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Stochastic heating in asymmetric capacitively coupled RF discharges

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Abstract

Electron dynamics in a strongly asymmetric capacitively coupled radio-frequency (RF) discharge at low pressures is investigated by a combination of various diagnostics, analytical models and simulations. Electric fields in the sheath are measured phase and space resolved using fluorescence dip spectroscopy in krypton. The results are compared with a fluid sheath model. Experimentally obtained input parameters are used for the model. The excitation caused by beam-like highly energetic electrons is measured by phase resolved optical emission spectroscopy (PROES) and compared with the results of a hybrid Monte Carlo model based on the electric field resulting from the sheath model. The plasma itself is characterized by Langmuir probe measurements in terms of electron density, electron mean energy and electron energy distribution function (EEDF). The RF voltage and the current to the chamber wall are measured in parallel. At low pressures the plasma series resonance (PSR) effect is observed. It leads to high frequency oscillations of the current (non-sinusoidal RF current waveforms) and, consequently, to a faster sheath expansion. The measured current is compared with an analytical PSR model. Another analytical model using experimentally obtained input parameters determines the influence of beams of highly energetic electrons on the time averaged isotropic EEDF as measured by Langmuir probes. The main result is the observation of beams of highly energetic electrons during the sheath expansion phase, that are enhanced by the PSR effect. The paper shows that the nature of stochastic heating is closely related to electron beams and the PSR effect.

1. Introduction

Capacitively coupled radio frequency (CCRF) discharges have been used for processing applications for a long time and a long record of experimental and theoretical investigations exists. However, despite its technological importance, the complexity of power coupling mechanisms in CCRF discharges at low pressures is not yet fully understood. Several theories of stochastic heating exist [1–8]. A standard assumption in most of the work except [8] is a sinusoidal current waveform. This is well justified at relatively high pressures or generally in symmetric discharges. However, higher harmonics in the 100 MHz regime can arise and become even dominant in asymmetric discharges at low pressures of less than typically 10 Pa. The regime, in which the plasma series resonance (PSR) effect plays an important, non-negligible role, corresponds to the standard conditions for industrial applications. Therefore, a detailed investigation of this effect and the phenomena related to it is needed. In this paper stochastic heating and the PSR effect are investigated by combining various experimental diagnostics, analytical models and simulations.

In order to obtain a closer insight into these fundamental processes, one needs to investigate cause and effect of electron heating in CCRF discharges. The cause of electron stochastic heating is the electric field in the plasma boundary sheath and its temporal as well as spatial evolution within one radio-frequency (RF) period [9–12]. Its effect is the time and space dependent ionization in the plasma, which can be probed.
through excitation [10–17]. Insight into power dissipation requires temporal resolution on a nanosecond time scale within the RF period for the investigation of the electric fields as well as the excitation dynamics.

In order to measure electric fields in the sheath the Stark splitting of high Rydberg states of krypton was investigated experimentally and theoretically [18, 19]. Using the resulting Stark maps the electric fields within the sheath are measured by fluorescence dip spectroscopy (FDS). Here the technique is applied to krypton as a probe gas for the first time. Similar methods for electric field measurements have already been developed and applied to other molecular and atomic probe gases like BCI [20], NaK [21], CS [22], He [23–25], H [26, 27], argon [19, 28–32] and Xe [33]. Knowledge of the field allows the determination of voltages, charge densities and currents.

The excitation dynamics is investigated by phase resolved optical emission spectroscopy (PROES). Similar investigations of excitation dynamics have been performed before [10–17, 34–37]. Both diagnostics are non-intrusive and provide high temporal and spatial resolution on a nanosecond and sub-millimetre scale.

The plasma itself is characterized by radially resolved Langmuir probe measurements of the electron density, the mean electron energy and the electron energy distribution function (EEDF). The EEDF is a key parameter for understanding basic discharge dynamics. It determines dissociation, excitation and ionization processes. Temporal changes of the high energy tail of the EEDF in CCRF discharges within the RF period have been previously observed by time resolved measurements [10–17, 34–37]. PROES has demonstrated sensitivity to such phenomena with high spatial and temporal resolution.

The current to the chamber wall as well as the RF voltage applied to the powered electrode are measured in parallel. At low pressures, strong high frequency oscillations of the current are observed and the current is no longer sinusoidal [8, 10, 12, 38–42].

The measured spatio-temporal profiles of the electric field in the sheath are compared with the results of a fluid sheath model [44, 45] with experimentally obtained input parameters (pressure, voltage, electron density and electron temperature). The density of electrons above 12 eV is calculated space and time resolved by a hybrid Monte Carlo simulation using the electric fields resulting from the fluid model as input parameters [10, 46]. The result is compared with the experimentally determined spatio-temporal excitation. An analytical PSR model [38] is used to describe the measured RF current. Another analytical model, that also uses experimentally obtained input parameters, describes the influence of highly energetic electron beams on the time averaged EEDF in the bulk as measured by a Langmuir probe.

Space and phase resolved profiles of the electric field in the sheath and the excitation under various conditions are presented and phenomena such as electron beams and the PSR effect are discussed based on the information from probe, current and voltage measurements. The synergistic effect of all these diagnostics in combination with analytical models and simulations yields a more complete picture of electron heating in CCRF discharges at low pressures (stochastic heating).

The paper is structured in the following way: in section 2 the experimental setup for all diagnostics is described separately. In section 3 the PSR effect is introduced and in section 4 the results of this work are presented. Here the focus lies on the comparison between two different sets of parameters (10 and 1 Pa), for which all diagnostics were applied. The results of the different diagnostics are compared with each other and to simulations yielding a better understanding of electron heating under these conditions. Finally, conclusions are drawn in section 5.

