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Diagnostics of the plasma series resonance effect in radio-frequency discharges

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Abstract. The intention of the paper is to give an example on how different plasma diagnostics can be combined in a synergistic way in order to investigate new physics. The link between the individual diagnostics has to be provided by theoretical concepts that predict certain relations between the different plasma parameters. The example chosen here is the effect of self-excited plasma series resonances in asymmetric capacitively coupled RF discharges. These resonance oscillations lead to high frequency current oscillations and are caused by a series resonance between the capacitive sheath and the effective inductance of the bulk which results from electron inertia. The non-linearity of the sheath is essential for the self-excitation of these oscillations. Laser spectroscopic electric field measurements, phase and space resolved optical emission spectroscopy, current, voltage, and Langmuir probe measurements are combined. The synergistic effect of these diagnostics in combination with a simple analytical model for the modification of the electron energy distribution function by electron beams yields information on cause and effect of electron heating and a better understanding of these fundamental phenomena.

1. Introduction
Capacitively coupled radio frequency (CCRF) discharges have been applied for processing applications for a long time and a long record of experimental and theoretical investigations exists. A standard assumption in most of the work is a sinusoidal current waveform. This is well justified at relatively high pressures or generally in symmetric discharges. However, in asymmetric discharges at low pressures of less than typically 10 Pa higher harmonics in the 100 MHz regime can arise and become even dominant. In this paper it will be shown how the phenomenon is investigated by combining laser spectroscopic electric field measurements for measuring the sheath electric field, a so called "SEERS" current sensor, phase and space resolved optical emission spectroscopy for determination of the local excitation rate, Langmuir probe measurements for the determination of electron energy distribution functions (EEDF) and a high voltage probe for determination of the RF voltage.

In general, despite its technological importance, the complexity of power coupling mechanisms in CCRF discharges is not yet fully understood. In particular, there is a lack of experimental investigations with regard to heating and excitation phenomena in these discharges at low pressures.

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 heating is the
electric field in the plasma boundary sheath and its temporal as well as spatial evolution within one RF cycle [1]. Its effect is the time and space dependent ionisation in the plasma, which can be probed through excitation [2, 3, 4]. Insight into power dissipation requires temporal resolution on a nanosecond time scale within the RF cycle 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. Using the resulting Stark maps the electric fields within the sheath are investigated by fluorescence dip spectroscopy (FDS). Here the technique is applied to krypton as a probe gas for the first time [5, 6]. Similar methods for electric field measurements have already been developed and applied to other molecular and atomic probe gases like BCl [7], NaK [8], CS [9], He [10, 11, 12], H [13, 14], argon [6, 15, 16, 17, 18, 19] and Xe [20]. Knowledge of the field allows the determination of e.g. voltages, charge densities and currents.

The excitation dynamics (effect of electron heating) is investigated by phase resolved optical emission spectroscopy (PROES). Similar investigations of excitation dynamics have been performed before [21, 22, 23, 2, 3, 4, 24]. Both diagnostics are non-intrusive and provide high temporal and spatial resolution on a nanosecond and sub-millimeter scale.

The plasma itself is characterised by radially resolved Langmuir probe measurements of electron density, electron temperature and electron energy distribution function. The EEDF is a key parameter for understanding basic discharge dynamics. It determines dissociation, excitation and ionisation processes. Temporal changes of the high-energy tail of the EEDF in single frequency RF discharges within the RF cycle have been previously observed by time resolved measurements [21, 22, 23, 2, 3, 4, 24]. Phase resolved optical emission spectroscopy (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 [25, 26].

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 plasma series resonances (PSR) are discussed based on the information from probe, current and voltage measurements. The corresponding experimental results are compared to an analytical PSR model [26] showing good agreement. The synergistic effect of all these diagnostics can yield a more complete picture of electron heating in CCRF discharges.

The paper is structured in the following way: In the second paragraph the experimental setup for all diagnostics is described separately. At the beginning the setup used for the electric field measurements and the investigation of the Stark effect in krypton is presented. Then the equipment used for PROES, current-, voltage- and probe measurements is described. In the third paragraph the PSR effect and the analytical model to describe this effect are introduced. The fourth paragraph comprises a description of the basics of FDS in krypton and PROES. A database of Stark shifts of high Rydberg states in dependence on the electric field is presented and the algorithm used for calculating the excitation out of the measured emission is introduced. In the fifth paragraph the results of this work are presented. Here the focus lies on the comparison between two different sets of parameters (10 Pa and 1 Pa), for which all diagnostics were applied. The results of the different diagnostics are compared to each other yielding a better understanding of electron heating under these conditions. Finally conclusions are drawn.

