</td>
<td>PMT, ICCD</td>
</tr>
</tbody>
</table>
3. Setup

3.1. Discharge chamber and gas supply

The discharge chamber used for experiments in the frame of this thesis consists of a cylindrical stainless steel housing of 25 cm inner diameter which is uprightly mounted on an experiment table. The end planes are each made of 13.25 inch ConFlat (CF) flanges, whereas the top flange is designed as removable cover with handle bars to access the inner part. It is sealed with a reusable viton gasket. The lateral area of the discharge chamber is equipped with several CF flanges of different sizes in a 45°-geometry with respect to the axis of symmetry for diagnostic and optical access. Discharge chamber and gas supply are sketched in figure 3.1.

A pumping system consisting of a diaphragm vacuum pump (Pfeiffer MD 4TC) and a turbomolecular drag pump (TMP, Pfeiffer TMU 520 PC) allows to evacuate the discharge chamber to a final pressure of $10^{-6}$ mbar. It is mounted to the bottom flange of the discharge chamber. A pneumatic gate valve (VAT series 10.8) can separate the chamber from the pumping system. A wire mesh guard in front of the valve avoids access of particles to the pumping system.

Pressure is monitored by a combination of an active Pirani gauge (Edwards APG-MP-16-ST, $10^{-4}$ mbar) and an active inverted magnetron gauge (Edwards AIM-S-NW25, $10^{-2} - 10^{-8}$ mbar) in the low pressure regime and by a capacitive gauge (Vacuubrand DVR 5, 0.1 - 1100 mbar) in the range of a few mbar up to atmospheric pressure.

Due to the high spatial resolution and small focus length of the optical setup (see section 3.3), a precise alignment of investigated devices is important. A guide rail is installed at the bottom of the discharge chamber. It can be horizontally rotated and locked in 45° positions perpendicular to the access flanges. A manual three-orthogonal-axis manipulator is mounted on a sliding bed and allows positioning of investigated devices within the discharge chamber. An additional piezoelectric high-precision relative positioning system is mounted...
on top of this manipulator. It consists of a translational three-orthogonal-axis (mechonics MX 35) and a two-axis tilt (mechonics MT 25) positioner and is capable of translational steps of 30 nm and rotational steps of 0.3 µrad. Investigated devices can either be fixed uprightly or horizontally on the positioning system. It is used to align the devices normal or parallel to the optical axis of a camera system.

A high voltage feedthrough flange is used to ensure power supply in the discharge chamber.

The gas feed is realised by means of a feedthrough flange close to the top flange of the discharge chamber. Process gas is provided by an argon gas cylinder through a gas line whereat the gas used exhibits impurities less than $6 \cdot 10^{-5}$. Gas addition is controlled by a valve. It can be by-passed for quick evacuation of the system as well as cut-off using additional quarter-turn plug valves. The whole gas line system consists of 6 mm stainless steel tubes and respective components compatible with Swagelok fittings. The TMP and the whole system respectively can be vented with laboratory dry air.

An optical fibre feedthrough flange is installed to enable fibre optic measurements inside the discharge chamber.
3. Setup

3.2. Power supply

An electric circuit diagram for the setup used in the frame of this thesis can be found in figure 3.2. A commercial arbitrary function generator (Tektronix AFG 3021B) is connected to a broadband amplifier (FM Elektronik DCU 600-40 HF) using a 50 Ω BNC cable. The amplification of the broadband amplifier can be controlled steplessly. The amplified signal is directly connected to the high voltage feedthrough flange via measuring lines. Inside the discharge chamber, flexible copper wires (3 mm diameter) with insulating varnish are used to connect investigated devices via clip terminals. The copper wires are additionally covered with ceramics protection to prevent short circuits or discharges in between.

In any case, the devices are powered using a bipolar triangular voltage waveform to keep $dU/dt$ constant, where the Ni grid serves as powered electrode and the Si wafer is grounded. The choice of a bipolar triangular excitation waveform is motivated by the strong dependence of the current pulse frequency in pulse trains observed in both, pulsating Townsend and pseudoglow mode of atmospheric pressure DBDs (see section 2.2.2). By choosing a constant voltage growth rate within each half period of the excitation frequency, the influence of the applied voltage waveform on current pulse frequencies within a current pulse train can be neglected. It was shown that general discharge behaviour of atmospheric pressure DBDs is comparable for bipolar triangular and sinusoidal excitation [74], so that results of this thesis are representative for both kinds of excitation. In the following, the excitation period is divided into positive ($pHP$) and negative ($nHP$) half period referring to respective Ni grid polarities.

Commercial capacitive voltage (Tektronix P6015A) and inductive current (Tektronix P6021) probes are inserted between amplifier and feedthrough flange. Measured voltage and current are monitored and recorded by a digital storage oscilloscope (DSO, Hewlett-Packard HP 54540C).
3. Setup

3.3. Optical setup

3.3.1. Photomultiplier tube and USB spectrometer

A sketch of the optical setup used in the frame of this thesis is shown in figure 3.3. The discharge chamber is equipped with two BK7 window flanges for optical access to investigated devices. To record time-dependent spectrally integrated optical emission behaviour, a photomultiplier tube (PMT, Hamamatsu R3896) with spectral response between 185 nm and 900 nm is installed in front of one of these flanges. The emission signal is imaged onto the PMT using a magnesium fluoride (MgF$_2$) lens doublet and monitored and recorded by the DSO.

A digital USB grating spectrometer (Ocean Optics HR 4000) is used to monitor optical emission spectra of investigated devices. The spectrometer has an effective spectral resolution of 0.22 nm and a detector range of 200 - 1100 nm. An f/2 fused silica collimating lens (Ocean Optics 74-UV, 200 - 2000 nm) is used to focus optical emission of investigated devices onto the spectrometer entrance slit. It is installed inside the discharge chamber and connected with the spectrometer via a 200 µm optical fibre using the optical fibre feedthrough flange.
3. Setup

Phase resolved optical emission spectroscopy on investigated devices is performed using an intensified charge-coupled device (ICCD) camera (LaVision PicoStar HR 16) with high gating rate. Investigated devices are imaged onto the ICCD chip (512x512 pixels) via a long-distance microscope (LDM, Questar QM1). This setup allows an effective maximum spatial resolution of 2 µm per pixel when additional lenses (1.5x and 2x Barlow) are inserted between camera and LDM. Both, the ICCD camera as well as the LDM are mounted on a guide rail, allowing to vary the working distance from the centre of the discharge chamber between 0.56 m and 1.52 m, corresponding to a numerical aperture of 0.0288 and 0.0580 of the LDM. This way, stepless control of the magnification is possible up to 12 times at the image plane. The whole camera setup and optical path are housed in a light-proof black polyoxymethylene (POM) housing to prevent the detection of stray light.

Figure 3.4 illustrates the configuration of the gateable intensifier of the employed ICCD camera. Photons entering the camera through the entrance window induce electrons at a photocathode due to the photoelectric effect.

3.3.2. PROES setup
3. Setup

Configuration of the gateable intensifier of the ICCD camera.

Figure 3.4: Configuration of the gateable intensifier of the ICCD camera.

A switchable voltage between photocathode and the so-called ‘microchannel plate’ (MCP) makes the intensifier gateable. When shut, a positive potential (+3 V) repels electrons while a negative potential (-22 V) accelerates the electrons from the photocathode towards the MCP. The MCP consists of a thin (0.5 mm) glass plane with electrodes on both sides and embeds a multitude of (10⁶) parallel channels of 10 µm diameter. These channels are sloped towards the main axis of the MCP in an angle of 8° so that multiple electron-wall-collisions lead to a multiplication of electrons and light transmitted through the photocathode is blocked. The factor of multiplication can be controlled via the applied MCP voltage whereupon a typical MCP voltage of 1 kV results in a multiplication factor of 10³. Secondary electrons produced this way are once more accelerated with 5 - 6 kV before they hit a phosphor layer whose radiation is led to the camera’s CCD chip using fibre optics. A thin aluminium film in front of the phosphor layer serves as a mirror. It reflects backwards emitted light originating from the phosphor layer so that it can also be used for detection. Moreover, it avoids that parasitic light hits the CCD chip. During the integration time of several milliseconds of the CCD chip, the MCP allows gate times of a few 100 ps at gating rates of up to 110 MHz. The intensifier limits the usage of the ICCD camera to detecting photon intensities, while photon energies cannot be distinguished.

Spectral discrimination of the images taken with the ICCD camera can be op-
tionally realised using a liquid crystal tunable filter (CRi VariSpec NIR) in between camera and LDM. This filter is composed of two coupled liquid crystal polarisation filters. The filter is based on linear polarisation filters at the entrance and exit of the filter combined with voltage-dependent alignment of a liquid crystal medium and thus controllable optical activity in between. It has a spectral range of 550 - 1000 nm at a bandwidth of 0.75 nm FWHM and a tuning accuracy of 0.5 nm.

However, during the cause of this study, the ICCD camera turned out to show interference when exposed to monochromatic light which did not allow to use a filter for wavelength discrimination. The interference most probably occurs in between the entrance window and the MCP of the camera, when photons penetrate the entrance window and are reflected by the MCP. This way, an etalon is created.

Gating of the ICCD camera for PROES measurements is realised via a commercial digital delay generator (Stanford Research Systems DG535). The synchronised output of the arbitrary function generator serves as trigger for the delay generator, where the zero crossing of the driving voltage is chosen as synchronisation point. Thus, the delay generator allows to generate a TTL trigger signal of determined pulse width and delay in relation to the respective zero crossing of the driving voltage. This signal is used to externally gate the ICCD camera. As the delay generator does not allow phase delays of $2\pi$, one whole excitation period cannot be recorded using one synchronisation point. Accordingly, the triggered slope of the driving voltage has to be switched after a phase delay of $\pi$ is reached.

The system of ICCD camera, DSO and delay generator is remotely controlled via the GPIB parallel port. This is realised with a software routine which has been developed in the frame of this study. It allows the synchronisation of the enlisted devices and automated recording of PROES image and DSO oscillogram series.

### 3.3.3. Synthesis of PROES plots

The synthesis of so-called ‘PROES plots’ which show the one-dimensionally space and phase resolved evolution of emission intensity is exemplarily shown in figure 3.5.
3. Setup

Synthesis of PROES plots exemplarily shown for one-dimensionally spatially resolved imaging: Images of the emission intensity of a microplasma array section are recorded at different times $t$ within the excitation period. A straight line (red vertical line) is defined and the emission intensity along this line is plotted versus the corresponding time within the excitation period for each image. The emission intensity perpendicular to the defined straight line is averaged over several pixels (green hatched area). The resulting false colour PROES plot shows the emission intensity spatially resolved along the defined straight line and temporally resolved within the excitation period.

Figure 3.5.: Synthesis of PROES plots exemplarily shown for one-dimensionally spatially resolved imaging: Images of the emission intensity of a microplasma array section are recorded at different times $t$ within the excitation period. A straight line (red vertical line) is defined and the emission intensity along this line is plotted versus the corresponding time within the excitation period for each image. The emission intensity perpendicular to the defined straight line is averaged over several pixels (green hatched area). The resulting false colour PROES plot shows the emission intensity spatially resolved along the defined straight line and temporally resolved within the excitation period.
3. Setup

For each transient position within the excitation period, an image of the emission intensity of the device is recorded with the ICCD. Typical gate times are in the order of 100 ns while the phase delay increase for each successive image complies with respective gate times. Thus, an excitation period of 100 µs resulting from a driving frequency of 10 kHz is resolved in 1000 single images. Although PROES allows to record images at very small gate times, reasonable registered intensities can only be acquired by integrating over several excitation periods and accumulating registered intensities at a fixed phasing within the excitation period. Thus, this technique is limited to observation of periodical phenomena while transient processes of statistical nature cannot be detected.

As it is obvious from the multitude of images recorded in one series and the information contained, analysis of this kind of image series requires sophisticated tools. Due to the lack of availability of such tools, an analysis software has been developed in the frame of this thesis. It allows the extensive analysis of PROES image series as well as of single ICCD camera images and data preparation.

Figure 3.5 shows a detail of the graphical user interface of the analysis software. For the assembly of a PROES plot, a straight line (red vertical line) is defined at a fixed position within the images of a PROES series. The evolution of the recorded emission intensity spatially resolved along the straight line is then plotted against the phasing and relative time within the excitation period respectively. The resulting PROES plot shows the recorded emission intensity spatially resolved along the chosen straight line and temporally resolved within the excitation period. Averaging over pixels in the dimension perpendicular to the resolved spatial dimension is possible (green hatched area).

Apart from spatially resolved PROES plots, the software also allows to spatially average over either single pixels and a defined number of adjacent pixels as well as groups of two-dimensionally equidistant pixels including respective neighboured pixels. This allows, for example, to temporally resolve the emission intensity of a single microplasma array cavity or of a group of cavities as well as of the Ni grid surface or of the whole image. These kinds of PROES plots only feature the phase resolved respective emission intensity without spatial resolution.

The ICCD camera setup can also be used to record time integrated images by choosing gate times of several excitation period durations.
4. Results

This chapter describes experimental results and respective interpretations of the investigated MSE array devices. In the first instance, the discharge mode of the devices is determined from voltage and current characteristics as well as PMT measurements which are compared to well-known DBDs in parallel plate geometry. Subsequently, the strong dynamics and collective as well as individual cavity behaviour of the array devices is investigated by means of two-dimensionally spatially resolved PROES. In particular, interaction of adjacent cavities and the dynamics of single cavities and their mutual impact are discussed.

4.1. Discharge characterisation

In the following section, general discharge properties of the investigated microplasma array devices such as voltage-current characteristics and emissivity are examined in dependence of outer parameters such as applied peak-to-peak voltage, excitation frequency, and gas pressure in order to determine discharge operation regimes and modes.

4.1.1. General discharge properties

For all investigated devices, preliminary measurements of typical voltage-current behaviour as well as time-dependent integral optical emission intensity are performed. Each device is operated in pure Ar and voltage, current as well as PMT signal are recorded using the DSO. Recorded characteristics are exemplarily shown in figure 4.1.

The characteristics shown for 50x50 50 µm (a) and 100 µm (b) base pyramidal cavity array devices, for a discharge channel device (c) as well as for a 32x32
Figure 4.1.: Typical U-I-characteristics of 50x50 50 µm (a) and 100 µm (b) base pyramidal cavity array devices, of a discharge channel device (c) as well as of a 32x32 50 µm circular base straight cavity array device. For each device, recorded applied voltage, total current and PMT signal in arbitrary units are displayed.
50 µm circular base straight cavity array device (d) exhibit the same general discharge behaviour.