2. Experimental setup

2.1. Electric field measurements

Electric fields in the boundary sheath of a strongly asymmetric CCRF discharge in krypton were measured via FDS using the Stark splitting of high Rydberg states of krypton atoms. The technique and the laser system used for these measurements are described in detail elsewhere [18]. For these investigations the $1\text{S}_0 \rightarrow 5\text{p}^2[3/2]_2$ transition was pumped at $2 \times 204.13$ nm (two-photon excitation). The fluorescence from the $5\text{p}^2[3/2]_2$ state to the $5\text{s}^2[1/2]_1$ state at 826 nm was monitored. A second tunable dye laser was used to excite from the $5\text{p}^2[3/2]_2$ state to high Rydberg states, which are shifted due to the Stark effect. Scanning the wavelength yields the dip spectrum. Different Rydberg levels with different field sensitivities were used depending on the electric field strength in the sheath at a given spatial position and phase.

The frequency-doubled output beam (532 nm) of a seeded Nd: YAG laser (Continuum, 9020) with a pulse width of 7 ns and a pulse energy of 900 mJ at a repetition rate of 20 Hz is split by a ratio of 2 : 1 to pump two tunable double-grating dye lasers (Radiant Dyes, Narrow Scan). The first one emits a fundamental wavelength of 612.4 nm, which is frequency-tripled for the two-photon excitation step (204.13 nm, 3 mJ).

The fundamental of the second dye laser (615–645 nm) is focused into a tube filled with 10 bar of H$_2$ gas, converting it into the desired wavelength range of 490–509 nm by stimulated anti-Stokes Raman scattering at approximately 150 µJ. A filter and dielectric mirrors block both the fundamental and Stokes component. The second laser beam is switched on and off by a mechanical shutter, so that drifts of the intensity (on the timescale of seconds) can be compensated. Both beams are guided collinearly into a modified GEC reference cell [47].

The metal cylinder surrounding the ICP antenna and the dielectric window were replaced by a monolithic quartz housing (figure 1). Here the inductive coupling was not used. The RF voltage is applied only to the lower electrode and the whole chamber wall acts as a grounded electrode. Therefore, the discharge is strongly asymmetric and almost the entire voltage drops across the sheath at the powered electrode. The electrode radius and the gap between electrode and quartz are both 5 cm.

The Q-switch of the Nd: YAG pump laser is synchronized with the RF generator (13.56 MHz) using a frequency divider
Figure 1. Experimental setup: GEC reference cell including all diagnostics (FDS, PROES, Langmuir probe, current and voltage measurements). The arrows indicate the direction of the current flow.

A wavelength scan of the second dye laser over a certain wavelength interval with a stepwidth of 3 pm was performed in the case of every measurement at a certain phase. The wavelength interval was chosen corresponding to the respective Rydberg state and its potential shift. For these measurements only nd'-levels were used because of their distinctive dips [18]. The fluorescence light was accumulated on the CCD for several seconds at each wavelength of the second dye laser and two images were taken, one with the shutter being open and one with the shutter being closed. An edge filter in front of the camera was used, which blocked all wavelengths below 780 nm. Then a difference image was calculated in order to determine the fluorescence decrease. Pixels in the horizontal direction were binned in order to reduce the noise. The spatial resolution was achieved by using a laser beam that covered 2 mm of the sheath and by moving the entire discharge chamber up and down leaving the laser beams at their original position. Using this technique the entire sheath ($s_{\text{max}} \approx 1$ cm under the conditions investigated) could be scanned. Finally, the wavelength position of the dips were identified at each spatial position and at each phase, respectively. The resulting shifts were then compared with a database of Stark shifts of Rydberg states in krypton [18] and the electric fields are determined spatially and temporally resolved.

Figure 2 shows an example for such a measurement in an argon discharge with 10% krypton admixture at the phase of maximum sheath expansion. The setup used for PROES is identical to the one shown in figure 1, but the laser system is not used and only the emission from the plasma is detected by an ICCD camera (Andor iStar). The camera is synchronized with the RF generator via a frequency divider (30 KHz). The Kr2p5-state at 758.7 nm is observed using an interference filter. The lifetime of 21.5 ns is short enough to resolve electron dynamics within one RF period at 13.56 MHz ($T_{\text{RF}} = 74$ ns).

The internal delay generator of the ICCD camera sets a certain delay between the trigger and the camera gate. Here the minimum camera gate width of 4.2 ns was used. The signal is acquired at a certain phase during several thousand RF periods. Then the delay is increased and the next phase is scanned. Here a step width of 2.1 ns was chosen. 38 steps were performed in order to scan a bit more than one RF period. Typical exposure times were 10 s. All images were binned in the horizontal direction in order to reduce the noise resulting in one dimensional spatial resolution along the discharge axis. The diagnostic of PROES is explained in detail elsewhere [12].

2.2. Phase resolved optical emission spectroscopy

In order to characterize the plasma in terms of electron density, electron mean energy and EEDF under each condition investigated, a Langmuir probe (SmartProbe, Scientific Systems) was inserted into the discharge from the side. Radial scans through the entire discharge volume were performed 2.5 cm above the powered electrode well outside the sheath.

Simultaneously with every other measurement, the RF voltage was measured at the output of the matching box directly in front of the powered electrode using a high voltage probe (LeCroy). The current to the chamber wall was also measured simultaneously by a SEERS current sensor (Plasmetrex) [48], which was integrated into the side wall of the GEC cell (see figure 1).
3. The plasma series resonance

In this paper, the focus lies on the synergistic use of different diagnostic techniques in combination with analytical models and simulations in order to study electron heating in CCRF discharges at low pressures. In that respect the PSR effect plays an important role. In asymmetric CCRF discharges the PSR arises from a periodic exchange between kinetic electron energy in the bulk and electric field energy in the sheaths. Due to its analogy to a series resonance between a capacitor and an inductance it is called plasma series resonance. In that context the space charge of the sheath provides a capacitive characteristic and the bulk can be considered as a series of inductance (electron inertia) and resistance (electron–neutral collisions). Different from a normal capacitor the charge voltage relation in the sheath is non-linear \[8, 38–43\].