2. Experimental setup
2.1. Setup for 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 is described in detail elsewhere [5]. For these investigations the $^1S_0$ to $5p^2[3/2]_2^2$ transition was pumped at $2 \times 204.13 \text{ nm}$ (two-photon excitation). The fluorescence from the $5p^2[3/2]_2$ state to the $5s^2[1/2]_0^1$ state at $826 \text{ nm}$ was monitored. A second tunable dye laser was used to excite from the $5p^2[3/2]_2$ state to high Rydberg states, which were shifted due to the Stark effect. Scanning the wavelength yields the dip spectrum. Different Rydberg levels with different field sensitivity were used depending on the electric field strength in the sheath at a given spatial position and phase.

2.1.1. Laser system, GEC reference cell and calibration cell

The experimental setup is shown in figure 1. For the investigation of the Stark splitting of high Rydberg states of krypton at known electric fields the calibration cell shown in figure 1 was used. For measurements of the electric field in the plasma sheath the calibration cell was replaced by a GEC cell (see figure 2).

![Figure 1](image)

Figure 1. Experimental setup used for the investigation of the Stark splitting of high Rydberg states of krypton at known electric fields. For measurements of the electric field in the plasma sheath the calibration cell was replaced by a GEC cell.

The frequency-doubled output beam (532 nm) of a seeded Nd:YAG laser (Continuum, 9020) with a pulse width of 7 ns and 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 $H_2$ 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 [28].

The metal cylinder surrounding the ICP antenna and the dielectric window were replaced by a monolithic quartz housing (figure 2). Here the inductive coupling was not used. The RF voltage is applied only to the lower electrode and the whole chamber wall acts as grounded electrode. Therefore, the discharge is strongly asymmetric and almost the entire voltage drops across the sheath at the powered electrode.

The investigation of the Stark splitting of high Rydberg states of krypton in dependence of a known electric field was done in a calibration cell and a photomultiplier was used for the detection of the fluorescence light.
The calibration cell is a vacuum chamber filled with krypton, where measurements with known electric field strength are performed. It contains a parallel-plate capacitor-like structure (d = 9.8 mm) providing a homogeneous DC electric field up to 3 kV/cm without electrical breakdown. The laser beams can pass the cell at the center of the capacitor through quartz windows on opposite sides. The fluorescence light can leave the chamber perpendicular to the beams through a standard BK7-window and is detected by a red-sensitive photomultiplier (Hamamatsu, R943-02). An interference filter blocks scattered light from the visible laser beam.

2.1.2. Synchronization for phase resolved measurements

The Q-switch of the Nd:YAG pump laser was synchronized with the RF generator (f = 13.56 MHz) using a frequency divider (20 Hz) and a delay generator (Stanford, DG-535). Moreover, the gate of an ICCD camera (Princeton Instruments) was synchronized with the laser pulse (figure 2). The variable delay between Q-Switch and RF generator allowed phase resolved measurements within the RF cycle. The spatial resolution of this setup is 50 $\mu$m.

![Diagram of the calibration cell and measurement setup](image)

**Figure 2.** GEC reference cell accessible for FDS and PROES

A wavelength scan of the second dye laser over a certain wavelength interval with a stepwidth of 3 pm was performed in 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 [5]. 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, that blocked all wavelengths below 780 nm. Then a difference image was calculated in order to determine the fluorescence decrease. Pixels in horizontal direction were binned in order to reduce the noise. On the one hand spatial resolution was achieved by the fact, that the laser beam itself covered 2 mm of the sheath and on the other hand by moving the entire discharge chamber up and down leaving the laser beams at their original position. Using this technique the entire sheath ($s_{max} \approx 1$ 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 to a database of Stark shifts of Rydberg states in krypton [5] and the electric fields are determined spatially and temporally resolved.
2.2. Setup for phase resolved optical emission spectroscopy

![Figure 3](image.png)

**Figure 3.** Principle of phase resolved measurements

The setup used for PROES is identical with the one shown in figure 2, but the laser system was not used and only the emission from the plasma was detected by an ICCD camera (Andor iStar). The camera was synchronized with the RF-generator via a frequency divider (30 KHz). The $Kr2p_5$-state at 758.7 nm was observed using an interference filter. The lifetime of 21.5 ns is short enough to resolve electron dynamics within one RF cycle at 13.56 MHz ($T_{RF} = 74$ ns). Figure 3 shows the principle of phase resolved measurements. The internal delay generator of the ICCD camera sets a certain delay between trigger and camera gate. Here the minimum camera gate width of 4.2 ns was used. Signal is acquired at a certain phase during several thousands of RF cycles. 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 cycle. Typical exposure times were 10 s. All images were binned in horizontal direction in order to reduce the noise resulting in one dimensional spatial resolution along the discharge axis.