After passing a certain threshold voltage in the positive half period, the discharge ignites and emission is registered with the PMT. A short interval of some µs in between, the discharge collapses and the emission intensity decreases, before a new emission peak evolves for a slightly increased applied voltage. This pulsing of the discharge stops upon reversal of the slope of the applied voltage. As it is not externally triggered, it is referred to as ‘self-pulsing’ of the discharge. This behaviour repeats for the negative half period.

The simultaneously registered current is dominated by displacement current. Due to the capacitive character of the electrode design of the investigated devices, the displacement current is determined by the derivative of the applied voltage waveform. For each occurring emission pulse, a synchronous current pulse superposing the displacement current is observed, where the current pulse is typically significantly smaller than the displacement current. Current peaks are smaller for devices with less effective discharge area and single cavities respectively.

This general discharge behaviour is observed for all investigated devices. Voltage, current, and emissivity are in the same order of magnitude for comparable operation parameters. Thus, their general comparability and compatibility is ensured. It can be exploited to investigate basic phenomena such as voltage-current-characteristics or the impact of an array arrangement and cavity dimensions on discharge dynamics of Si-based micro-structured electrode array devices.

### 4.1.2. Typical emission spectra

In the frame of this study, all discharges are operated in pure argon atmosphere at or close to atmospheric pressure. Thus, a brief discourse on atmospheric pressure argon discharges is held in the following. Moreover, typical emission spectra of investigated devices are discussed.

For pulsed operation (see figure 4.1) of a low-temperature atmospheric pressure argon discharge, the species present shortly after excitation mainly comprise neutral atoms (Ar), atomic ions (Ar^+), electrons (e^-), and excited atoms in low (Ar^*) and high energetic states (Ar**) as well as metastable states (Ar^m). Small
impurities of the process gas may also be present. Here, nitrogen plays the most important role. Thus, excited and ionised states of molecular and atomic nitrogen will be present. However, their densities are comparably small but can still play a role in energy transfer. Argon excimer (Ar₂⁺) and excimer ion (Ar⁺₂) densities may build up due to three-body collisions after excitation. In particular at atmospheric pressure, excimers play an important role due to the high probability of three-body collisions which promote excimer production (see table 4.1).

Table 4.1 gives an overview over elementary processes and associated reactions in argon discharges at atmospheric pressure. The listed reaction scheme is not complete and conduces to clarity. If nitrogen chemistry is added, the system gets much more complex. Similar reactions as shown for argon also exist for nitrogen. The most important reactions with small nitrogen impurities comprise Penning ionisation of nitrogen atoms through metastable argon species (Ar⁺ + N → Ar + N⁺ + e⁻) [85]. Due to the high surface-to-volume ratio of the single discharge cavities, surface effects such as neutralisation of excited species and secondary electron emission at surfaces can also play an important role in the discharges examined in the frame of this study.

In order to get an idea of species present, emission spectra of whole devices have been recorded using the USB spectrometer. A typical spectrum of an array device is exemplarily illustrated in figure 4.2 for a 50x50 100 µm pyramidal cavity array device and operation in 750 mbar argon at an excitation frequency of 30 kHz and 800 V peak-to-peak voltage.
4. Results

Figure 4.2.: Typical emission spectrum of investigated devices as recorded for a 50x50 100 µm pyramidal cavity array device operated in 750 mbar argon at an excitation frequency of 30 kHz and a peak-to-peak voltage of 800 V using the USB spectrometer. Source energy levels of observed emission lines are indicated together with typical excitation energies.
Emission spectra in the range of 300-400 nm originate from the second positive band of nitrogen (N\textsubscript{2}). It indicates that the molecular nitrogen impurities contained in the process gas are efficiently excited in the discharge. Typical excitation energies of the emitting second positive band of N\textsubscript{2} are in the order of 12 eV. Apart from the emission caused by the second positive band of N\textsubscript{2}, atomic argon emission lines are observed in the range of 690 - 1000 nm. These emission lines exclusively originate from argon 2p levels (in Paschen notation). Typical excitation energies of these energy levels are in in the order of 13 eV. Emission lines of argon ions, which are expected between 400 nm and 500 nm are not observed. Here, excitation energies are in the order of 21 eV. It can thus be estimated that the average electron energy of the discharge is in the order of a few eV. In case of higher electron energies, argon ion emission lines would be observable. Excimer emission lines lie in the vacuum UV and are not accessible with the diagnostics used in this study. However, due to the collisional regime, high excimer densities are presumably present in the discharge (see above).

Due to the BK7 windows in the experimental setup, neither the PMT nor the ICCD camera can observe the N\textsubscript{2} emission bands, as transmission is negligible in this wavelength region. Thus, all observations made in the following based on these diagnostics and emission intensity measurements reflect the behaviour of the argon 2p emission lines.

### 4.1.3. Discharge mode

The determination of the discharge mode of Si-based MSE microplasma arrays is essential in order to understand discharge dynamics and to optimise the design of the devices with respect to envisaged applications. All devices investigated in this work exhibit dielectrically coated electrodes and are thus by nature DBDs. However, their complex geometry significantly deviates from parallel plate symmetry and the array arrangement of thousands of micro-DBDs makes them sophisticated microplasma sources. In the following, the discharge mode of the investigated devices is determined on the basis of time-resolved voltage and current as well as PMT measurements and compared to behaviour of DBDs in parallel plate geometry.

The setup employed in this thesis only allows to measure voltage and current outside the discharge chamber. This restriction implicates that only applied voltage and total current through whole devices are directly accessible, where
the latter may be influenced by stray capacities in the setup. The total current is, due to the dielectric coatings on the electrodes, made up by a superposition of displacement and discharge current. The latter itself is a superposition of the discharge currents of single microplasmas in the array structure. A calculation of single cavity or whole device capacity is not trivial due to the complex electrode geometry and layer structure. It is thus sophisticated to conclude single microplasma gap voltage and discharge current values. However, as applied voltage waveforms are chosen to be bipolar triangular, the displacement current naturally follows a bipolar rectangular waveform. This allows to deduce the integral discharge current through the whole device from the total current signal.

Although neither current nor voltage information of single microplasmas in the array arrangement can be accessed, integral values for the whole device allow deduction of information on the single microdischarges. In first approximation, it can be assumed that the active discharge area of the array devices is made up of the sum of the single cavity base areas, neglecting the complex cavity and electrode geometry. This is supported by temporarily and spectrally integrated photographs perpendicular to the devices principle plane.

Figure 4.3 exemplarily shows a photograph of a 4x5x16 rectangular straight cavity array device operated in atmospheric pressure argon at an excitation frequency of 5 kHz and 600 V peak-to-peak voltage. Cavity sizes are 25, 100, 150 and 50 µm times 150 µm. The photograph reveals that for each cavity, independent of cavity size, approximately the whole cavity base area is filled with emission features. Thus, current peak values can be related to an effective discharge area which allows calculation of current densities. This approximation implies that calculated current densities represent lower threshold values in the case of smaller actual discharge areas.

The imaging in figure 4.3 illustrates homogeneous emission behaviour for the single microplasmas in an array arrangement. However, this does not imply the formation of a homogeneous DBD.

To definitely exclude a filamentary type of discharge, images recorded with a gate time in the range of 100 ns, the typical lifetime of filaments in DBDs, would be necessary. In order to be able to spatially resolve filaments, these images would have to be taken with high magnification, showing only small excerpts of a microplasma array device. The comparably low emissivity of the area of interest as well as the short gate time require strongly intensified
4. Results

Photograph of a 4x5x16 rectangular straight cavity array device operated in atmospheric pressure argon at an excitation frequency of 5 kHz and 600 V peak-to-peak voltage.

Figure 4.3.: Photograph of a 4x5x16 rectangular straight cavity array device operated in atmospheric pressure argon at an excitation frequency of 5 kHz and 600 V peak-to-peak voltage.
single shot cameras which are hardly available. Thus, this evidence cannot be provided in the frame of this study via optical measurements.

Apart from the homogeneity of the emission of single microplasmas, voltage and current characteristics of single microplasmas and whole microplasma arrays respectively allow more reliable determination of the DBD discharge mode and are easier to access.

**Voltage and current characteristics**

While the image shown in figure 4.3 clearly illustrates the area of active luminescence, it does not allow to deduce information on temporal emission behaviour of the microplasma arrays. As will be shown in section 4.2.1 and thus not discussed in detail here, the ignition of single microplasmas in an array arrangement does not occur synchronously. In fact, phase resolved emission spectroscopy reveals that their ignition occurs successively [86, 87]. It typically starts with one cavity and subsequent wave-like isotropic ignition of adjacent cavities. These emission waves propagate over the entire surface of the array device, where each cavity ignites once per wave. This way, time-integrated emission of the whole microplasma array appears homogeneous.

This has significant impact on the interpretation of the measured discharge current peaks and associated emission intensity and PMT signal peaks respectively. They are temporally broadened and exhibit smaller peak values compared to the case of synchronous ignition of all cavities. However, due to the comparably high propagation velocity of the wave-like feature, deduced current peak and FWHM values are in first approximation extreme lower and upper values respectively. Thus, their determination allows adequate statements concerning possible discharge modes.

The determination of discharge current peak values and respective full width half maximum (FWHM) values on the basis of oscillograms of applied voltage and total current is exemplarily illustrated in figure 4.4 for a 50x50 100 µm base pyramidal cavity array operated in atmospheric pressure argon at an excitation frequency of 10 kHz and a peak-to-peak voltage of 800 V. For each half period, a linear fit of the displacement current plateau level serves as base line. The difference of the respective total current peak and the corresponding base line current value specifies the discharge current peak value, denominated $I_{\text{peak}}$ in the figure. The FWHM of the discharge current peak is then determined at current values equal to half the discharge current peak value added to the base
4. Results

Figure 4.4: Illustration of discharge current determination. Oscillograms of applied voltage (black line) and total current (blue line) of a 50x50 100 µm base pyramidal cavity array operated in atmospheric pressure argon at an excitation frequency of 10 kHz and a peak-to-peak voltage of 800 V are exemplarily shown (a). A magnified excerpt of the oscillograms marked with the red rectangle is displayed for better exemplification (b). A linear fit (red line) of the plateau of the displacement current serves as baseline.

Discharge current values determined this way are a superposition of all single microplasma discharge currents on the array device.

At low excitation frequencies, discharge current peaks are too small to be distinguishable from signal noise. This is due to discharge onset at lower overvoltages for smaller frequencies, as will be shown in section 4.1.4. Thus, investigation of discharge current peaks is performed for excitation frequencies higher or equal to 5 kHz. However, this frequency range limits the number of observed discharge current pulses per pulse train and half period respectively to a maximum of 2. Here, the respective second current peak of a pulse train may not be fully developed due to the simultaneous reversal of the slope of the applied voltage waveform. As a consequence, only respective fully developed first discharge current peaks of current pulse trains are taken into consideration in the following.

Figure 4.5 exemplarily illustrates (a) the small current peak values for low frequency excitation of a 50x50 100 µm base pyramidal cavity array device operated in 750 mbar Ar at 1 kHz and 800 V\textsubscript{PP} as well as (b) the disruption of second order discharge current peak development by reversal of the slope of the applied voltage waveform for a 50x50 50 µm base pyramidal cavity array device operated in 500 mbar Ar at 20 kHz and 800 V\textsubscript{PP}. While the small current peak values in (a) do not allow distinction from current signal noise, the corresponding integral optical emission peaks registered with the PMT are
4. Results

Figure 4.5.: Illustration of the limitation of current peak value determination. Oscilloscopes of applied voltage (black line), PMT signal (red line), and total current (blue line) of (a) a 50x50 100 µm base pyramidal cavity array operated in 750 mbar Ar at 1 kHz and 800 V_{PP} as well as of (b) a 50x50 50 µm base pyramidal cavity array operated in 500 mbar Ar at 20 kHz and 800 V_{PP} are exemplarily shown.

well-defined and indicate the existence of the former.

A disruption of the respective second current peak and associated PMT signal peak for both, positive and negative half period, can be observed in (b). Moreover, an asymmetry of PMT signal as well as of total current with respect to electrode polarity and half period respectively is obvious from these oscilloscopes. It can be related to the asymmetric electrode design and will be discussed in detail in section 4.2.3 and is thus not further investigated at this point.

Peak and FWHM values of respective first discharge current peaks of current pulse trains in the positive and negative half period are determined as depicted in figure 4.4 and described above for investigated devices under variation of gas pressure, peak-to-peak voltage as well as excitation frequency in order to compare these quantities to typical values for different DBD discharge modes (see table 2.1).

For a regime spanned by operation parameters of 500 - 990 mbar gas pressure, 5 - 30 kHz excitation frequency and 700 - 800 V peak-to-peak voltage, resulting typical discharge peak values I_{peak} are summarised in table 4.2 together with corresponding calculated current densities j_{peak} for 50x50 50 µm and 100 µm pyramidal cavity devices. The calculation of the j_{peak} is performed under the assumption of an effective device discharge area made up of the sum of its single cavity base areas as stated above.
4. Results

Table 4.2.: Regimes of typical discharge current peak values \( I_{\text{peak}} \) and current densities \( j_{\text{peak}} \) as determined for several MSE devices operated in argon under variation of pressure, excitation frequency and peak-to-peak voltage.

<table>
<thead>
<tr>
<th>MSE Device</th>
<th>( I_{\text{peak}} ) [mA]</th>
<th>( j_{\text{peak}} ) [mA cm(^{-2})]</th>
</tr>
</thead>
<tbody>
<tr>
<td>50x50 50 µm pyramidal cavity</td>
<td>0.8 - 9.6</td>
<td>12.8 - 153.6</td>
</tr>
<tr>
<td>50x50 100 µm pyramidal cavity</td>
<td>2.8 - 29.4</td>
<td>11.2 - 117.6</td>
</tr>
</tbody>
</table>

In comparison with typical discharge current densities for different parallel plate DBD operation modes as stated in table 2.1, the values received strongly indicate operation of the investigated devices in APGD mode. This assumption is supported by the distinct periodicity of the discharge current with respect to the excitation frequency which suggests a pulsating homogeneous DBD discharge mode according to section 2.2.2. Although the registered current is a superposition of the discharge currents of the single discharges in the array arrangement and the displacement current of the whole device, its periodicity allows the conclusion that the single contributing components are coherent and have the same periodicity. Thus, devices investigated in this study can be assumed to operate in pseudoglow mode within the given operation parameter range. Detailed analysis of current behaviour under operation parameter variation yields further proof and is discussed in the following.