The PSR effect leads to high frequency oscillations superimposed on the normal RF current. The frequency of these oscillations is of the order of 100 MHz and its amplitude is limited by collisions of the oscillating bulk electrons with the neutral background gas. These oscillations lead to a faster sheath expansion compared with regimes where sinusoidal RF voltage in the sheath is non-linear \[8, 38–43\].

These oscillations can be detected by a small sensor integrated in the reactor wall that picks up a fraction of the RF current (see figure 1). This diagnostic can be used as a non-invasive process monitoring technique in industry. It is known as self excited electron resonance spectroscopy (SEERS) \[48, 49\].

Figure 3 shows an example of such a measurement in a strongly asymmetric CCRF discharge in krypton at 0.5 Pa and 8 W. Due to the low pressure and, consequently, low damping strong high frequency oscillations of the current during the entire RF period are observed.

These oscillations can be described by a one dimensional analytical PSR model. In this model the so-called PSR equation is solved for current and charge \[38\]:

\[
\sin^2 \left( \frac{\Omega t}{2} \right) = q^2 + 2(\dot{q} + \kappa \dot{q}). \tag{1}
\]

Here \(q = Q/Q_m\) is the charge in the sheath normalized to the space charge \(Q_m = A_c \sqrt{2e\phi_0/\Phi_0}\) in the sheath at the phase of maximum negative applied voltage \(-\Phi_0\). \(\tau = \omega_0\), where \(\omega_0 = \gamma \omega_p\) is the plasma frequency reduced by a dimensionless geometry factor \(\gamma = \sqrt{s_m A/L A_s} < 1\) with \(s_m = \sqrt{2e\phi_0/\kappa n}\). \(A_s\) is the surface area of the powered electrode, \(A\) an effective value for the discharge area, \(L\) the discharge length and \(n_t\) the effective density in the sheath. \(\Omega = \omega_{RF}/\omega_0\) is the normalized RF frequency and \(\kappa = \nu/\omega_0\) the normalized electron–neutral elastic collision frequency.

The PSR equation is a balance equation between the externally applied voltage and the voltage that drops across the plasma in a totally asymmetric discharge. The voltage drop across the plasma is the sum of the voltage, that drops across the sheath and the bulk. The plasma is considered to be a series of a non-linear capacitor (sheath) and a resistance and inductance (bulk). In this context the voltage that drops across the sheath is assumed to be proportional to \(\sigma^2\), where \(\sigma\) is the surface charge density. This assumption is based on a static ion density profile with the approximation of a quasi-constant ion density and a step-function for the electron density. This presumption will be verified experimentally in this paper.

In this work this model will be used to describe measured currents theoretically by varying the two parameters \(\Omega\) and \(\kappa\) in equation (1).

4. Results

The focus of this work lies on the investigation of electron (stochastic) heating at low pressures including the PSR effect. Various plasma parameters are investigated in detail applying different diagnostics, analytical models and simulations.

Knowledge of the time and space resolved electric field in the sheath of CCRF discharges is essential for the understanding of basic mechanisms such as electron heating. It allows the determination of voltages, charge densities and currents. Furthermore, the field itself and its spatial as well as temporal evolution are the cause of electron stochastic heating. Therefore, a novel technique of FDS in krypton for space and phase resolved electric field measurements was developed \[18\] and is applied to a CCRF discharge for the first time. The results are compared with a fluid sheath model. Furthermore, excitation dynamics, voltage, current and other plasma parameters such as electron density, electron mean energy and EEDF are studied in a strongly asymmetric CCRF discharge under various conditions and compared with theoretical results.

The focus lies on the comparison of two specific sets of conditions, for which detailed investigations applying all diagnostics were done. Those are measurements in a strongly asymmetric krypton discharge at 10 Pa, 8 W and 1 Pa, 8 W, respectively. These two different pressures were chosen, since the PSR effect is more pronounced at low pressures. At 10 Pa
4.1. Results at 10 Pa

4.1.1. Current and voltage measurements. At low pressures the current starts to deviate from the form shown in figure 4. Figure 5 shows current and voltage measurements at 10 Pa and 8 W. At this pressure some small high frequency oscillations superimposed on the sinusoidal RF current become visible. These oscillations can be attributed to the PSR effect. However, the pressure and, therefore, the collision frequency at 10 Pa is still quite high and the oscillations are damped fast.

The figure also shows a comparison with an analytical PSR model \[38\] presented in section 3. At higher pressures the measured RF current waveforms are sinusoidal.

4.1.2. Langmuir probe measurements. Figure 6 shows radially resolved profiles of electron density and electron mean energy in the plasma bulk of a strongly asymmetric CCRF discharge at 10 Pa and 8 W in pure krypton 2.5 cm above the powered electrode. The maximum density is of the order of \(n_e \approx 7 \times 10^9 \text{ cm}^{-3}\) and the electron mean energy is of the order of \(\langle \varepsilon \rangle \approx 3 \text{ eV}\). Both profiles show shapes typical for CCRF discharges.

Figure 7 shows the measured EEDF \(E^{-1/2}\) in the radial centre of the discharge. It has a bi-Maxwellian shape typical for CCRF discharges at low pressures \[50–54\]. The enhancement of the high energy tail can be interpreted as the influence of stochastic heating, which is efficient at low pressures. Further details regarding the nature of stochastic heating will be presented in this paper. Besides the electron mean energy of all electrons the electron temperature \(T_e\) of the low energetic part of the EEDF and the electron temperature of the high energetic part \(T_h\) can be determined out of the respective slopes of the EEDF. In the radial centre those temperatures are \(T_e \approx 1 \text{ eV}\) and \(T_h \approx 5 \text{ eV}\).