2.3. Probe-, voltage- and current measurements

![Figure 4](image.png)

**Figure 4.** Realization of current measurements using a SEERS current sensor

In order to characterize the plasma in terms of electron density, electron mean energy and EEDF under each conditions 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 to every 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). Also simultaneously the current to the chamber wall was measured by a SEERS sensor (ASI), that was integrated into the side wall of the GEC cell (see figure 4).

3. The Plasma Series Resonance

In this paper the focus lies on the synergistic use of different diagnostic techniques in order to study electron heating in CCRF discharges at low pressures. In that respect the PSR plays an important role. In asymmetric CCRF discharges (see figure 4) 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.

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 can be detected by a small sensor integrated in the reactor wall, that picks up a fraction of the RF current (see figure 4). This diagnostic can be used as a non-invasive process monitoring technique in industry. It is known as Self Excited Electron Resonance Spectroscopy (SEERS) [29, 30].

![Figure 5. Current measured by a SEERS sensor in a strongly asymmetric CCRF discharge in krypton at 0.5 Pa and 8 W](image)

Figure 5 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 cycle can be observed. These oscillations can be described by a one dimensional analytical PSR model [26]. In this model the so-called PSR equation is solved for current and charge. 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 $\sigma^2$. 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.

The corresponding equation for an externally applied potential $\Phi$ is [26]:

$$\Phi = -\frac{1}{2e\varepsilon_0}\pi_s A_s^2 Q^2 + \frac{mL}{e^2\pi A} \left( \frac{\partial^2 Q}{\partial t^2} + \nu \frac{\partial Q}{\partial t} \right)$$

(1)

Here $\pi_s$ is an effective quasi-constant ion density in the sheath (matrix sheath approximation), $A_s$ the surface area of the powered electrode, $Q$ the charge, $L$ an effective discharge length, $\pi$ and $A$ effective values for the discharge density and area and $\nu$ is the electron-neutral elastic collision frequency, that is assumed to be independent of the drift velocity. The first term in equation 1 corresponds to the voltage drop across the sheath and the second term to the voltage drop across the plasma bulk.

By introducing a sinusoidal voltage

$$\Phi = -\frac{\Phi_0}{2} [1 - \cos(\omega_{RF} t)]$$

(2)

and dimensionless quantities equation 1 reduces to its final form, which is called PSR equation:

$$\sin^2 \left( \frac{\Omega \tau}{2} \right) = q^2 - 2(\ddot{q} + \kappa\dot{q})$$

(3)

Here $q = \frac{Q}{Q_m}$ is the charge normalized to the space charge $Q_m$ in the sheath at the phase of maximum negative applied voltage $-\Phi_0$. $\tau = \omega_0 t$ where $\omega_0 = \gamma \omega_p$ is the plasma frequency reduced by a dimensionless geometry factor $\gamma = \sqrt{s_m A_s} < 1$ with $s_m = \sqrt{2e\varepsilon_0 \phi_0 m}$. $\Omega = \frac{\omega_{RF}}{\omega_0}$ is the normalized RF frequency and $\kappa = \frac{\omega}{\omega_0}$ the normalized electron-neutral elastic collision frequency.

In this paper this model will be used to describe measured currents theoretically by varying the two parameters $\Omega$ and $\kappa$ in equation 3.

4. Basics for diagnostics

4.1. Electric field measurements

In order to measure electric fields in the sheath of a CCRF discharge a novel diagnostic of FDS in krypton was developed. In comparison to other probe gases krypton has various advantages: As a rare gas krypton does not influence the discharge conditions much due to its high ionization energy. Compared to molecular gases such as $H_2$ the influence on the EEDF is low, since krypton does not have any low energetic vibrational or rotational levels. Furthermore, the degree of dissociation of molecular gases in low-temperature CCRF discharges is low. Therefore, generally much higher partial pressures of molecular gases are needed compared to atomic gases. In comparison to other noble gases like He or Ar the first excitation step in krypton starts from the ground state and not from a metastable level [10, 17]. Therefore, in case of krypton, a calibration without plasma, simply in the neutral gas, is possible.

In order to produce a database of Stark shifts of high Rydberg states of krypton, the line splitting of these levels was investigated in a calibration cell with known electric fields.