**Parameter variation**

Discharge current peak and respective FWHM values for a variation of excitation frequency are exemplarily shown in figures 4.6 and 4.7 respectively for a 50x50 50 µm base pyramidal cavity array device which is operated in argon at pressures between 500 and 1000 mbar. The peak-to-peak voltage is in each case 780 V. For each excitation frequency and pressure, discharge current peak and FWHM values for both, negative and positive half period, are determined as indicated in the figures.

The discharge current peak values show a uniform behaviour and increase in first approximation linearly with increasing excitation frequency, independent of respective half periods. Determined peak values increase from approximately 1 mA for an excitation frequency of 1 kHz to values around 8 mA at 30 kHz. At an elevated excitation frequency of 30 kHz, however, discharge current peak values in the negative half period are slightly smaller than those in the positive half period. This may be due to an enhanced memory effect.
4. Results

Figure 4.6.: Dependence of discharge current peak values on gas pressure and frequency for a 50x50 50 µm base pyramidal cavity array device operated in argon at a peak-to-peak voltage of 780 V.

Figure 4.7.: Dependence of discharge current peak FWHM values on gas pressure and frequency for a 50x50 50 µm base pyramidal cavity array device operated in argon at a peak-to-peak voltage of 780 V.
at higher excitation frequencies which originates from the comparably smaller periods in between successive half periods. Thus, the asymmetry of the electrode design can have a stronger impact and lead to the observed phenomenon. Although investigations at frequencies higher than 30 kHz would allow further analysis, the resulting higher discharge current peak values are experimentally found to be likely to destroy the thin Si$_3$Ni$_4$ dielectric layers. This can lead to DC currents in the device resulting in thermal destruction of the latter. Thus, measurements at higher frequencies are omitted here.

Correlating the respective FWHM in figure 4.7 with discharge current peak values in figure 4.6, one can conclude that for increasing frequency and thus peak values, corresponding FWHM values decrease. Deviations between the FWHM values for different Ni grid polarities are negligible.

The phenomenon of discharge current pulses exhibiting smaller FWHM and higher peak values with increasing excitation frequency needs closer investigation, in particular at lower frequencies, in order to understand involved processes. Before the low frequency regime is investigated in detail, further observations deduced from discharge current peak investigations are reported.

While the influence of excitation frequency on discharge current peak behaviour is obvious, that of gas pressure is comparably small. Deviations of discharge current peak values in dependence of gas pressure never exceed 2 mA for the same electrode polarity. However, the operation regime of the investigated devices is limited with regards to pressure. For pressures lower than 500 mbar, parasitic discharges at the edges of the active device area are likely to occur. Due to pd scaling, discharge onset on comparably higher scales becomes more probable. This is in particular critical close to the edges of the Si wafer where defects in the dielectric coatings are likely due to cutting of the devices. Once a parasitic discharge covers an area of defect dielectric layers, the resulting DC current leads to thermal destruction of the device. Thus, investigations in this thesis are carried out at gas pressures of at least 500 mbar.

Apart from excitation frequency and pressure variation, a change in peak-to-peak voltage has an influence on discharge current peak behaviour as well. Figure 4.8 exemplarily shows determined discharge current peak values for a variation of applied peak-to-peak voltage as recorded for a 50x50 100 µm pyramidal cavity array device in argon at atmospheric pressure for various excitation frequencies. Current peak values are in each case determined for both half periods of the excitation period as indicated in the figure.
Data points in brackets represent discharge current peaks which are not fully developed due to insufficient peak-to-peak voltage. These peaks are cut off by the reversal of the slope of the applied peak-to-peak voltage. Naturally, the current peak value for these data points is smaller than that of the first successive fully developed discharge current peak. However, such undeveloped peaks are only observed in the positive half period, indicating an asymmetric ignition behaviour of the array device. The applied voltage necessary for discharge ignition is smaller in the negative half period.

Moreover, a strong asymmetry of discharge current peak values with regards to respective half periods is again observed. The peak values in the positive half period always exceed those in the negative one. Again, the frequency dependence of this deviation is obvious. It increases with increasing excitation frequency.

The impact of peak-to-peak voltage on discharge current peak values is mirrored in the strong dynamics of the latter. An increase of peak-to-peak voltage allows discharge current peaks which have been cut off at smaller voltages to fully develop, leading to an increase of current peak values in figure 4.8. Once the peak is fully developed, further increase of peak-to-peak voltage has no significant influence on current peak values. This changes when a second discharge current peak evolves. At this point, a decrease of current peak values
of the first peak is observed, irrespective of Ni grid polarity. This transition occurs between 750 and 775 V_{pp} for excitation frequencies of 5 and 10 kHz and between 775 and 800 V_{pp} for excitation at 20 kHz.

As an impact of second discharge current peak evolution within the same Ni grid polarity on the respective incipient current peaks can be excluded, this phenomenon indicates a mutual influence of consecutive discharges of reverse polarity. As the discharge ignition occurs at earlier phasing within the excitation period for increasing peak-to-peak voltage, further voltage increase after extinction of the incipient discharge burst may be sufficient to give rise to a second discharge onset. Thus, the occurrence of a second discharge current pulse per pulse train implies a shorter period of time between current pulse trains of reverse polarity. As a consequence, higher remanent species concentrations may be present upon discharge re-ignition. Here, metastables can lower the ionisation threshold, lead to Penning ionisation or cause secondary electron emission upon impinging the dielectric surfaces. At the same time, the evolution of a second current peak of the same Ni grid polarity implies higher surface charge densities. Consequently, both species can reduce the gap voltage where discharge onset occurs. Less overvoltage upon discharge onset yields lower electric fields in the gap. The respective current peak value is thus reduced [78]. Further increase of peak-to-peak voltage at a constant excitation frequency implies increasing $dU/dt$ values which competes against lower ionisation threshold with regards to impact on current peak values. This effect becomes dominant at some point and finally leads to an increase in discharge current peak values until a third discharge current peak may evolve.

The time lag between successive pulses of reverse Ni grid polarity by far exceeds the natural lifetime of argon metastables in an atmospheric pressure argon atmosphere ($\sim 200 \text{ ns}$ [88]). Even if radiation trapping is considered under conditions similar to those in this study, effective metastable lifetimes are only of the order of 10 µs [89]. The lifetime of excimers is closely connected to that of metastable levels, as the latter are an effective source of population. Nevertheless, figure 4.8 suggests communication between discharges of reverse polarity. A possible source may lie in the high surface to volume ratio of the single discharge cavities and adsorbed argon ions. These ions as well as those trapped in the discharge gap (see section 2.2.2) could be converted to excimers and metastables through the following reduced reaction scheme which may be an explanation for remanent excited species [89]:
4. Results

\[
\begin{align*}
\text{Ar}^+ + 2\text{Ar} & \rightarrow \text{Ar}_2^+ + \text{Ar} \\
\text{Ar}_2^+ + e^- & \rightarrow \text{Ar}^{**} + \text{Ar} \\
\text{Ar}^{**} & \xrightarrow{hv} \ldots \xrightarrow{hv} \text{Ar}^m \\
\text{Ar}^m + 2\text{Ar} & \rightarrow \text{Ar}_2^* + \text{Ar} 
\end{align*}
\]

(4.1) (4.2) (4.3) (4.4)

Upon a three-body collision of an argon ion with two argon atoms, an argon excimer ion and a ground state argon atom are produced (equation 4.1). The excimer ions can be dissociated into a highly excited argon atom and a ground state argon atom by electron impact (equation 4.2). These highly excited argon species fall into metastable argon levels via cascades (equation 4.3). The metastable argon species can again be converted to argon excimer ions and neutral argon atoms in three-body collisions with two ground state argon atoms (equation 4.4). This reaction scheme allows enhanced effective metastable and excimer lifetimes of more than 10 µs [89] under experimental conditions comparable to that in this study and could be one source of excited species with long lifetimes. Due to the scheme above, metastable lifetimes can be deduced from measurements of the emission intensity of argon excimer continuum radiation at 127 nm [89]. However, in the frame of this study, observation of radiation at 127 nm is not possible.

The investigation of discharge current peaks and the implied restriction to excitation frequencies of 5 kHz or higher and pulse trains consisting of a maximum number of two peaks respectively does not allow detailed analysis of the self-pulsing behaviour of the investigated devices. As the emission bursts associated with respective discharge current peaks by nature have the same periodicity and can be observed at lower frequencies using the PMT, their behaviour can be exploited to analyse the low excitation frequency regime. While deduction of discharge current peak periodicity is readily possible, limited information on discharge current pulse FWHM and peak values can be concluded as the emission pulses are influenced by lifetimes of excited states, quenching, electron density and EEDF, and other plasma parameters. In the following, this method is applied to investigated devices in order to describe the observed self-pulsing.
Summary

The discharge mode of investigated devices is examined. Time integrated imaging of the emission shows homogeneous emission features. Moreover, the periodicity of voltage and current suggests operation in a homogeneous DBD mode. Voltage and current measurements yield current densities which are typically associated with APGDs [60]. Here, the current peak behaviour under frequency variation suggests operation in pseudoglow mode [70]. Strong evidence for the influence of long-lived excited species generated in antecedent half periods on successive ones is found.

4.1.4. Self-pulsing

In this section, the self-pulsing of the investigated MSE array devices is examined on the basis of discharge emission bursts as registered with the PMT as well as simultaneously measured voltage oscillograms.

Influence of excitation frequency

At first, the discharge burst behaviour under frequency variation is investigated. It is similar for all investigated devices and exemplarily illustrated in figure 4.9 for a 50x50 100 µm pyramidal cavity array device which is operated in atmospheric pressure argon at a constant peak-to-peak voltage of 800 V and excitation frequencies of (a) 1 kHz, (b) 2 kHz, (c) 3 kHz and (d) 5 kHz in the form of oscillograms of applied voltage and PMT signal in arbitrary units.

It is obvious that the number of emission bursts per half period diminishes with increasing excitation frequency from four at 1 kHz to two at 5 kHz for the recorded operation parameters. Further increase of excitation frequency finally leads to the formation of only one emission burst per half period at 20 kHz. For convenience, the number of discharge bursts per half period in dependence of excitation frequency is summarised in figure 4.10 for the same device and operation parameters.

The reduction of the number of emission bursts with increasing frequency can be interpreted with the help of investigations of emission burst and thus current pulse onset behaviour as well as of time lag between incipient and second emission burst per pulse train under frequency variation.
4. Results

Figure 4.9.: Oscillograms of PMT signal [a.u.] and applied voltage for a 50x50 100 µm pyramidal cavity array device operated in atmospheric pressure argon at a constant peak-to-peak voltage of 800 V and excitation frequencies of (a) 1 kHz, (b) 2 kHz, (c) 3 kHz and (d) 5 kHz.
Figure 4.10.: Number of emission bursts per half period as registered with the PMT for a 50x50 100 µm pyramidal cavity array device operated in atmospheric pressure argon at a constant peak-to-peak voltage of 840 V under frequency variation.

Apart from a reduction of the number of emission bursts with increasing frequency, figure 4.9 reveals, that for low-frequency excitation the emission bursts superpose a kind of continuous emission during the pulse train. The intensity of this continuous background increases with increasing number of successive bursts. As emission bursts within one pulse train approximately occur with a fixed frequency, this indicates a continuous supply of energy to excite argon species. A possible source are metastables and excimers which are built up and accumulate with successive discharge current pulses. Moreover, ions trapped in the discharge gap (see section 2.2.2) may lead to this phenomenon through recombinatory excitation. This increasing background strongly suggests that higher numbers of emission bursts lead to build-up of higher excited species concentrations with comparably long lifetimes.

The ratio of this continuous background and emission peak values increases with increasing frequency. This is a combined effect and can be ascribed to an increased frequency of emission bursts within one pulse train for increasing excitation frequencies (see table 4.3). As a result, the time-separation between successive emission bursts diminishes and leads to increasing overlap of the latter, which explains the observed behaviour.

The determination of discharge ignition on the basis of applied voltage and total current as measured in the frame of this study is not readily possible. How-
4. Results

Figure 4.11.: Applied voltage upon incipient discharge pulse onset. Voltages are determined at the peak value of incipient discharge current pulses for 50x50 50 µm and 100 µm pyramidal cavity array devices operated in atmospheric pressure argon at a peak-to-peak voltage of 800 V.

ever, applied voltage and gap voltage are interrelated by the memory effect. Thus, the determination of applied voltages upon incipient discharge current pulse onset allows to draw conclusions on the former. Figure 4.11 shows such voltages in dependence of the excitation frequency for respective half periods of 50x50 50 µm and 100 µm pyramidal cavity array devices respectively. Here, the devices are operated in atmospheric pressure argon at a peak-to-peak voltage of 840 V. The applied voltage upon incipient discharge current pulse onset is determined at the peak value of corresponding discharge current pulses. Other choices of points in time of discharge pulse onset (for example 10% of the peak value) are difficult to determine due to the superposition of the continuous emission intensity background and emission peaks and are thus inconvenient.

It is discerned that with increasing excitation frequency the applied voltage upon incipient emission bursts increases until a certain plateau level is reached. Here, cavity size seems to play a role, as the plateau level is reached at 5 kHz for the device with 100 µm cavity base length while it is reached at 40 kHz for the device with 50 µm cavity base length. Moreover, an asymmetry of incipient emission burst occurrence with respect to Ni grid polarity is observed. While emission bursts in the positive half period occur at smaller applied voltage val-
4. Results

Table 4.3.: Time lag $\Delta t_{1-2}$ and voltage rise $\Delta V_{1-2}$ between incipient and second emission burst for a 50x50 100 µm pyramidal cavity array device operated in atmospheric pressure argon at 800 V peak-to-peak voltage under frequency variation. Respective quantities are distinguished in two categories depending on respective Ni grid polarity ($pHP$ and $nHP$).