The bi-Maxwellian shape of the measured EEDF is reproduced well by a hybrid Monte Carlo model of a symmetric CCRF discharge \[10,46,55\]. As only qualitative comparisons are performed here, it is important to note that bi-Maxwellian EEDFs are observed in the experiment and simulation.

4.1.3. Electric field measurements. Figure 8 shows the spatial and temporal evolution of the electric field in the boundary sheath of an asymmetric CCRF discharge at 10 Pa and 8 W in krypton at the powered electrode. Four different phases were investigated. The electric fields in the sheath shortly after the collapse at 7.5 ns, during the phase of sheath expansion (21.5 ns), shortly after the phase of maximum sheath expansion (39.5 ns) and during the sheath collapse (57.5 ns) are shown. The maximum sheath width at the phase of full sheath expansion can be identified to be \(s_{10 \text{ Pa}} \approx 5 \text{ mm}\). The phases of sheath expansion and collapse are clearly visible. In particular there is a big jump of the sheath edge between 7.5 ns and 21.5 ns.

The solid lines in figure 8 are the result of a fluid sheath model \[44,45\] using experimentally obtained input parameters such as pressure, voltage, electron density and electron temperature. Good agreement between experiment and simulation is found.

Following Poisson’s equation, \[2\]

\[
\frac{dE}{dx} = -\frac{e}{\varepsilon_0} (n_i - n_e),
\]

a steeper decrease of the electric field in the sheath \((n_e = 0 \text{ cm}^{-3})\) corresponds to a higher ion density. Here \(e\) is the elementary charge, \(\varepsilon_0\) the dielectric constant, \(n_e\) the electron density and \(n_i\) the ion density. Therefore, the ion density is lowest at the electrode and increases towards the sheath edge due to flux conservation. Close to the sheath edge high theory and experiment is good. At lower pressure the PSR oscillations are much more pronounced (see section 4.2.1).
energetic electrons can enter the sheath \((n_e \neq 0 \text{ cm}^{-3})\) and the absolute value of the slope decreases again. However, in this range the electric fields are close to the sensitivity limit of 50 V cm\(^{-1}\). Qualitatively, this agrees very well with the spatial shape of the electric field distribution shown in figure 8.

In order to check the reliability of the electric field measurements the voltage applied to the bottom electrode was measured at the output of the matchbox using a high voltage probe. Furthermore, the measured electric fields were integrated over the entire sheath at each phase, at which FDS measurements were performed. Under the conditions investigated experimentally here the electrostatic approximation holds \([56]\). Following

\[
E = -\nabla \Phi, \tag{3}
\]

this integration should yield the momentary RF voltage, if the measurement is correct.

The comparison of both ways of determining the momentary sheath voltage is shown in figure 9 and very good agreement is found.

In the sheath \((n_e = 0 \text{ cm}^{-3})\) Poisson’s equation is

\[
\frac{dE}{dx} = \frac{e}{\varepsilon_0} n_i. \tag{4}
\]

Integration of equation (4) from 0 to the momentary sheath edge \(s(t)\) yields

\[
E_{el} = \frac{e}{\varepsilon_0} \int_0^{s(t)} n_i \, dx, \tag{5}
\]

where \(E_{el}\) is the electric field at the electrode. The surface charge density \(\sigma\) at the electrode is defined as

\[
\sigma = e \int_0^{s(t)} n_i \, dx. \tag{6}
\]

Substitution of equation (6) into equation (5) yields a direct relation between the electric field at the electrode and the surface charge density:

\[
\sigma = \varepsilon_0 E_{el}. \tag{7}
\]
Using equation (7) and the measured electric fields at the electrode at different phases, the surface charge density $\sigma$ can be determined at different phases. The result is plotted against $\sqrt{U_s}$, where $U_s$ is the momentary sheath voltage. The result is shown in figure 9. Obviously, $U_s \propto \sigma^2$. (8)

This relation is an important fundamental assumption for a one dimensional PSR model [38] and has now been verified experimentally.

4.1.4. Excitation dynamics. Figure 10 shows the spatio-temporal excitation into Kr2p$_5$ close to the powered electrode under the same conditions as before.

The Kr2p$_5$-state was chosen, since the discharge is operated in pure krypton, this state has a short lifetime of 21.5 ns [57], and cascade contribution to its population density can be neglected [58]. Excitation out of metastable states is also neglected for the qualitative investigations performed here. Based on these assumptions the excitation $E_i(t)$ can be determined [14,34]:

$$E_i(t) = \frac{1}{A_{ik}} n_0 \left( \frac{dn_{ph,i}(t)}{dt} + A_i n_{ph,i}(t) \right).$$ (9)

Here $A_{ik}$ is the transition probability of the observed emission, $n_0$ the ground state density, $n_{ph,i}(t)$ the measured number of photons per unit volume and time, and $A_i = 1/\tau$ the decay rate, which is given by the inverse of the lifetime $\tau$.

An absolute phase calibration between the FDS and PROES measurements was performed, so that the time scales in figures 8 and 10 are identical. Based on figure 10 the sheath width can be estimated to be 5 mm. This agrees very well with the maximum sheath width $s_{10\text{ Pa}} \approx 5$ mm determined from the electric field measurements.