<table>
<thead>
<tr>
<th>Excitation frequency [kHz]</th>
<th>$\Delta t_{1-2}$ [µs]</th>
<th>$\Delta V_{1-2}$ [V]</th>
<th>$\Delta t_{1-2}$ [µs]</th>
<th>$\Delta V_{1-2}$ [V]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>20</td>
<td>32</td>
<td>22</td>
<td>35</td>
</tr>
<tr>
<td>2</td>
<td>13</td>
<td>42</td>
<td>17</td>
<td>54</td>
</tr>
<tr>
<td>3</td>
<td>11</td>
<td>53</td>
<td>13</td>
<td>62</td>
</tr>
<tr>
<td>5</td>
<td>6.8</td>
<td>54</td>
<td>7.2</td>
<td>58</td>
</tr>
</tbody>
</table>

At the same time, the time lag and the accompanying voltage rise between incipient and second emission burst under frequency variation are determined from emission burst peak positions in the oscillograms of PMT signal and applied voltage. Although discharge current peak and emission burst peak position may not exactly temporally overlap, this approximation sufficiently reflects discharge current periodicity as well as frequency behaviour. Both, time interval $\Delta t_{1-2}$ between incipient and second emission burst as well as the associated voltage rise $\Delta V_{1-2}$ are exemplarily summarised in table 4.3 as deduced from figure 4.9 for a 50x50 100 µm pyramidal cavity array device operated in atmospheric pressure argon at 800 V peak-to-peak voltage and varying excitation frequencies.

Table 4.3 illustrates that the time lag between incipient and second emission burst and thus discharge current peak decreases with increasing excitation frequency. At the same time, the associated voltage rise between the successive pulses increases. This behaviour is observed for both Ni grid polarities, while time lag and voltage rise are higher for the nHP, again indicating asymmetry of the discharge with respect to Ni grid polarity.

The observed behaviour can be interpreted as a consequence of ion inertia. For higher excitation frequencies, the slope $dU/dt$ of the applied voltage is higher as well, allowing a faster voltage rise which is necessary to overcome the reduced electric field in the gap and to ignite the second discharge cur-
rent pulse. However, remanent ions in the discharge gap affecting the local field play a more important role on shorter time scales, so that a comparably higher voltage rise between successive pulses can result for higher excitation frequencies. Thus, less discharge current pulses can evolve after the incipient one before the slope of the applied voltage reverses. Consequently, smaller densities of excited species with comparably long lifetimes are build up until the reversal of the slope of the applied voltage.

As a result, the voltage rise needed to achieve breakdown for the first discharge current pulse of reverse Ni grid polarity is comparably higher. The resulting higher overvoltage across the discharge gap upon breakdown gives rise to discharge current peaks of comparably higher peak and smaller FWHM values. A similar correlation of overvoltage and discharge current peak and FWHM values has been found and investigated for parallel plate DBD discharges in helium by Bartnikas et al. [78].

Following this line of argumentation, the incipient discharge ignition for respective Ni grid polarities takes place at a later phasing within the excitation period for increasing excitation frequencies as observed in figure 4.11 due to a reduced number of remanent long-lived excited species. Apart from the increased voltage rise necessary in between successive discharge current pulses of the same Ni grid polarity, this additionally limits the number of pulses that can evolve within one pulse train.

The plateau level in figure 4.11 and consequently a constant applied voltage upon incipient emission bursts could be reached once the temporal proximity of reverse Ni grid polarities compensates the smaller density of remanent excited species remaining.

The asymmetry of the applied voltage upon incipient emission burst with respect to Ni grid polarity (see figure 4.11) suggests, that the choice of an appropriate peak-to-peak voltage would allow the generation of a discharge in only one half period. However, if the applied peak-to-peak voltage is gradually reduced in experiment, the discharges in both half periods extinguish simultaneously, as soon as one of them cannot be sustained. This effect verifies the assumption of the existence of long-lived charged species at the dielectric surfaces which are generated in antecedent half periods and influence subsequent ones (see section 4.1.3).
4. Results

Influence of applied peak-to-peak voltage

Apart from excitation frequency, the applied peak-to-peak voltage determines the self-pulsing of the microplasma arrays. Figure 4.12 exemplarily shows a series of oscillograms of applied voltage and PMT signal in arbitrary units as recorded for a 50x50 100 µm pyramidal cavity at an excitation frequency of 1 kHz and peak-to-peak voltages of (a) 700 V, (b) 725 V, (c) 750 V, (d) 775 V and (e) 800 V.

It can be perceived that an increase of applied peak-to-peak voltage gives rise to the formation of an increasing number of emission bursts per half period. While for a peak-to-peak voltage of 700 V only one emission burst is observed, an increase of 100 V in steps of 25 V allows the successive formation of up to four emission bursts. To analyse this phenomenon, the time lag $\Delta t_{1-2}$ and associated voltage rise $\Delta V_{1-2}$ between incipient and second emission burst as well as the absolute value of the applied voltage $V_i$ upon incipient emission burst occurrence are determined from emission burst peak values as described above for frequency variation and summarised in table 4.4.

The table illustrates that the applied voltage upon incipient emission bursts diminishes with increasing applied peak-to-peak voltage. Moreover, a reduction of the time lag between incipient and second emission burst can be discerned. The asymmetry of discharge behaviour with regards to Ni grid polarity is mirrored in occurrence of incipient emission bursts at lower applied voltages and earlier phasing within the excitation period respectively and increased lag time and voltage rise between successive emission bursts of the same Ni grid polarity in the nHP compared to the pHP.

Again, this behaviour can be interpreted under the assumption of excited species with effective lifetimes longer than the time lag between last and incipient emission burst of reverse polarity. An increase of applied peak-to-peak voltage at constant excitation frequency accounts for an increase of the slope $dU/dt$ of the applied voltage waveform. As the ignition voltage of incipient discharges is exceeded at earlier phasing within the excitation period, the successive voltage rise until the reversal of the applied voltage waveform slope may be sufficient to give rise to a second discharge of the same Ni grid polarity. This leads to an increasing number of emission bursts per half period as well as higher surface charge densities at the dielectric surfaces [90]. The resulting reduced time lag between pulse trains of reverse polarity allows higher remanent excited species densities in subsequent half periods. Moreover, the increased
Figure 4.12.: Oscillograms of PMT signal and applied voltage for a 50x50 100 µm pyramidal cavity array device operated in argon at 500 mbar, a constant excitation frequency of 1 kHz and peak-to-peak voltages of (a) 700 V, (b) 725 V, (c) 750 V, (d) 775 V and (e) 800 V.
4. Results

Table 4.4.: Time lag $\Delta t_{1-2}$ and voltage rise $\Delta V_{1-2}$ between incipient and second emission burst as well as absolute value of applied voltage upon incipient emission burst $V_i$ for a 50x50 100 µm pyramidal cavity array device operated in 500 mbar argon at 1 kHz excitation frequency under variation of peak-to-peak voltage. Respective quantities are distinguished in two categories depending on respective Ni grid polarity (pHP and nHP).

<table>
<thead>
<tr>
<th>Peak-to-peak voltage [V]</th>
<th>pHp</th>
<th></th>
<th>nHP</th>
</tr>
</thead>
<tbody>
<tr>
<td>$V_i$ [V]</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\Delta t_{1-2}$ [µs]</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\Delta V_{1-2}$ [V]</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>700</td>
<td>339</td>
<td>-</td>
<td>-</td>
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<tr>
<td>725</td>
<td>309</td>
<td>24</td>
<td>35</td>
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<tr>
<td>750</td>
<td>297</td>
<td>24</td>
<td>36</td>
</tr>
<tr>
<td>775</td>
<td>291</td>
<td>22</td>
<td>34</td>
</tr>
<tr>
<td>800</td>
<td>282</td>
<td>22</td>
<td>35</td>
</tr>
</tbody>
</table>

number of emission bursts per pulse train leads to additional build-up of long-lived excited species densities. Together with higher surface charge densities, both effects lead to a decrease of ionisation threshold and effectively reduce the applied voltage necessary to initiate incipient discharge pulses. With increasing applied peak-to-peak voltage and number of pulses per pulse train respectively, the overall remanent excited species density may increase and lead to a weak reduction of voltage rise between successive pulses due to further reduced ionisation threshold as observed and documented in table 4.4.

Although the investigated devices are composed of a multitude of single discharge cavities in an array structure, their integral behaviour in general coincides with that of parallel plate DBDs in homogeneous mode. Measured current densities suggest operation of the whole device as well as of single microplasmas in pseudoglow mode. The frequency and voltage dependence of the self-pulsing of the discharges agrees reasonably well with observations of Radu et al. for helium pseudoglow discharges in parallel plate geometry [70] and supports this assumption. However, in case of the MSE array devices investigated in the frame of this study, their behaviour is composed of that of a multitude of single cavities. Strong dynamics of the single microplasmas in the array structure as well as coupling between adjacent cavities in form of non-simultaneous wave-like ignition are observed. These phenomena have not been investigated and explained yet. They cannot be detected by integral diagnostics and are thus investigated by means of phase and space resolved optical emission spectroscopy in the following.
4. Results

Summary

The self-pulsing of investigated MSE array devices is investigated on the basis of PMT measurements. It is found, that the number of emission bursts per half period decreases with increasing frequency. This is ascribed to smaller densities of remanent long-lived excited species and resulting higher breakdown voltages. A continuous emission background is observed to build up with increasing number of emission bursts per pulse train. This is interpreted as a successive build-up of long-lived excited species which accumulate during one pulse train. Apart from excitation frequency, applied peak-to-peak voltage determines the number of emission bursts per half period. An increasing peak-to-peak voltage accounts for an increasing number of bursts. Here, the breakdown at earlier phasing within the excitation period allows the evolution of additional emission bursts and thus leads to higher densities of remanent long-lived excited species which again reduce the gap voltage necessary for incipient breakdown.

The emission burst behaviour under voltage and frequency variation again agrees well with that observed for helium pseudoglow discharges in parallel plate geometry [70].

4.2. Discharge dynamics

The dynamics of single microplasma cavities in an array structure as well as the coupling of adjacent cavities are investigated by means of phase and space resolved optical emission spectroscopy using the ICCD camera with attached LDM. Complimentary current and voltage as well as PMT measurements are performed at the same time. Several transient phenomena are observed, independent of the type of array device. In the following, observed phenomena and dynamics are exemplarily introduced before they are separately discussed in more detail.

In order to investigate the dynamics of the single microplasmas and their behaviour in an array arrangement, ICCD camera and attached LDM are aligned under an angle of 90° to the principal plane of investigated devices. Phase resolved images of the two-dimensionally spatially resolved and spectrally integrated emission intensity of the operated devices are recorded.
Figure 4.13 exemplarily illustrates the transient phenomena observed for a 50x50 100 µm pyramidal cavity array device operated in atmospheric pressure argon at an excitation frequency of 1 kHz and 800 V peak-to-peak voltage. The excitation period is temporally resolved in 500 ns steps while gate times of the camera are chosen to be 500 ns as well. Images are recorded with a spatial resolution of 3.6 µm per pixel covering a section of 1843x1843 µm of the device surface. The figure comprises (a) oscillograms of applied voltage and integral emission intensity of the recorded PROES images in arbitrary units in the centre as well as false colour PROES images. The temporal and spatial evolution of emission intensity during the incipient emission burst of the positive half period is illustrated by the image sequence at the top of the figure (b) from left to right. The time separation between single images of the sequence is 1 µs. The columns of images to the left and right of the oscillograms comprise PROES images recorded at the points in time of maximum emission of the successive emission bursts in the positive (c) and negative (d) half period. The sequence from top to bottom corresponds to the temporal order of the emission burst maxima within the respective half period.

The oscillograms (a) show the expected self-pulsing of the discharge which is abruptly interrupted upon reversal of the slope of the applied voltage. The continuous emission background which builds up with increasing number of successive emission bursts within one pulse train as well as the asymmetry of emission intensity with regards to Ni grid polarity are distinguishable as well.

From (b) it can be discerned that cavities in an array arrangement do not simultaneously ignite during the evolution of one emission burst. In fact, ignition onset starts at particular cavities, affects adjacent cavities and finally spreads isotropically over the array area in a wave-like manner. This has significant impact on the interpretation of integral diagnostic data and thus has to be described and analysed in detail. In this case, the wave-like ignition starts in the centre of the right image border. These wave-structures are referred to as ionisation waves in the following. Although the observed emission intensity only allows to conclude excitation, it is most probable that ionisation comes along with it. As this feature is reproducible and detectable with the PROES setup due to its strong periodicity, it is not a random feature. Detailed analysis of the ionisation waves will be presented in section 4.2.1.

Moreover, it can be identified in (b) as well as in (c) and (d) that emission is more intense at the cavity edges compared to the cavity centre, where the
4. Results

Figure 4.13.: False colour PROES images and oscillograms (a) of applied voltage and integral emission intensity deduced from the former resolving one excitation period for a 50x50 100 µm pyramidal cavity array device operated in atmospheric pressure argon at an excitation frequency of 1 kHz and 800 V peak-to-peak voltage. The gate time and temporal resolution of the PROES images are in each case 500 ns. The top row of PROES images (b) shows the temporal evolution of emission for the incipient emission burst of the pHP in time steps of 1 µs from left to right. PROES images in columns to the left and right of the oscillograms are recorded at the points in time of maximum emission of the successive emission bursts of the pHP (c) and nHP (d) respectively, where the sequence from top to bottom corresponds to the temporal order of emission burst maxima. They are identified by P or N depending on positive or negative Ni grid polarity and a number representing the corresponding emission burst order.
observed quadratic emission features are smaller in the negative half period compared to the positive one. This asymmetry has not been observed yet and will be discussed in section 4.2.3 together with the other effects observed with regards to Ni grid polarity.

Another typical feature of the array devices is observable in (c) and (d). With increasing order of emission bursts, the emission at the cavity edges becomes less pronounced in relation to that in the cavity centre. This feature is first stated in the frame of this study and will be addressed in section 4.2.2

It is obvious that the microplasma array devices exhibit transient collective and individual discharge cavity behaviour which has to be investigated and analysed in order to tailor devices to specific application needs. In the following, the above observations are thus discussed in detail.

4.2.1. Ionisation waves

While time integrated emission intensity of single microplasma cavities in an array arrangement recorded with the PROES setup is the same for different cavities as also suggested by figure 4.3, this is not true for single transient positions within the excitation period.

Two-dimensionally space and phase resolved optical emission spectroscopy reveals that for each emission burst, a wave-like ignition of the single cavities is observed. Initially, few cavities ignite followed by a successive ignition of adjacent cavities. This successive ignition and ensuing extinction occurs isotropically until every cavity is ignited once per wave, where these ionisation waves are reproducible for same operation parameters. The propagation direction of the wave is not influenced by the locations of electrical contacts on the Si wafer and Ni grid surface.

The starting point of these ionisation waves and initial ignition of single cavities strongly depends on gas pressure but is arbitrary for different devices. However, no significant influence of excitation frequency and applied peak-to-peak voltage can be observed. This suggests that the initial ignition of cavities is determined by $pd$ scaling and Paschen’s law respectively. Some cavities with fabrication defects may preferably ignite at certain gas pressures, whereas the same may apply for other cavities at different operation parameters.
4. Results

The observed collective behaviour and underlying interaction of adjacent cavities has significant influence on device performance. The exchange of energy between adjacent cavities may be favourable in certain applications, while it may be unwanted in others. Thus, processes involved in cavity interaction and wave propagation have to be understood in order to promote or prohibit the latter. In the following, the dependence of wave propagation on external parameters is investigated and underlying processes are identified.

**Ionisation wave velocity**

The parameter which is best accessible with the experimental PROES setup in use is the ionisation wave propagation velocity. Moreover, it can help to determine processes that promote ionisation wave propagation as the velocity is directly connected with typical timescales of involved reactions. However, the identification of an ionisation wave front is necessary in order to determine the wave velocity.

The PROES setup allows to record two-dimensionally space and phase resolved detection of optical emission intensity. Investigated devices are imaged as a whole under an angle of 90° with respect to the Si wafer principal plane. This way, the evolution of emission intensity of each cavity in the array arrangement can be observed. The convention chosen in this study constitutes the determination of the wave front location by local maxima of emission intensity in time. This means, the ionisation wave velocity is determined from the temporal delay of emission intensity maxima of two spatially separated cavities in the array arrangement.

Figures 4.14 and 4.15 exemplarily illustrate the determination of the ionisation wave velocity for the first emission burst in the positive half period of a 32x32 50 µm cylindrical straight cavity array device. It is operated in atmospheric pressure argon at an excitation frequency of 20 kHz and a peak-to-peak-voltage of 675 V. A series of PROES images recorded at gate times and time steps of 50 ns serves as the basis for ionisation wave velocity determination. The images are recorded at an effective spatial resolution of 14.79 µm per pixel.

Figure 4.14 shows selected images of this series which allow to track the wave during the first emission burst in the positive half period. Each image shows the integral optical emission intensity distribution of the array device in false colours at a specific phasing within the excitation period. The delay of the gate
4. Results

Figure 4.14.: Series of PROES images (gate time: 50 ns) recorded for a 32x32 50 µm cylindrical straight cavity array device operated in atmospheric pressure argon at an excitation frequency of 20 kHz and a peak-to-peak voltage of 675 V during the first emission burst in the positive half period. The images show the integral emission intensity distribution of the array device in false colours. Delays indicated at the top of the images refer to the point in time of synchronisation. The cavities marked 1 and 2 which are indicated by white circles are used for determination of the ionisation wave velocity.
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Figure 4.15.: Emission intensity evolution for cavities 1 (solid black line) and 2 (solid red line) as indicated in figure 4.14 during the first emission burst (32x32 50 µm cylindrical straight cavity array device, 1000 mbar argon, 20 kHz, 675 Vpp). Emission intensities are normalised to the emission intensity maximum of the respective cavity in this time interval. The delay of emission intensity maxima $\Delta t_w$ is indicated.

time to a random synchronisation point in time with respect to the excitation period is indicated at the top of each image. It allows to determine the phase relationship of the single images among themselves.

It can be discerned that in this case emission is first observed at the lower left part of the array device at 295 ns. It spreads along the lower and left device edges (695 ns). Finally, the ionisation wave concentrically crosses the device surface at 1095 ns, initiated at the device edges. The intensity in the resulting area of increased emission intensity at the device centre decreases at 1895 ns and the emission burst extinguishes.

In this case, the cavities used for determination of the ionisation wave velocity are indicated in the images by white circles and numbered 1 and 2 respectively. The delay of emission intensity maximum between cavity 1 and cavity 2 is used to determine the ionisation wave velocity.

Figure 4.15 shows the evolution of emission intensity for cavity 1 (black solid line) and cavity 2 (red solid line) during the first emission burst. In each case, the emission intensity is normalised to the maximum emission intensity registered for the respective cavity in this time frame. The delay $\Delta t_w$ between emission intensity maxima of both cavities is indicated in the figure. The ionisation
Ionisation wave velocities in dependence of excitation frequency as determined for a 32x32 50 µm cylindrical straight cavity array device from PROES imaging as described above. The device is operated in atmospheric pressure argon at a peak-to-peak voltage of 675 V.

Ionisation wave velocities are determined on the basis of the method introduced above for all investigated array devices under variation of gas pressure, excitation frequency as well as peak-to-peak voltage. Ionisation wave velocities are found to be in the order of a few km s$^{-1}$ for all devices. While no significant influence of gas pressure and peak-to-peak voltage can be discerned from the measurements, a correlation between excitation frequency and ionisation wave velocity is found. This is partly accounted for by the comparably small percentaged variation of gas pressure and peak-to-peak voltage within possible operation parameters.

Figure 4.16 exemplarily shows deduced ionisation wave velocities for incipient emission bursts in the positive half period under variation of excitation frequency of a 32x32 50 µm cylindrical straight cavity array device. It is operated in atmospheric pressure argon at a peak-to-peak voltage of 675 V. Error bars are calculated from spatial and temporal resolution of the recorded PROES images using Gaussian error propagation.

It can be discerned that the ionisation wave velocity increases with increasing excitation frequency. This behaviour is observed for all investigated array
4. Results

devices. Here, a possible explanation may lie in higher overvoltages upon incipient breakdowns at higher excitation frequencies (see section 4.1.4) and the associated current peaks of higher amplitude. The latter can lead to excited species generation on a shorter timescale after incipient breakdown in the first cavity. As adjacent cavities where breakdown has not occurred yet still exhibit vacuum potential, charge carriers drift towards the latter. Due to higher overvoltages the electric field gradient between ignited and adjacent cavity under vacuum field conditions is higher at higher excitation frequencies. This allows higher ion and electron drift velocities. Together with a faster species generation this can yield an increased effective ionisation wave velocity.

Determined ionisation wave velocities are significantly smaller than the speed of light and too high to be caused by sound waves, thermal velocity of gas particles or diffusion of metastable species which is demonstrated in the following.

The velocity of sound waves $c_s$ in a neutral gas is described by [91]:

$$c_s = \left( \frac{\gamma RT}{M} \right)^{\frac{1}{2}} \quad (4.5)$$

where $\gamma$ is the adiabatic invariant, $R = 8.314511 \text{ J K}^{-1} \text{ mol}^{-1}$ [92] is the ideal gas constant, $T$ is the gas temperature, and $M$ is the molar mass of the gas molecules.

For argon as an atomic gas, the only degrees of freedom for particle motion are translation in spatial directions ($\gamma = 1.67$ for standard conditions). Considering a molar mass of $M = 3.995 \times 10^{-2} \text{ kg}$ [92] and assuming a gas temperature of 300 K, the respective sound velocity in argon can be derived:

$$c_{s,Ar} = 323 \frac{\text{m}}{\text{s}} \quad (4.6)$$

The mean thermal velocity $\bar{v}$ of neutral gas particles under the assumption of a Maxwellian distribution is [93]:

$$\bar{v} = \left( \frac{8k_BT}{\pi m} \right)^{\frac{1}{2}} \quad (4.7)$$
where \( k_B = 1.380658 \cdot 10^{-23} \text{ J K}^{-1} \) [92] is the Boltzmann constant, and \( m \) is the molecular mass of the gas.

For argon at 300 K, the mean thermal velocity can be calculated:

\[
\bar{v}_{Ar} = 399 \frac{\text{m}}{\text{s}}
\]  

(4.8)

The metastable species diffusion velocity \( v_D \) is determined by the binary diffusion coefficient \( D \) and metastable density \( n \) [93]:

\[
v_D = D \frac{\nabla n}{n}
\]  

(4.9)

where \( D \) is determined by the reduced collision diameter and the collision integral of the metastable species and the process gas [94].

Using tabulated Lennard-Jones parameters [95], the diffusion constant for argon metastables can be calculated:

\[
D = 1.85 \cdot 10^{-5} \frac{\text{m}^2}{\text{s}}
\]  

(4.10)

Under the assumption of a metastable density depletion over a distance of 100 µm, the diffusion velocity \( v_{D,Ar}^{m} \) for argon metastables can be derived to:

\[
v_{D,Ar}^{m} = 3.7 \cdot 10^{-1} \frac{\text{m}}{\text{s}}
\]  

(4.11)

It is obvious, that the velocities derived above are significantly smaller than that of the observed ionisation waves. The underlying processes thus have to deviate from those discussed above.

In order to exclude energy transfer through radiation as a source of wave propagation, the optical thickness of the investigated microplasmas would have to be determined. In the optically thin case, the wave velocity is too small to be induced by radiation and radiation absorption is low. In contrast, for an optically thick plasma, radiation trapping can be effective and thus reduce the ‘radiation velocity’. In this case, radiation could contribute to the observed
4. Results

ionisation wave propagation. However, in order to determine whether the plasma is optically thick or thin, the self-absorption of respective emission lines would have to be determined. Due to geometrical restrictions of the design of the investigated array devices, measurements based on transmission of radiation are not possible. Determination of the optical thickness based on line-broadening is in this case inhibited due to the high pressure regime and resulting pressure broadening of emission lines. Thus, a contribution of radiation absorption to ionisation wave propagation cannot be excluded. However, the comparably small ionisation wave velocity rather suggests a non-radiative mechanism. Moreover, preferably resonant transitions are susceptible to re-absorption of radiation due to the highly populated ground state in atmospheric pressure low-temperature discharges. The theoretical energy gain upon absorption alone would not be sufficient to lead to ionisation as needed in case of the observed ionisation waves. In the later course of this study, it is demonstrated that ionisation wave propagation due to radiation transport can be excluded (see section 4.2.3). However, photodesorption of surface charges can further enhance electron generation and contribute to ionisation wave propagation [96].

As diagnostic access to ionisation waves is rather limited, further investigation of this phenomenon with the help of plasma simulations is performed in the following.

Simulation of ionisation waves

Plasma simulation of the investigated devices in three dimensions is rather complex and exceeds available computer power. In order to simulate a microplasma array, the simulated geometry has to be simplified and degrees of freedom have to be reduced respectively. However, observation of ionisation waves in simulations requires at least two dimensions in space. Here, it is sufficient to examine a sub-system of linearly arranged cavities in order to observe ionisation waves. The two dimensions comprise the direction perpendicular to the wafer principal plane as well as one direction in the wafer principal plane. However, the choice of this geometry neglects the finite expansion of the cavities in the third dimension. In fact, this geometry reflects an arrangement of three parallel infinitely expanded discharge channels with equilateral triangular cross section. However, this geometry approximates the pyramidal cavity arrangement as much as possible in two dimensions. Moreover, the
general discharge behaviour of the discharge channel device is found to be in accordance with that of the array devices.

To quantify the validity of these statements, a two-dimensional simulation of the vacuum potential characteristics of a single 50 µm pyramidal cavity is performed using the commercial simulation software ‘COMSOL Multiphysics 3.5’ for two cases. In one case, rotational symmetry is assumed, where the axis of symmetry is chosen in the centre of the cavity, perpendicular to the Si wafer principal plane. In the second case, mirror symmetry is assumed for the same geometry. While rotational symmetry best approximates the real tridimensional geometry of the cavity in a two-dimensional model, mirror symmetry equals the conditions in the envisaged simulation of the ionisation wave. In the COMSOL simulation, Poisson’s equation is solved for a potential of 320 V applied to the Ni grid and a grounded Si wafer. Periodical boundary conditions are chosen for remaining boundaries of the computational domain. The resulting contour plots for the potential are shown in figure 4.17. The potential contour plot linearly scales from 320 V (red) to 0 V (blue). Equipotential lines are indicated by solid white lines. Black arrows indicate electric field strength (length of the arrow) and direction (direction the arrow points at). It can be discerned that potential and electric field characteristics calculated for both symmetries slightly deviate. However, the fundamental characteristics are preserved and deviation of absolute values is comparably small. Thus, the approximation of the complex three-dimensional array geometry by the simple two-dimensional model in case of the simulation of the ionisation waves is adequate.

In cooperation with the Institute of Theoretical Electrical Engineering at the Ruhr-Universität Bochum, a two-dimensional simulation of a 50x50 50 µm pyramidal cavity array is set up and run by Alexander Wollny [97, 98]. The dynamics of the device are studied on the basis of the hybrid modelling platform ‘nonPDPsim’. The platform is based on a fluid model and described in detail in the literature [26, 99, 100]. Here, only relevant physical equations and species taken into account are briefly discussed.

In case of this simulation, a linear array of three cavities is examined. The simulated array geometry is consistent with that depicted in figure 1.1 for three cavities instead of two. The cavities are denominated I to III in the order of their spatial alignment from left to right. The computational domain extends 600 µm to the left and right hand side of the wafer as well as 900 µm perpendicular to the Si wafer principal plane. As mentioned above, this cavity geometry
4. Results

Figure 4.17.: Vacuum potential contour plot as simulated in rotational and mirror symmetry for a 50 µm pyramidal cavity. The rotational and mirror axis respectively is indicated by the dashed black line. The potential applied to the Ni grid is 320 V (red) while the Si wafer is grounded (blue). Equipotential lines are indicated by solid white lines. Electric field direction and strength are indicated by black arrow direction and length respectively.
is only an approximation of the experimental one. The Si wafer which is in contact with the boundary exhibits a conductivity of 0.17 Ωcm, where the secondary electron coefficient is 0.15 at all surfaces.

The model treats Maxwell’s equations in the electrostatic approximation. This means, Poisson’s equation is used to calculate the electrostatic potential and is self-consistently coupled with drift-diffusion equations for charged particles as well as with the surface charge balance equation. A Newton iteration technique is used to simultaneously integrate this set of equations. Subsequently, the electron energy equation is solved and thus the electron temperature is implicitly updated. Non-Maxwellian electron energy distribution functions are taken into account in form of electron transport coefficients and rate coefficients for chemical reactions which are obtained from solving the zero-dimensional Boltzmann’s equation for the former. An atmospheric pressure argon gas atmosphere at 300 K is assumed. The species considered in the model and simulation are electrons, neutral atoms (Ar), an effective metastable atomic state (Ar*), an effective highly excited atomic state (Ar**), ions (Ar+), excimers (Ar2*) as well as excimer ions (Ar2+). Among these species, 28 chemical reactions are taken into account [101].