During the phase of fastest sheath expansion (7.5–21.5 ns, see figure 8) the most intense excitation is observed. At this phase a beam of highly energetic electrons is generated by the expanding sheath that penetrates into the plasma bulk (straight line in figure 10). During the time interval from 7.5 ns to 21.5 ns almost all electrons that have been located in the sheath region before are pushed out of the sheath by the electric field. The maximum excitation does not occur directly at the sheath edge (5 mm), but after one mean free path. Further inside the bulk the excitation decreases and the highly energetic directed beam electrons lose energy and their directed character through collisions. Similar phenomena have been observed in hydrogen [14, 15, 27] before. In the case of investigations performed in hydrogen, electron beams could not be directly observed, since the pressure was too high and the beams were immediately stopped behind the sheath. However, from these investigations at low pressures, the nature of this excitation mechanism in terms of electron beams is now obvious. From the slope of the beam trajectory the electron drift velocity
can be estimated. Under these conditions the drift velocity is
\(v_{d,10 \text{ Pa}} \approx 5 \times 10^5 \text{ m s}^{-1}\).

In this context the sheath edge is defined according to Brinkmann [45] as spatial position \(s\), where the following condition is fulfilled:

\[
\int_0^s n_e(x) \, dx = \int_s^{\infty} (n_i(x) - n_e(x)) \, dx,
\]

(10)

where \(x = 0\) is the position of the electrode. Here the sheath width is roughly estimated from spatio-temporal plots of the excitation to be the distance between the electrode and the spatial position where the excitation starts to increase significantly.

Figure 11 shows the result of a hybrid Monte Carlo model in terms of the integration of the calculated EEDF above 12 eV in a symmetric discharge in argon operated at 9.3 Pa [50]. Only the fraction of the discharge close to the bottom electrode is shown here. Similar to the experiment trajectories of beams of highly energetic electrons are observed, that are generated by the expanding sheath. As the simulated discharge is symmetric, an electron beam is also generated during the sheath expansion phase at the grounded electrode 180° phase shifted in comparison with the beam generated at the powered bottom electrode. In contrast to the experiment in the simulation a sinusoidal sheath motion is observed due to the fact that a symmetric discharge geometry is used. In the experiment the discharge is asymmetric and the PSR leads to very fast sheath expansions and non-sinusoidal sheath motions at low pressures. The same effect is even more pronounced at 1 Pa.

4.2. Results at 1 Pa

4.2.1. Current and voltage measurements. Figure 12 shows a comparison between the measured current to the plasma wall (SEERS sensor implemented into the side wall of the GEC chamber) and the modelled current using equation (1) at 1 Pa and 8 W. The measured RF voltage is also shown.

At high pressures (45 Pa) the current is nearly sinusoidal, as it is usually assumed in most studies, and the agreement with the PSR model is good (see figure 4). At an intermediate pressure of 10 Pa small high frequency oscillations superimposed on the sinusoidal shape become observable (see figure 5). At low pressures (1 Pa) the temporal characteristics of the current are completely different compared with 45 Pa. There are strong high frequency oscillations superimposed on the sinusoidal shape. These high frequency oscillations are caused by the PSR effect and have been observed theoretically before [8,38–42]. The current oscillations are well reproduced by the analytical PSR model (equation (1)) under the corresponding conditions. These high frequency oscillations lead to a faster sheath expansion and, therefore, enhance the generation of beams of highly energetic electrons [10].

4.2.2. Langmuir probe measurements. Figure 13 shows a comparison between an experimentally obtained EEDF measured close to the radial centre of the discharge at 1 Pa and 8 W 2.5 cm above the powered electrode and a theoretically calculated EEDF using an analytical model, which is explained in section 4.2.5. The agreement is good and a bi-Maxwellian shape is observed in both experiment and theory. At 1 Pa the electron mean energy is \(\langle \varepsilon \rangle \approx 5.1 \text{ eV}\), the temperature \(T_c\) of the low energetic part of the EEDF is \(T_c \approx 2.3 \text{ eV}\) and the electron temperature of the high energetic part \(T_h\) is \(T_h \approx 7 \text{ eV}\). The maximum electron density is \(n_e \approx 2 \times 10^9 \text{ cm}^{-3}\).

Bi-Maxwellian EEDFs are also observed using a hybrid Monte Carlo model of a symmetric discharge at similar pressures [55].

4.2.3. Electric field measurements. Figure 14 shows the spatial and temporal evolution of the electric field in the boundary sheath of an asymmetric CCRF discharge at 1 Pa and 8 W in krypton at the powered electrode. The markers correspond to measured fields using FDS and the solid lines to the result of a fluid sheath model [44,45] using experimentally determined input parameters. Good agreement between experiment and theory is found. Similarly to the situation at 10 Pa, several different phases were investigated.

Due to the lower pressure the maximum sheath width is bigger than in the 10 Pa case (see figure 8) and the electric
The surface charge density also at lower pressures. As before, the sheath voltage is proportional to the square of powered electrode using equation (7). The result is shown in figure 14. Integration of the time averaged field yields the time averaged potential $\Phi$ in the sheath at different positions (figure 16). As the ions in the discharge are much heavier than the electrons, the ions can only react to this time averaged potential.

Based on the conservation of ion energy and flux the following expression for the ion density $n_i(x)$ in the sheath as a function of the time averaged potential $\Phi(x)$ is derived:

$$n_i(x) = n_0 \left(1 - \frac{2e\Phi(x)}{Mv_0^2}\right)^{\frac{1}{2}}. \quad (11)$$

Here $M$ is the ion mass, $n_0$ the ion density in the bulk, which is equal to the electron density, and $v_0$ the Bohm-velocity, at which the ions enter the sheath.

The result in terms of the space resolved ion density in the sheath is shown in figure 17. The ion density is $2 \times 10^6$ cm$^{-3}$ at the electrode and increases by one order of magnitude towards the sheath edge located 1 cm away from the electrode due to flux conservation. The absolute value at the sheath edge of $n_i \approx 2 \times 10^6$ cm$^{-3}$ is the same as the electron density in the bulk ($n_e \approx 2 \times 10^9$ cm$^{-3}$) measured by a Langmuir probe (see section 4.2.2).