Boundary conditions of the computational domain are chosen to be ground for the electric potential. As the ionisation wave as observed in experiments is expected to cover the distance of three cavities on a very short time scale, the change of applied voltage in this time interval is negligibly small. Thus, a constant potential of −500 V is applied to the Ni grid in the simulation which sufficiently approximates the voltage behaviour in experiment. The Si wafer in the simulation is grounded. Consequently, the simulation covers the negative half period of the excitation period in the experiment. The simulated time is chosen to be 300 ns. In order to initiate a discharge in cavity I, an initial Gaussian quasi-neutral charge density distribution of argon ions and electrons with a peak value of 5 · 10^10 cm^{-3} and an extension of 10 µm is assumed at the left vertex of the cavity.

The temporal evolution of the electron density in proximity of the discharge cavities as derived from the simulation is illustrated in figure 4.18. The temporal evolution of two-dimensional maps of the electron density are plotted in images (a) to (f). The corresponding scale reading is given at the bottom of the figure. The height scaling of the images starts at 50 µm, while the Si wafer further extends to 0 µm. Similarly, the width scaling starts at 600 µm and ends at 900 µm while the computational domain extends from 0 µm to 1500 µm.
4. Results

Simulated temporal evolution of electron density for a 3x1 linear 50 µm pyramidal cavity array [98].

**Figure 4.18.** Simulated temporal evolution of electron density for a 3x1 linear 50 µm pyramidal cavity array [98].
It can be discerned from the simulation, that the initial electrons are drawn into cavity I while the ions drift towards the Ni grid due to electrode polarity (a). Once the ions impinge on the dielectric surface of the Ni grid, secondary electron emission is triggered. These generated electrons are repelled by the negative Ni grid potential and accelerated towards the centre of cavity I as well. Once these electrons gain sufficient kinetic energy, they can excite and ionise background gas. This leads to further ion and electron generation and an avalanche multiplication effect while the discharge spreads on top of the dielectric as expected in a DBD. Generated electrons accumulate inside cavity I until a critical electron density of $10^{15}$ cm$^{-3}$ is reached (b). At this point, a quasi-neutral area fills cavity I due to shielding of potential, which can be interpreted as a transition from Townsend to glow discharge. The discharge spreads over the cavity edges by charging of the top dielectric. Thus, a horizontal component of the electric field is produced. The small path between the quasi-neutral area and the Ni grid is now dominated by a strong cathode fall which promotes volume ionisation. Ions generated above the dielectrically coated Ni grid further cause secondary electron emission. Electrons generated this way at the Ni grid between cavity I and cavity II drift into cavity II due to the strong electric field between the top of the dielectric and the bottom of the cavity and further excite and ionise on their trajectory. While electrons accumulate in cavity II, a Townsend discharge is formed between the cavity centre and the Ni grid to its left hand side (c). Again, once a critical electron density is reached in cavity II, a transition from Townsend to glow discharge is observed. Generated ions drift towards the Ni grid separating cavity II and cavity III (d). The process described above for cavity II repeats for cavity III, where a Townsend discharge forms (e) 85 ns after glow mode transition in cavity II. Finally, a glow discharge forms in cavity III (f), whereupon all three cavities are connected by a quasi-neutral area. Thus, the successive discharge onset and ionisation wave propagation consist of two phases. In the first phase, secondary electrons freely drifting into the adjacent cavity and initiating an avalanche dominate the propagation. In the second phase, ions produced inside the cavity drift towards the cathodes. The effective ionisation path is roughly covered by electron and ion drift in equal parts. Due to the comparably smaller ion mobility, the ionisation wave propagation velocity is limited by the drift velocity of the ions. For a mean reduced electric field of $E/n = 2.1 \cdot 10^2$ Td as derived from the simulation, one finds an argon ion drift velocity $v_{d,Ar^+}$ of [102]:
4. Results

Simulated potential for a 3x1 linear 50 µm pyramidal cavity array corresponding to image (c) in figure 4.18 [98].

\[ v_{d,Ar^+} = \mu E = 570 \, \text{m/s} \]  \hspace{1cm} (4.12)

The comparably faster electron drift can be assumed to occur instantaneously. Thus, the combined ion and electron drift velocity agrees fairly well with the ionisation wave propagation velocity observed in the experiment and in the simulation.

To illustrate the different discharge modes described above, figure 4.19 shows a two-dimensional map of the electric potential corresponding to image (c) and \( t = 75 \, \text{ns} \) in figure 4.18. Three stages of discharge development can be distinguished: While in cavity III the electric field is dominated by the vacuum potential, a glow discharge with quasi-neutral bulk has formed in cavity I. Most of the potential in cavity I drops across the dielectric coatings of anode and cathode. Cavity II exhibits a conducting channel which extends from its centre towards cavity I and indicates the onset of a transition from Townsend to glow discharge.

The electron densities of up to \( 10^{15} \, \text{cm}^{-3} \) derived from this simulation exceed the typical values given in table 2.1 by several orders of magnitude. However, it has to be considered that the discharge dimensions and electrode gap size in case of the microplasma arrays are much smaller (\( \sim 10 \, \mu\text{m} \)) compared to the
4. Results

parallel plate geometries considered in the table (~ 1 mm). As the devices are operated at similar peak-to-peak voltages, the microplasma arrays can exhibit higher electric fields and thus higher electron densities.

Moreover, the observed discharge behaviour agrees well with the experimental findings. The transition from Townsend to glow discharge during the evolution of one emission burst for the cavities is well reproduced in the simulation. Furthermore, ionisation wave propagation is observed as well and can for the first time be ascribed to ion drift. The ionisation wave propagation velocity derived from the simulation is approximately 1 km s\(^{-1}\). It is slightly smaller than the velocities observed in experiment (compare figure 4.16) but still in the same order of magnitude. This deviation can likely be explained by the fact that the ionisation wave velocity derived from the simulation results from a first ignition of the investigated cavities, while in experiments a steady state is observed which is influenced by the memory effect. Moreover, the simulation only covers a linear array of three cavities which results in a comparably higher error compared to experimental determination of the ionisation wave velocity comprising a significantly higher number of cavities. Apart from that, the simulation does not take into account nitrogen impurities or radiation. While the absence of the former leads to a different plasma chemistry compared to that in the experiment, the absence of the latter does not allow photodesorption of surface charges.

The propagation and formation of the ionisation waves investigated in this section as well as integral observations strongly depend on single cavity dynamics. In order to understand the impact of single cavity dynamics on collective array behaviour, single cavity behaviour in an array arrangement is thus investigated by means of PROES and PMT measurements in the following section.

Summary

For each emission burst observed for investigated array devices, a sequential ignition of adjacent cavities in form of an isotropic ionisation wave is observed irrespective of Ni grid polarity. The starting point of these waves is most likely determined by fabrication irregularities of single cavities which are thus more likely to ignite due to Paschen’s law and is found to strongly depend on gas pressure. The ionisation wave velocity is found to increase with increasing excitation frequency and is in the order of 1-10 km s\(^{-1}\). Simulation of a linear cav-
ity array reveals that the driving mechanism of ionisation wave propagation is ion drift. The simulation further reproduces the transition from Townsend to glow discharge during one emission burst for single cavities.

### 4.2.2. Individual microplasma behaviour

Apart from the observed collective phenomenon of ionisation waves, single cavities in investigated array arrangements show a strong modulation of individual emission behaviour. In the following, this modulation is investigated on the basis of two-dimensional PROES measurements perpendicular to the devices principal plane combined with PMT measurements. This way, the individual cavity emission behaviour can be compared to the collective emission of the whole device.

#### Integral and individual cavity emission characteristics

For all investigated array devices, the device surface is imaged using the PROES setup with a spatial resolution of 3.58 µm and gate times and temporal resolution of 100 ns respectively. The emission intensity behaviour of single cavities during the excitation period is investigated by integration of the emission intensity over the area of one cavity in the PROES images. Consequently, the emission characteristics of a single cavity comparable to PMT measurements is obtained. At the same time, PMT measurements of the integral device emission intensity are performed.

Figure 4.20 exemplarily illustrates obtained emission characteristics of a single cavity in comparison to the integral device for the positive (a) and negative (b) half period of a 50x50 100 µm pyramidal cavity array device. It is operated in atmospheric pressure argon at an excitation frequency of 2 kHz and 800 V peak-to-peak voltage. Displayed emission intensities are in each case normalised to the respective maximum emission intensity in the positive half period. Both graphs also show the applied voltage waveform for better orientation.

For both half periods, three emission bursts are observed for the integral device as well as for the single cavity. While the periodicity of the emission bursts is the same for both signals, the FWHM of respective emission bursts is significantly smaller for the single cavity. This is due to the delayed successive ignition of the single cavities in the array arrangement in form of an ionisation
4. Results

Figure 4.20.: Integral and individual emission intensity oscillograms for the positive (a) and negative (b) half period of a 50x50 100 µm pyramidal cavity array device operated in atmospheric pressure argon at an excitation frequency of 2 kHz and a peak-to-peak voltage of 800 V. Integral emission intensity is registered with the PMT while individual emission intensity is derived from PROES measurements with a spatial and temporal resolution of 3.58 µm and 100 ns respectively by integration over the area of a single cavity. Both signals are normalised to the respective maximum emission intensity in the positive half period. The applied voltage is in each case indicated for better orientation.
4. Results

Comparison of incipient emission burst properties of the array as a whole and of a single cavity as recorded for a 50x50 100 µm pyramidal cavity array device for positive and negative half period. The device is operated in atmospheric pressure argon at an excitation frequency of 2 kHz and 800 V peak-to-peak voltage. Displayed quantities are derived from figure 4.20, where PMT signals and PROES recorded with the ICCD camera are used to document the emission behaviour of the device as a whole and that of a single cavity.

<table>
<thead>
<tr>
<th></th>
<th>pHP Peak value [a.u.]</th>
<th>pHP FWHM [µs]</th>
<th>nHP Peak value [a.u.]</th>
<th>nHP FWHM [µs]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Whole device</td>
<td>1</td>
<td>5.33</td>
<td>0.56</td>
<td>5.82</td>
</tr>
<tr>
<td>Single cavity</td>
<td>1</td>
<td>2.16</td>
<td>0.44</td>
<td>2.43</td>
</tr>
</tbody>
</table>

Significant difference between integral and single cavity emission characteristics is observed with respect to the relative ratios of emission burst peak values and superposing increasing continuous emission within one half period. While a moderate increase of the continuous emission is observed for single cavities (< 15% of maximum emission intensity), the increase registered for the integral emission reaches almost half of the maximum intensity. This can be understood as at any time after the incipient onset, the number of cavities which simultaneously exhibit a burst is small compared to the number of cavities which have already passed this phase. Thus, the simultaneous contribution of continuous emission background from single cavities is comparably higher for the integral device.

Before the increasing continuous emission is investigated in more detail, table 4.5 quantifies the ratio of incipient emission burst peak and FWHM values for the positive and negative half period comparing single cavities and integral devices as derived from figure 4.20.

As observed earlier in section 4.1.3, the FWHM of emission bursts in the nega-
4. Results

tive half period is higher than of those in the positive half period for the integral device. This is now confirmed for single cavities as well. Thus, statements as to the periodicity and relative FWHM characteristics of emission bursts of the integral devices and associated conclusions are still valid for single cavities, although absolute values may deviate.

Moreover, it can be discerned that the ionisation wave propagation velocity is slightly smaller in the negative half period from the comparably higher emission burst FWHM value for the whole device. This can be understood, as discharge onset occurs at comparably smaller overvoltages in this half period which is deduced from the higher FWHM and smaller peak values for emission bursts of single cavities. This implies a smaller electric field gradient between ignited cavities and neighboured cavities under vacuum electric field conditions. Thus, a comparably smaller ion drift and ionisation wave propagation velocity results in the negative half period compared to the positive one for the 50x50 100 µm pyramidal cavity array device. A possible explanation for the asymmetric ignition behaviour of single cavities will be discussed in section 4.2.3.

These results in combination with the observation of the ionisation waves demand a separate consideration of single cavities as well as of collective phenomena such as the ionisation waves. Although integral diagnostics suggest a similar behaviour of the devices as a whole compared to DBD setups in parallel plate geometry, dynamics of the array devices investigated in this thesis are much more complex. Thus, models which can be used for parallel plate setups cannot be directly transferred to the array devices and have to be modified.

Build-up of continuous emission background

The described and discussed build-up of the continuous emission background with increasing number of emission bursts within a pulse train per half period is investigated in more detail in the following. As it is obvious from figure 4.20, this phenomenon is not only observed for the device as a whole but also for single cavities. In order to resolve the spectrally integrated optical emission of a single cavity in time and space, PROES image series are recorded for various MSE array devices perpendicular to the devices’ principal plane and with high spatial resolution. As the observations made are similar for all devices, the results obtained for a 50x50 100 µm pyramidal cavity array device are discussed in detail.
4. Results

The device is operated in atmospheric pressure argon at an excitation frequency of 1 kHz and 800 V peak-to-peak voltage. PROES image series are recorded using gate times and time steps of 500 ns with a spatial resolution of 3.58 µm.

Figure 4.21 comprises the relevant findings. At the top of the figure, a false colour PROES plot shows the temporal evolution of the emission intensity spatially resolved along the perpendicular bisector of the cavity base side. The plot is divided into pH and nHP corresponding to respective Ni grid polarities. Only the times within the excitation period when emission is observed are displayed in the plot (80 µs in the positive half period, 100 µs in the negative one). The resulting discontinuity of the abscissa is indicated by a dashed white line. For each half period, the abscissa scaling is 10 µs.

In each half period, a series of five emission bursts can be identified, where the fifth burst is cut off in the positive half period. Moreover, pronounced emission at the cavity edges is observed for each emission burst, irrespective of Ni grid polarity. While the gradient of emission intensity drop towards the exterior of the cavity is rather steep, significant emission intensity is located in the cavity centre. It can also be discerned from the plot, that with increasing number of emission bursts a continuous emission background builds up in the cavity.

The time intervals between successive emission bursts in the negative half period are larger. Furthermore, maximum emission intensity in the negative half period is smaller than in the positive one. This is consistent with the findings in section 4.1.4. Emission features are more spacious and emission maxima more distant in the positive compared to the negative half period.