4.2.4. Excitation dynamics. Analogously to the investigations performed at 10 Pa and presented in the previous section, the effect of electron heating in terms of the spatio-temporal excitation, probed through the Kr 2p$^5$-state, is also investigated at 1 Pa. The result is shown in figure 18.

The maximum sheath width obtained from the spatio-temporal contour plot of the excitation is $s_{1\text{Pa}} \approx 1$ cm. This is again in good agreement with the electric field measurements (see figure 14). Similarly to the situation at a higher pressure of 10 Pa (see figure 10), the generation of a highly energetic electron beam is observed. From the slope of the corresponding trajectory the propagation velocity of the beam electrons can be estimated to be $v_{d,e,1\text{Pa}} \approx 2.3 \times 10^6$ m s$^{-1}$. In comparison with the observed drift velocity at higher pressures ($v_{d,e,10\text{Pa}} \approx 5 \times 10^5$ m s$^{-1}$), the beam electrons are substantially faster at lower pressures. This is caused by the bigger sheath width and the PSR effect, which plays an important role at 1 Pa. Both arguments cause a faster sheath expansion. The corresponding drift energy at 10 Pa is $E_{d,10\text{Pa}} \approx 1.5$ eV, whereas the drift energy at 1 Pa is $E_{d,1\text{Pa}} \approx 11$ eV. Because of this difference in energy, which is deposited in the plasma by beam electrons, it is well understandable that stochastic heating plays a significant role only at low pressures.

In contrast to the situation at 10 Pa there are additional excitation maxima during the later part of the RF period (20–74 ns) that occur after the tilted maximum caused by the electron beam. Those straight maxima are directly correlated with the high frequency oscillations of the RF current due to the PSR effect, that are well pronounced at 1 Pa in contrast to the 10 Pa case, where no additional excitation maxima are
Figure 15. Left graph: comparison between measured RF voltage (high voltage probe and sinusoidal fit) and sheath voltage determined out of the integration of the measured electric fields at different phases. Right graph: relation between sheath voltage $U_s$ and surface charge density at the electrode $\sigma$ at 1 Pa and 8 W.

Figure 16. Time averaged electric field (left graph) with a polynomial fit and time averaged potential (right graph) in the sheath determined from the fit at 1 Pa and 8 W in a pure Kr discharge.

observed. This phenomenon will be discussed in more detail in section 4.3 at even lower pressure.

The solid orange line in figure 18 is the sheath width as a function of time. It is the result of the following algorithm using data from current and electric field measurements.

The values of the current shown in figure 12 do not correspond to the actual current in the discharge, but are proportional to it. Consequently, the following relation between measured current density $j_{\text{meas}}$ and actual current density $j$ is valid:

$$j = C \cdot j_{\text{meas}},$$

where $C$ is a constant. The surface charge density at the electrode surface $\sigma$ at a certain time $\tau_m$ within the RF period can be calculated out of the current density:

$$\sigma_{\tau_m} - \sigma_0 = \int_0^{\tau_m} j \, dt = C \cdot \int_0^{\tau_m} j_{\text{meas}} \, dt,$$

where $\sigma_0$ is the surface charge density at the beginning of the RF period at $\tau = 0$. The surface charge density at different phases is known from figure 15. Using equation (13), the constant $C$ is determined to be $C = 2.1 \times 10^4$ and the actual current in the discharge is known.

The sheath velocity $w(t)$ in dependence on time is given by

$$w(t) = \frac{j(t)}{en_s(t)},$$

where $n_s(t)$ is the ion density at the sheath edge, which is equal to the electron density at this point due to quasi-neutrality. $j(t)$ is known from equation (12) and $n_s(t)$ can be determined out of current and electric field measurements.

The surface charge density $\sigma$ can be determined out of the integration of the current density $j$ over time (equation (13)). However, $\sigma_0$ is unknown. Due to electron inertia the sheath collapse at low pressures does not take place at $\tau = 0$, when the applied voltage is zero, but is slightly phase shifted and takes place later. Therefore, $\sigma_0$ is not zero, but has some unknown value. Under these conditions the electrode is assumed not to charge up positively at any phase within the RF period (no field reversal during sheath collapse [11,59]). Therefore, a constant
is subtracted, so that \( \sigma \leq 0 \) during the entire RF period. This constant corresponds to \( \sigma_0 \). In fact, \( \sigma \) is always negative during the entire RF period, since the sheath never collapses completely due to the dynamic floating potential \( \Phi_f \) [38]:

\[
\Phi_f = -\frac{k_B T_e}{2e} \left[ -\ln \left( \frac{M}{2\pi k_B T_e} \right) - \ln \left( 2\pi e U / k_B T_e \right) \right].
\] (15)

For an electron temperature of 2.3 eV, a krypton mass of \( M = 1.39 \times 10^{-25} \) kg and an amplitude of the applied voltage \( U \approx 260 \) V the floating potential is \( \Phi_f \approx 4.1 \) V. This is small in comparison with the applied voltage and, therefore, neglected here. Equation (15) is based on the assumption of sinusoidal sheath voltages [38]. Due to the PSR effect the sheath voltages are not sinusoidal at 1 Pa in an asymmetric discharge. Therefore, equation (15) only yields an approximate result for the floating potential.

\( \sigma \) can also be determined out of the integration of the charge density in the sheath over the sheath:

\[
\sigma = \int_0^{\Phi_f} n_s \, dz.
\] (16)

4.2.5. Simple analytical model. The way that fast electron beams can change the isotropic part of the distribution function in the plasma bulk can be demonstrated using a simple analytical model. The distribution function \( f \) in the plasma bulk is assumed to consist of a time independent and isotropic Gaussian part \( f_0 \), an anisotropic part \( f_1 \) attributed to the ohmic current and a beam part \( f_{\text{beam}} \) resulting from the fast extension of the sheath just after the collapse:

\[
f = f_0 + f_1 \cos \theta + f_{\text{beam}}.
\] (17)

\[
\cos \theta = \frac{v_z}{v}.
\] (18)

The beam part is assumed to be a shifted version of the time independent distribution function:

\[
f_{\text{beam}}(\vec{v}) = \alpha f_0 \left[ (\vec{v} - \vec{u})^2 \right] \Theta(\vec{v} \cdot \vec{u}).
\] (19)

Here \( s(t) \) is the sheath edge position in dependence on time. The surface charge density at the electrode is assumed to be equivalent to the overall charge in the sheath. The left graph in figure 19 shows \( \sigma \) as a result of equation (13) and the right graph as result of equation (16).

The following procedure is now applied in order to determine \( n_s(t) \) and, consequently, \( u(t) \).

At a certain phase \( \sigma \) is determined from the left graph in figure 19. Then, this value is used in order to determine the sheath width \( s \) at this phase from the right graph in figure 19. Finally, this sheath width is used to determine \( n_s \) at this phase from figure 17. Knowing \( n_s(t) \) the sheath velocity \( u(t) \) is calculated using equation (14). One RF period is now separated into discrete time steps and a linear interpolation between the sheath velocities at different phases is performed. Based on this interpolation \( s(t) \) is determined. The results are shown in figures 18 and 20 (orange line). The PSR oscillations of the current are reflected in sheath voltage and width. Following the Hard Wall model [1,2], the propagation velocity of the electron beam should be twice the sheath velocity during the time interval of sheath expansion. The value of \( V_b \approx 2 \times 10^6 \) m/s, determined from the tilt of the beam trajectory in figure 18, is a time averaged value. The corresponding value for the drift velocity resulting from this algorithm (figure 20) is also of the order of \( 2 \times 10^6 \) m/s.

Figure 21 shows the result of a hybrid Monte Carlo model in terms of the integration of the calculated EEDF above 12 eV in a symmetric discharge in argon operated at 2.7 Pa [50]. Again only the fraction of the discharge close to the bottom electrode is shown. Similar to the experiment trajectories of beams of highly energetic electrons are observed, which are generated by the expanding sheath also at lower pressures.
The beam contribution makes the above distribution $f$ non-isotropic. In order to determine the isotropic part $g_{\text{beam}}$ of the beam part, the distribution function has to be averaged over the full solid angle. Taking $u$ in the $z$-direction and substituting $y = \cos \theta$, where $\theta$ is the angle from the $z$-axis, leads to

$$g_{\text{beam}}(v) = \alpha \int_0^1 f_0(v^2 + u^2 - 2vu y) \, dy.$$  \hspace{1cm} (20)

Here all velocities are normalized to the thermal velocity $\sqrt{2kT_e/m}$. Further on $f_0$ is assumed to be a Maxwell–Boltzmann distribution $f_0 = N \cdot e^{-v^2}$, where $N$ is a normalization factor. Then the above integral becomes

$$g_{\text{beam}}(v) = \alpha f_0(v^2) e^{-u^2} \frac{e^{2vu} - 1}{2vu}.$$  \hspace{1cm} (21)

The distribution function measured by Langmuir probes does not resolve the fast temporal oscillation of $u$, but performs a time average over the RF period $T$. According to our measurements, $w$ and correspondingly also $u$ are high only during the short interval of fast expansion following the collapse. This can be described by the PSR effect [38], but functional details are complicated. As a reasonable analytical approximation $u$ is assumed to be a constant $u_0$ for $0 \leq t \leq \tau$ and zero otherwise. In this approximation, the time averaged
isotropic part of the distribution function in the bulk \( g_{\text{bulk}} = \langle f \rangle \) finally has the following form

\[
g = f_0(v^2)(1 + \alpha h(v)), \quad (22)
\]

\[
h(v) = 1 + \frac{\tau}{T} \left( e^{-\frac{v}{v_0}} - 1 - \frac{1}{2v_0^2} - 1 \right). \quad (23)
\]

Typical values for the parameters can be extracted from different measurements presented earlier in this paper. The time averaged density \( n_s \) in the sheath can be estimated in two different ways. It can be determined out of the measured electric field at the electrode at a given phase assuming a constant \( n_s \) and using equation (5). Alternatively, \( n_s \) is given by the average value of the ion density in the sheath, that was calculated in section 4.2.3 (figure 17). Both ways yield \( n_s \approx 4 \times 10^8 \text{ cm}^{-3} \). The density in the bulk is \( n_0 \approx 2 \times 10^9 \text{ cm}^{-3} \) (section 4.2.2). Therefore, \( \alpha = 0.2 \) is assumed. \( \tau/T \) can be determined out of the excitation (figure 18) or current measurements (figure 12). In both cases an electron beam is observed during 15% of the RF period. Therefore, \( \tau/T = 0.15 \) is assumed. The average sheath expansion velocity \( w \) can be determined out of the excitation (figure 18) or figure 20 to be \( 1.1 \times 10^6 \text{ m s}^{-1} \). The electron temperature of the low energetic part of the EEDF, which determines the thermal velocity, is \( kT_e = 2.3 \text{ eV} \) (section 4.2.2). Normalizing \( u = 2 \cdot w \) by the resulting thermal velocity yields \( u_0 \approx 2.5 \).

The resulting form of \( \langle f \rangle \) depends critically on the velocity \( u_0 \). For \( u_0 < 1 \), the beam has little effect and the distribution function is close to \( f_0 \). However, for \( u_0 > 1 \) a strong tail develops at high energies. This is clearly shown in figure 22 where the distribution function is plotted as a function of energy normalized to \( kT_e \). In the figure, the normalization factor \( N \) is set to \( N = 1/[1 + \alpha(1 - \tau/T)] \). The form of the distribution function at different times is shown in figure 22 for the case of \( u_0 = 2.5 \). This bi-Maxwellian agrees well with the measured EEDF at low pressures (see figure 7).