In order to quantify and discuss the observations in the PROES plot, emission intensity profiles along the perpendicular bisector of the cavity base side (along the ordinate of the plot) at points in time of maximum emission for each emission burst are deduced from the PROES plot. Resulting profiles are displayed in (a) and (b) for positive and negative half period respectively, where emission intensity is normalised to the maximum emission intensity peak value. The spatial range displayed in each case comprises the cavity cross section as well as half of the spacing towards adjacent cavities. The locations of the cavity edges are indicated by dashed lines in the graphs.

For the positive half period (a) the emission is peaked close to the cavity edges for each emission burst. The peak intensity is significantly higher for the incipient emission burst compared to subsequent bursts. While the peak inten-
Figure 4.21.: False colour PROES plot (at the top) of the emission intensity of a single cavity of a 50x50 100 µm pyramidal cavity array device operated in atmospheric pressure argon at an excitation frequency of 1 kHz and 800 V peak-to-peak voltage. While the ordinate represents spatial resolution along the perpendicular bisector of the cavity base side, the abscissa represents time (10 µs scaling). Temporal and spatial resolution are 3.58 µm and 500 ns respectively. The plot is divided into pH and nHP corresponding to respective Ni grid polarity, where a discontinuity of the abscissa is marked with a dashed line in the image. Emission intensity profiles along the spatial dimension derived from the PROES plot at the points in time of maximum emission for each emission burst are given for pH (a) and nHP (b).


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sity close to the cavity edges does not significantly deviate for the subsequent bursts, the emission intensity in the inner part of the cavity increases with increasing number of bursts. Moreover, the emission intensity does not decrease to zero in between adjacent cavities.

For the negative half period ($b$) the same general behaviour is observed. However, the emission maxima are in this case located closer to the cavity centre. The increase of the emission intensity in the cavity centre compared to the peaked intensity close to the edges is more obvious in this half period. For the fifth burst which is not fully developed in the positive half period, a rather flat intensity profile is observed. No peaks close to the cavity edges can be distinguished any longer. Other than in the positive half period, in the negative half period the emission intensity decreases to zero between adjacent cavities.

The observed peaked emission close to the cavity edges is most probably due to direct electron impact excitation. The vacuum electric field is highest at the cavity edges, as the separation between powered Ni grid and grounded Si wafer is smallest at this point. This can be discerned from figure 4.17. For pseudoglow rare gas DBDs in parallel plate geometry, it is observed that the amplitude of discharge current peaks within one pulse train successively diminishes [70]. This originates from an increased number of remanent excited species in the discharge volume with higher pulse order. Thus, less overvoltage is needed in order to initiate emission bursts of higher order and consequently associated discharge current peaks decrease [78]. This might explain the decreasing peak values of emission intensity at the cavity edges. It can also be the reason for the increasing emission intensity measured in the cavity centre for increasing number of successive emission bursts. The number of remanent long-lived excited species which remain from previous emission bursts accumulates within the discharge cavity during one pulse train. Thus, for emission bursts of higher order, the contribution of such species to the total excitation increases. Outside the cavity, the effect of an increasing emission background is not observed.

These observations further encourage the interpretation of emission burst behaviour in section 4.1.4 which relates periodicity of emission bursts to an increasing number of long-lived species accumulating within one pulse train.

As the discerned asymmetry of emission behaviour comparing positive and negative half period will be discussed in detail in the following section, it is not further examined here.
4. Results

Figure 4.22.: Temporal emission intensity evolution at the cavity edge and centre in the positive ((a), (c)) and negative ((b), (d)) half period as derived from figure 4.21 for a 50x50 100 µm pyramidal cavity array device. Emission intensities are normalised to the emission intensity maximum at the cavity edges in the positive half period.
4. Results

Figure 4.21 shows a deviating emission intensity behaviour comparing cavity centre and cavity edge. To investigate this phenomenon, time resolved emission intensity profiles for the cavity edge and centre derived from figure 4.21 are illustrated in figure 4.22 for positive ((a), (c)) and negative ((b), (d)) half period. Emission intensities are normalised to the emission intensity maximum at the cavity edges in the positive half period.

The emission intensity evolution at the cavity edges in the positive (a) and negative (b) half period shows the same behaviour as observed for integral devices. With increasing number of successive emission bursts, the respective FWHM values increase. While the continuous emission background intensity increases, the superposing emission bursts exhibit decreasing peak values. This behaviour has been discussed in detail in section 4.1.4.

However, emission intensity in the cavity centre shows a deviating behaviour. For positive (c) and negative (d) half period, emission bursts occur with the same periodicity as observed at the cavity edges. Moreover, an increasing continuous background is observed and emission burst FWHM values increase for increasing number of emission bursts. While this agrees with the behaviour at the cavity edges, the emission burst peak values stay constant within a pulse train in the cavity centre. This is observed for the first time in the frame of this study and suggests that different excitation processes are involved here. The interpretation of different excitation mechanisms comparing cavity edge and centre demands spectroscopic investigations. This way, involved species may be identified. However, such examination cannot be performed in the frame of this thesis as previously discussed in section 3.3.2.

Moreover, the emission intensity in the cavity centre is significantly smaller compared to that at the cavity edges in the positive half period. This deviation is less pronounced in the negative half period. An explanation of the asymmetry comparing positive and negative half period requires further investigation of spatial single cavity dynamics and will be discussed in section 4.2.3.

**Summary**

Comparison of single cavity emission intensity behaviour to that of whole MSE array devices reveals that emission burst durations are significantly smaller for the single cavity, while the periodicity of emission bursts remains the same. This can be ascribed to the successive ignition of single cavities in an array arrangement in form of ionisation waves. Moreover, a continuous emission superposing the emission bursts which increases with increasing number of
successive bursts is found. It is associated with excitation through long-lived excited species which accumulate with increasing number of emission bursts during one pulse train. The emission intensity upon emission burst peaks is further peaked close to the cavity edges, where the vacuum electric field is maximal. These emission peaks are more pronounced for incipient emission bursts, which is interpreted as a cause of the diminishing amplitude of successive current pulses within one pulse train for pseudoglow discharges. An asymmetry of the spatial extension of the emission features within the cavity comparing positive and negative half period is observed and is discussed in the following section. Moreover, the emission intensity dynamics in the cavity centre deviates from that at the cavity edges, indicating that different excitation mechanisms are involved.

4.2.3. Asymmetry of positive and negative half period

Several asymmetric aspects with regard to the polarity of the Ni grid of investigated devices have been observed for MSE devices investigated in this study. Emission burst and current peak intensity are smaller in the negative half period (see figure 4.8 and figure 4.20). The applied voltage upon incipient emission burst onset deviates comparing positive and negative half period. Here, the device layout seems to influence if incipient emission burst onset occurs at lower applied voltages in the positive or negative half period (see figure 4.11). Independent of MSE array layout, emission features of single cavities in the positive half period are more spacious than in the negative one (see figure 4.21).

One reason for this asymmetric behaviour lies in the device geometry. Ni grid and Si wafer size and shape significantly deviate. Thus, the devices investigated here can be expected to show more asymmetric behaviour compared to DBDs operated in parallel plate geometry, which can be reduced to a one-dimensional problem. In particular, the asymmetric occurrence of emission features of single cavities comparing both half periods is discussed in the following.
Single cavity emission dynamics

Single cavity emission dynamics within one excitation period are observed using the ICCD camera. Pyramidal and straight cavity array devices are imaged under an angle of 90° with respect to the wafer principal plane using PROES with a spatial resolution of 3.58 µm. This way, a concept of single cavity discharge dynamics can be deduced from the image series.

Figure 4.23 exemplarily shows the results of ICCD PROES imaging of a 50x50 100 µm (a) and 50 µm (b) pyramidal cavity array device respectively. Both devices are operated in atmospheric pressure argon at an excitation frequency of 20 kHz and 800 V peak-to-peak voltage. The figure comprises false colour emission intensity distribution plots as recorded with the ICCD camera for a single cavity upon the intensity maximum of respective incipient emission bursts in the positive (at the top) and negative (at the bottom) half period for each device. In each case, the gate time of the ICCD camera is 100 ns. The central graphs show the emission intensity spatially resolved along the perpendicular bisector of the respective cavity base side averaged over the area indicated by red rectangles in the ICCD images. In (a), vertical black and red solid lines mark the position of emission maxima in the positive and negative half period respectively. In (b), they are given at the same relative positions of the cavity cross section as derived in (a) for better comparability of emission structure extension.

The 100 µm cavity exhibits peaked emission intensity close to the cavity edges, as already observed in figure 4.21. The spatial extension and intensity of the emission intensity feature are significantly higher in the positive half period. While the emission intensity here is peaked in a distance of 48 µm from the cavity centre, this distance is 32 µm in the negative half period. In case of the positive half period, significantly more emission intensity is observed exceeding the cavity edges compared to the negative half period.

The 50 µm cavity behaves similar to the 100 µm one. It exhibits a more spacious and intense emission feature in the positive half period which also exceeds the cavity edges. However, in case of the smaller cavity, no distinct peaked emission is observed close to the cavity edges. This is not an artifact of the imaging, but a real feature as the spatial resolution is sufficient to resolve peaked structures in this image. The smaller the cavity diameter, the closer are opposing cavity edges. Thus, once the cavity diameter falls below a critical value, electric fields and thus the peaked emission features at the cavity edges due to
4. Results

Figure 4.23.: Emission intensity profiles of single cavities of a 50x50 100 µm (a) and 50 µm (b) pyramidal cavity array device respectively. The devices are both operated in atmospheric pressure argon at an excitation frequency of 20 kHz and 800 V peak-to-peak voltage. Top and bottom false colour emission intensity distribution plots are recorded with the ICCD camera with gate times of 100 ns upon incipient emission burst maximum for the positive and negative half period respectively. Central graphs show the emission intensity spatially resolved along the perpendicular bisector of the cavity base side averaged over the area indicated by red rectangles in the ICCD images. Vertical black and red solid lines indicate the position of emission maxima in the positive and negative half period respectively in (a). In (b) they are given at the same relative positions of the cavity cross section as derived in (a) for better comparability of emission structure extension.
4. Results

Emission intensity distribution upon incipient emission burst maximum of the positive (a) and negative (b) half period in false colours for a 50x50 100 µm pyramidal cavity array device operated in atmospheric pressure argon at an excitation frequency of 20 kHz and 800 V peak-to-peak voltage. The images are recorded with the ICCD camera in parallel to the Si wafer principal plane with a spatial resolution of 3.58 µm and a gate width of 100 ns. The camera is focused on the very back row of cavities. The colour scaling in the images deviates: While in (a) the full range (blue to red) covers 32000 counts, that in (b) covers 2000 counts. Red solid lines indicate the location of the wafer surface (lower line) and maximum emission extension in the positive half period (upper line).

electron impact excitation merge, leading to the observed effect.

The observations made above are based on ICCD camera imaging with line integration along the principal axis. As single cavities have a complex tridimensional geometry, information on emission behaviour in direction of this axis is lacking. A consistent interpretation of the obtained data demands observations that allow to conclude emission behaviour in all three spatial dimensions. Before the observations made for figure 4.23 are further interpreted, results from PROES using the ICCD camera in an orientation parallel to the Si wafer principal plane are thus discussed in the following.

Figure 4.24 exemplarily shows false colour ICCD camera images taken upon incipient burst emission maxima in the positive (a) and negative (b) half period for a 50x50 100 µm pyramidal cavity array device operated at atmospheric pressure at an excitation frequency of 20 kHz and 800 V peak-to-peak voltage. The ICCD camera is aligned in parallel to the Si wafer principal plane and
focused on the very back row of cavities. The images are recorded at a spatial resolution of 3.58 µm and with a gate time of 100 ns. The colour scaling in the images deviates. While in (a) the full range (blue to red) covers 32000 counts, that in (b) covers 2000 counts. Horizontal red solid lines indicate the location of the wafer surface (lower line) and maximum emission extension in the positive half period (upper line).

It can be discerned from (a) that very intense emission is registered exceeding the cavities in the positive half period. Here, emission is observed up to 100 µm above the Si wafer surface (indicated by the upper red solid line). Moreover, the emission intensity increases comparing the cavities from right to left. This is a direct cause of an ionisation wave propagating from right to left. The bright homogeneously illuminated area of the image results from cavities which are not in the focus of the LDM. The brighter spot in the centre of the image is an artifact due to the optical imaging with the LDM.

In (b), the red solid lines indicate the wafer surface and a distance of 100 µm from the latter as well. Emission intensity registered in the negative half period is rather diffuse and much smaller compared to the positive one. Furthermore, the emission features of the single cavities do not exceed the cavities to the same degree. This has significant impact on the interpretation of the ionisation waves (see section 4.2.1). Radiative energy transport as a driving process for the ionisation waves can be excluded when this observation is implicated. As ionisation waves are observed in the negative half period as well while the emission does not exceed the cavity, radiative communication of adjacent cavities is prohibited.

These findings, together with the observations made in figure 4.23 are the same for straight cavity array devices. Both types of devices only disagree with regards to a critical cavity diameter, where peaked emission close to the cavity edges is not observed any longer. While this is the case for 50 µm cavities in pyramidal geometry, it is observed for 25 µm cavities in straight geometry. This is due to the different cavity geometry and resulting different electric field line characteristics of both types of cavities. Apart from cavity geometry, the choice of process gas as well as of dielectric materials can have an influence on the critical cavity size where formation of peaked emission at the cavity edges is observed.

In combination, figure 4.23 and figure 4.24 allow to deduce the tridimensional emission dynamics of single cavities. The observed behaviour suggests that
4. Results

Asymmetry of emission in positive and negative half period: Sketch of cavity cross sections in the positive (a) and negative (b) half period. Assumed electron trajectories are indicated by arrows, violet areas represent areas of most effective excitation.

Figure 4.25. Asymmetry of emission in positive and negative half period: Sketch of cavity cross sections in the positive (a) and negative (b) half period. Assumed electron trajectories are indicated by arrows, violet areas represent areas of most effective excitation.

in the positive half period excitation is most effective close to the upper cavity border while in the negative half period this is the case further inside the cavity. This can be understood when the Ni grid polarity and resulting electron trajectories are investigated.

Figure 4.25 shows a sketch of a single pyramidal cavity cross section and assumed electron trajectories (black arrows) as well as areas of most effective excitation (violet area) for positive (a) and negative (b) Ni grid polarity. For positive Ni grid polarity, electrons are accelerated out of the cavity centre towards the Ni grid. Electron density and energy reach an optimum for excitation close to the upper cavity edge leading to the observed emission features. As vacuum electric fields are strongest close to the cavity edges, peaked emission intensity is observed in this region. For reverse Ni grid polarity and electron trajectory direction, the electrons can most effectively excite further inside the cavity. This results in less spacious emission features as observed in figure 4.23. Moreover, only little emission intensity is observed outside the cavity (see figure 4.24).

This contributes to a higher emission intensity registered for emission bursts in the positive half period compared to the negative one. In the negative half period emission features are smaller.

Moreover, the discharge in the negative half period is more concentrated inside the cavity which accounts for a higher ratio of plasma-surface interface and plasma volume compared to the positive half period. As a consequence, the governing excitation and ionisation mechanisms in discharges of reverse Ni grid polarity might be different. This can result in deviating plasma parameters such as species densities and energies. Thus, the different surface-to-
volume ratios can be responsible for the different onset and current behaviour with respect to Ni grid polarity previously discussed.

Apart from the asymmetric emission dynamics, asymmetric electrode design also contributes to the observed behaviour. In contrast to discharges in parallel plate geometry, anode and cathode area significantly deviate in size for the devices investigated in this thesis. Moreover, the distance between both electrodes is not constant within the plasma volume. This can also lead to asymmetric discharge formation with regards to different Ni grid polarities.

As emission intensity is observed above the Si wafer surface (see figure 4.24), registered emission intensity features can contain patterns that are reflected at the Si wafer surface. In the following, an error estimation as to the observations made above is performed.

**Error estimation: Reflection of emission at the surface**

The incident emission intensity registered with the ICCD camera and attached LDM could either result from direct or reflected emission from the array surface. As shown in figure 4.24, emission structures are observed at a maximum distance \(d\) of 100 µm from the array surface. The LDM primary mirror diameter \(m\) of 96 mm and the working distance \(l\) of the LDM determine the maximum aperture angle \(\alpha\) for imaging as shown in figure 4.26. Here, \(\alpha\) derives from:

\[
\alpha = \arctan \left( \frac{m}{2l} \right) \quad (4.13)
\]

Thus, information of the device surface imaged onto one pixel of the sensitive part of the ICCD camera’s CCD chip could contain reflected light within the maximum aperture angle \(\alpha\) as shown in figure 4.27.

The resulting maximum error in the camera’s effective resolution relates to the displacement \(\Delta x\) of the reflected emission source in the wafer’s principal plane:

\[
\Delta x = d \cdot \tan (\alpha) \quad (4.14)
\]

\[
= d \cdot \frac{m}{2l} \quad (4.15)
\]
4. Results

Figure 4.26.: Sketch: LDM maximum aperture angle $\alpha$ resulting from working distance $l$ and primary mirror diameter $m$.

Figure 4.27.: Sketch: Influence of emission reflected at the device surface. For emission structures observed at a maximum distance of $d$ from the device surface, an uncertainty of $\Delta x$ in spatial resolution results from the LDM maximum aperture angle $\alpha$. 
4. Results

Accordingly, maximum resolution errors $\Delta x$ caused by reflected light from the device surface are in the range of 3.2 $\mu$m and 9.6 $\mu$m for working distances of 150 cm and 50 cm respectively. These errors are much smaller than the observed emission features, which measure at least 50 $\mu$m. Therefore, possible resolution errors caused by reflected light play a minor role and observed emission features can be attributed to direct incident emission originating from discharges in the cavities.

Summary

On the basis of two-dimensional PROES using an ICCD camera, the asymmetric emission behaviour of single cavities with respect to Ni grid polarity is investigated. It is found that in the positive half period, excitation is most effective close to the top of the cavity at the cavity edges. Significant emission intensity is registered outside the cavity, above the Si wafer surface. In the negative half period, excitation is most effective deeper inside the cavity. The resulting increased surface-to-volume ratio for negative Ni grid polarity is a possible explanation for different discharge onset and current behaviour comparing Ni grid polarities. Radiative energy transport as a source of ionisation waves can be excluded based on these observations. Moreover, it is found that no peaked emission intensity close to the cavity edges is observed, once the cavity size falls below a critical value.
5. Summary and outlook

In the frame of this study, it was achieved to investigate and describe the dynamics of single microplasma cavities in MSE array arrangements as well as their interaction under variation of operation parameters such as applied peak-to-peak voltage, excitation frequency, and gas pressure. This was realised by means of spectrally integrated two-dimensionally space and phase resolved optical emission spectroscopy using an ICCD camera and an attached long-distance microscope. The discharge mode of the investigated array devices was determined on the basis of voltage-current and photomultiplier tube measurements by comparison of observed behaviour to that of well-known dielectric barrier discharges in parallel plate geometry. The understanding of the processes responsible for single cavity dynamics and interaction is essential with respect to future tailoring of microplasma array devices to application demands and further scale-up.

The general comparability of the different MSE array devices was verified by voltage, current, and PMT measurements. All devices showed a synchronous self-pulsing of emission and current and comparable behaviour under variation of operation parameters. For each half period of the excitation period, a pulse train of current bursts was observed which is interrupted upon reversal of the applied voltage waveform slope.

In order to determine the discharge mode of the investigated devices, current peak behaviour under variation of operation parameters was examined. It was found, that with increasing excitation frequency the number of current peaks per pulse train decreases, their amplitude increases, and respective FWHM values decrease. It was thus discerned that at higher frequencies the number of long-lived excited species and surface charges originating from discharges in antecedent half periods is smaller so that higher overvoltages are required upon incipient burst onset. Consequently, less current peaks can evolve until the reversal of the slope of the applied voltage waveform. Typical derived current densities were in the order of 10 - 100 mA cm$^{-2}$.
The self-pulsing was further investigated by means of complementary PMT measurements. It was observed that continuous emission builds up during a pulse train and superposes the single emission bursts. This behaviour is attributed to the successive build-up of metastable species as well as surface charge densities during a pulse train. These species can directly ionise nitrogen impurity species through Penning ionisation or cause secondary electron emission at the dielectric surfaces and effectively reduce ionisation threshold. For an increase of applied peak-to-peak voltage, the number of emission bursts per half period increased while the applied voltage upon incipient burst onset decreased. The FWHM value of subsequent bursts within one pulse train increased with increasing order of the burst, while the peak values decrease. This is further verification for the successive build-up of surface charge and metastable densities with successive emission bursts, which reduce the over-voltage upon ignition and can influence subsequent half periods.

The observed voltage, current, and emission behaviour in general agrees with that observed for parallel plate dielectric barrier discharges in pseudoglow mode, which is thus assumed for the investigated array devices as well. This has significant influence on the interpretation of experimental observations, as for every emission burst, a quasi-neutral glow discharge is formed. It accounts for substantially different discharge behaviour compared to Townsend-like discharges, which exhibit almost undisturbed electric fields.

Interaction of adjacent cavities in the array arrangements was investigated using two-dimensionally space and phase resolved optical emission spectroscopy. For this purpose, a recording and evaluation software was developed. It was observed, that each emission burst is composed of a successive ignition of the single cavities in the array arrangements in an isotropic wave-like manner. These ionisation waves were reproducible for the same operation parameters and did not depend on the locations of electric contacts on the array device. The starting point strongly depended on gas pressure, so that a preferred ignition due to pd-scaling resulting from manufacturing irregularities of single cavities was concluded. Typical propagation velocities were in the order of 1 - 10 km s^{-1}.

In order to identify driving processes for ionisation wave propagation, experimental observations were complemented by a two-dimensional fluid simulation of a linear cavity arrangement. The simulation well reproduces the successive ignition of the adjacent cavities. The ionisation wave velocity resulting
from the simulation was $1 \text{ km s}^{-1}$. The combination of experiment and simulation allowed to attribute ionisation wave propagation to electron and ion drift. Moreover, the simulation reproduced the formation of a glow discharge, which agrees well with experimental observations.

In the experiment, the ionisation wave propagation velocity increased with increasing excitation frequency. This increase is attributed to higher overvoltages upon ignition at higher excitation frequencies. Consequently, the electric field gradient upon ignition towards neighboured cavities under vacuum electric field is higher and may promote faster charge carrier drift velocities.

All of the investigated array devices were found to exhibit an asymmetry of applied voltage upon incipient burst onset as well as of emission intensity comparing both half periods of the excitation period. Registered emission intensity was typically smaller for negative Ni grid polarity while the polarity of earlier onset was dependent on the array design. As single cavity dynamics determine the collective array behaviour, it was examined by means of two-dimensionally space and phase resolved optical emission spectroscopy in order to investigate its impact on wave propagation and on asymmetric behaviour.

For all devices, the general behaviour of single cavity emission under variation of operation parameters agreed well with the observations made for integral devices. While the periodicity is the same, pulse widths are smaller for single cavities due to the wave-like superposition which is registered for integral array devices. However, conclusions drawn for integral devices were found to be valid for single cavities as well.

Emission intensity was found not to be homogeneously distributed within single cavities, but to exhibit peak structures close to the cavity edges. In this area, vacuum electric field strength is highest due to the stacking of the electrode structures. It was thus discerned that these peaked emission structures mainly originate from electron impact excitation. While for positive Ni grid polarity emission was registered up to 100 µm above the cavity and peaked closely to the cavity edges, it was concentrated inside the cavity and peaked more to the centre of the cavity for negative Ni grid polarity. This has significant influence on the interpretation of the observed asymmetric behaviour as the surface to volume ratio is smaller for positive Ni grid polarity. Thus, weighting of different processes deviates comparing both Ni grid polarities which can result in different plasma properties and the observed asymmetries.
Moreover, radiation transport as main driving process of ionisation waves can be excluded based on this observation. Peaked emission at the cavity edges was only observed for cavities with a diameter bigger than 50 µm. For smaller cavity diameters, the peak structures superpose and build one effective central peak due to the proximity of opposite electrode edges.

Moreover, it was discerned that emission intensity in the cavity centre shows a significantly different temporal behaviour compared to that at the emission maxima. Both structures are superposed by an increasing continuous background and exhibit successively increasing FWHM values. While the peak intensity of the emission maxima decreases with increasing number of emission bursts per pulse train, that in the cavity centre remains constant. This indicates that substantially different excitation mechanisms prevail comparing emission maxima close to the cavity edges with the cavity centre.

Apart from the direct results presented before, this study illustrates the complexity of microplasma array systems due to single cavity and collective dynamics. The complicated diagnostic access due to geometric restrictions demands complementary simulations. However, the complex geometry and plasma chemistry of such MSE array devices cannot be fully implemented in current simulations due to lack of calculating capacity of present computers. Thus, simplifications of underlying models are required. In order to verify model assumptions, benchmark simulations, and verify processes responsible for the observed behaviour, further spectroscopic investigations are mandatory.

The results of this study suggest, that in particular the communication of subsequent half periods via long-lived species plays a major role in discharge dynamics. Here, metastable species and surface charges are most likely the main agents. If the drawback of interference of monoenergetic emission inside the ICCD camera and filter system can be resolved, this would yield huge application potential. It would allow a calibration of the system and thus phase and space resolved line-ratio measurements. In particular, in argon, the electron density \[83\], \[84\] and metastable densities \[103\] could be derived with temporal and spatial resolution.

Another way to temporally and spatially resolve argon metastable and argon ion densities on the dielectric surfaces and in the cavities could be provided by means of laser-induced fluorescence spectroscopy in combination with the ICCD camera filter system \[104\]. Ions could be excited from \(3d^2D_{9/2}\) to \(4p^2F_{7/2}\) at 661.5 nm and fluorescence to \(4s^2D_{5/2}\) can then be observed at 459 nm \[105\].
5. Summary and outlook

Excitation of neutral argon metastables can be realised in the range of 690 - 790 nm. One realised scheme is based on excitation at 772.38 nm ($1s_5$ to $2p_7$ in Paschen notation) and observation at 810.37 nm ($2p_7$ to $1s_4$) [106]. Determination of absolute species densities is sophisticated due to the collisional operation regime. However, relative profiles and trends can yield valuable information on species behaviour.

Due to the restricted diagnostic access, conclusions on dominant processes in MSE array devices so far are based on variation of operation parameters. Further progress could be made with the availability of new device layouts. A variation of inter-cavity spacing and cavity aspect ratios could yield valuable information on intra- and inter-cavity processes. Further knowledge of such processes would then allow the design of new devices which may exploit the former in order to achieve high duty-cycles at low peak-to-peak voltages.

In certain applications such as high-frequency imaging with homogeneous lighting or lab-on-a-chip integration, where the interaction with single cavities cannot be readily monitored, the ionisation waves may be unfavourable. A re-design of MSE array devices with dielectric walls in between single cavities would not allow ion and electron drift between adjacent cavities. Thus, the propagation of ionisation waves may be prevented.
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Appendix
A. Curriculum Vitae

Name: Henrik Böttner  
Date of birth: 7. November 1982  
Place of birth: Essen, Germany

Education

since 04/2008 Research assistant at the  
Institute for Experimental Physics II: Applied Plasma Physics,  
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Since 06/2008  
Fellow of the Ruhr University Research School,  
Doctoral Representative of the  
section Natural Sciences and Engineering

04/2008 – 06/2010  
Associate of the DFG Research Training Group 1051

10/2003 – 03/2008 Majored in physics at the Ruhr-Universität Bochum, Germany  
University degree: Diplom-Physiker, focus: Plasma physics

01/2007 – 03/2008  
Student assistant at the  
Institute for Experimental Physics II: Applied Plasma Physics,  
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Student assistant at the  
Institute for Experimental Physics V: Plasma and Atomic Physics,  
Ruhr-Universität Bochum, Germany

07/2002 – 04/2003 Civilian service at the St. Josef-Hospital, Bochum, Germany

08/1993 – 06/2002 Hellweg Gymnasium, Bochum, Germany  
Graduation: Allgemeine Hochschulreife (Abitur)
B. Publications

Parts of this work have already been published. The respective papers are marked with ♦.

• ‘Phase resolved optical emission spectroscopy of coaxial microplasma jet operated with He and Ar’

♦ ‘Excitation dynamics of micro-structured atmospheric pressure plasma arrays’
  H. Boettner, J. Waskoenig, D. O’Connell, T. L. Kim, P. A. Tchertchian, J. Winter
  and V. Schulz-von der Gathen
C. Acknowledgement

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