This model clearly shows that stochastic heating is closely related to highly energetic electron beams. The formation of these beams is influenced by the PSR effect, which causes the sheath to expand faster. Therefore, stochastic heating is also correlated with the PSR effect.

4.3. Results at 0.5 Pa

Current and PROES measurements have been performed at 0.5 Pa and 8 W in pure krypton. If the pressure is even lower than 1 Pa, the excitation related to the PSR oscillations of the RF current waveform is clearly pronounced. Figure 23 shows the correlation between current, excitation and deposited power in terms of \( I^2 \) at 0.5 Pa and 8 W in a pure krypton discharge. At the beginning of one RF period one can observe a direct correlation between the three graphs indicated by vertical lines. At the phases of maximum excitation, extrema of the current and maxima of the deposited power are observed. The first extremum of the current corresponds to the fast initial expansion of the sheath. Consequently, the generation of an electron beam is observed at this phase. When these beam electrons hit the opposing quartz cylinder, it is charged negatively and, consequently, a sheath develops in front of it. In contrast to the tilted excitation maximum caused by the electron beam the following maxima are straight. Their formation might be understood based on the following hypothesis.

At the phases of straight excitation maxima there are sheaths at the quartz and at the powered electrode. Consequently, electrons in the plasma bulk are confined within a potential well. At relatively low energies this potential can be assumed to be harmonic. In such a harmonic potential well, the oscillation period of the confined low energetic electrons does not depend on the spatial oscillation amplitude. Therefore, the observed excitation maxima are straight and the maximum excitation is observed in the discharge centre, where the absolute value of the potential is minimum and the kinetic energy is maximum. The correlation between excitation maxima and current extrema might show that additional energy is transferred to the oscillating electrons via the PSR oscillations of the sheath edge at the bottom powered electrode. Once these PSR oscillations stop, the
amplitude of the excitation maxima decreases because the energy loss via collisions is no longer compensated. This scenario might be similar to electron bounce resonance heating described in [60] with the difference that here the bounce resonance frequency is not the RF but the PSR frequency. However, this argumentation remains a hypothesis, which needs to be tested and verified against experiments and/or simulations. Similar measurements have been performed in hydrogen at 1.2 Pa and 60 W before [61]. These results are in good agreement with PIC simulations of the power density performed by Vender [62].

It should be noted that in this model the electron oscillation leading to the observed excitation pattern is perpendicular to the quartz surface. The current amplitude is measured outside the gap between electrode and quartz. Therefore, the measured current corresponds to electron oscillations parallel to the quartz and electrode surface. Although both directions are coupled, they do not necessarily have to be identical.

5. Conclusion

Cause and effect of electron (stochastic) heating at low pressures in an asymmetric CCRF discharges have been studied using the synergistic effect of various diagnostics, analytical model and simulations. FDS in krypton was applied for the first time in order to measure electric fields in the sheath at high spatial and temporal resolution. Generally, FDS in krypton can also be applied to other gas mixtures using only a small admixture of krypton.

PROES was used to measure the excitation into a specifically chosen krypton level spatially and temporally resolved within the RF period with one dimensional spatial resolution along the discharge axis. The combination of these two diagnostics allowed experimental investigations of cause and effect of electron stochastic heating in asymmetric CCRF discharges.

Simultaneously to each measurement the current in the plasma and the RF voltage were measured. Separately, the discharge is characterized by Langmuir probe measurements in terms of electron density, mean electron energy and EEDF. The measured EEDFs at 1 Pa and 10 Pa are bi-Maxwellian showing an enhanced high energy tail, which can be attributed to stochastic heating, that is efficient at such low pressures. The measured electric fields at 1 Pa and 10 Pa agree well with the theoretically calculated values using a fluid sheath model. Both experiment and simulation, show the fast sheath expansion and the sheath collapse. During the sheath expansion the generation of beams of highly energetic electrons by the expanding sheath is observed by PROES.

The measured spatio-temporal excitation is compared with results of a hybrid Monte Carlo model showing good qualitative agreement. Due to an absolute phase calibration of FDS and PROES the effect of electron stochastic heating (excitation) can be directly linked to its cause (spatio-temporal evolution of the electric field in the sheath). A simple analytical model demonstrates that these highly energetic electron beams lead to an enhanced high energy tail of the EEDF, which is usually attributed to stochastic heating. The input parameters for this model were extracted from measurements using different diagnostics.

At low pressure (1 Pa) the PSR effect is observed in terms of high frequency oscillations of the measured RF current waveform. It also affects the measured spatio-temporal excitation profiles. The measured currents at high (45 Pa), intermediate (10 Pa) and low (1 Pa) pressures are well reproduced by an analytic PSR model. The assumption, made in this model, that the sheath voltage is proportional to the square of the surface charge density at the electrode is verified experimentally.
Based on the time resolved electric field measurements the spatially resolved ion density in the sheath is calculated. Based on current and field measurements sheath velocity and width are calculated showing good agreement with data taken from PROES measurements.

Altogether, the synergistic use of different diagnostic techniques, analytical models and simulations provides a better understanding of electron (stochastic) heating in asymmetric CCRF discharges at low pressures. We have demonstrated, both experimentally and theoretically, that the nature of stochastic heating is closely related to electron beams that are generated during the sheath expansion phase. The PSR effect leads to sheath oscillations, whose frequency is about one order of magnitude higher than the RF of the applied voltage. As a consequence, a much faster initial sheath expansion takes place, which enhances the generation of energetic electron beams. Further, the PSR effect leads to high frequency current oscillations, which contribute to the ohmic heating.

Regarding industrial applications, it is important to point out that the regime investigated here is typical for industrial processing applications (low pressure, asymmetric discharge). The PSR and its effect on the parameters investigated here is, therefore, most relevant.

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