First of all the strong $nd^3/2\ell_3$ autoionizing Rydberg series in the zero field case has been measured for principal quantum numbers $15 \geq n \geq 50$. The corresponding spectrum is shown in figure 6. The fluorescence decrease is around 60% at $n = 15$, thus the transitions at lower $n$ are almost saturated without electric field [5].
Figure 6. Spectrum of the Rydberg series in krypton without electric field. The relative fluorescence decrease $\delta F$ is plotted versus the principal quantum number $n$ of the $nd'[5/2]_3$ levels.

Figure 7. Measured (left graph) and calculated (right graph) Stark map in krypton in the range $19d' - 21d'$. In case of the experimental results the fluorescence decrease is indicated by bright lines (brighter = higher $\delta F$). The straight line at $\lambda \approx 630.8 \text{ nm}$ is an artefact.

The most prominent features of the Stark effect in krypton can be discussed on the basis of a two-dimensional map of the fluorescence decrease (see figure 7). It consists of eleven spectra which were obtained at field strengths between zero and about $1000 \frac{V}{cm}$. The wavelength range covers the interval between the $19d'$ and $21d'$ levels in the zero field case. For comparison, a map of the calculated line positions is shown in figure 7. In order to keep the figure clear, the field dependent line strengths are not shown. At the left edge of figure 7, one can only recognize the strong $nd'$ and the much weaker $ns'$ levels. With increasing field strength, they are slightly shifted to longer wavelengths and disappear. Especially the $nd'$ levels disappear very rapidly at around $500 \frac{V}{cm}$. On the other hand, the forbidden $np'$ levels become visible at around $200 \frac{V}{cm}$, shifting with increasing field strength very strongly and getting brighter at the same time. A manifold of the higher, quasi-degenerated states with $l \geq 3$ appears above $500 \frac{V}{cm}$ between the $nd'$ and $(n + 2)p'$ states. The agreement between theoretically and experimentally obtained Stark maps is very good.
Figure 8. Quadratic Stark shift of \( np' \) levels. The energy differences \( \Delta \varepsilon \) between the observed shifted lines and the associated, extrapolated zero field energies have been plotted versus the square of the electric field \( E \).

As one example figure 8 shows the quadratic Stark shift of four different \( np' \) levels. These states only get visible, if an electric field is applied, since they get admixtures of the \( ns' \) and \( nd' \) states due to the Stark effect.

4.2. Excitation dynamics

For phase resolved investigations of the plasma emission the \( Kr2p_5 \)-state was chosen, since the discharge is operated in krypton and the contribution of cascades to the population density of this state is small [32]. Moreover, \( Kr2p_5 \) has a short lifetime of only 21.5 ns [33], which allows access to excitation dynamics within one RF cycle, since the lifetime is shorter than the length of one cycle (74 ns).

Therefore, the population density \( n_i(t) \) of the \( Kr2p_5 \)-state can be described by the following rate equation:

\[
\frac{dn_i(t)}{dt} = n_0 E_{0,i}(t) - A_i n_i(t)
\] (4)

In equation 4 \( E_{0,i}(t) \) is the electron impact excitation function for excitation out of the ground state. \( n_0 \) is the ground state density and \( A_i = \frac{1}{\tau} \) the decay rate, that is given by the inverse lifetime \( \tau \). Quenching is neglected because of the low pressure and the level is chosen such that excitation out of metastable levels is low and, therefore, also neglected.

The measured number of photons per unit volume and time \( n_{ph,i}(t) \) is given by:

\[
n_{ph,i}(t) = A_{ik} n_i(t)
\] (5)

Here \( A_{ik} \) is the transition probability of the observed emission.

Substituting \( n_i(t) \) using equations 5 and 4 the excitation into the \( Kr2p_5 \)-state can be directly determined out of the measured emission by the following equation:

\[
E_i(t) = \frac{1}{A_{ik} n_0} \left( \frac{dn_{ph,i}(t)}{dt} + A_i n_{ph,i}(t) \right)
\] (6)

In fact cascade contributions cannot be completely neglected in case of \( Kr2p_5 \) [2]. However, the error caused by the neglection of cascades is small and acceptable for the qualitative inves-
tigations performed here [32].

5. Results

The focus of this work lies on the investigation of the PSR effect and generally on electron heating at low pressures. For these investigations various plasma parameters are investigated in detail applying different diagnostics.

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 is the cause of electron heating. Therefore, a novel technique of FDS in krypton for space and phase resolved electric field measurements was developed and applied to a CCRF discharge for the first time.

In order to investigate the Stark Effect of high Rydberg states of krypton systematically in dependence of the electric field, measurements of the Stark Splitting of these levels were performed in a calibration cell with known electric field. (see chapter 4.1). Then this knowledge was used to perform spatially and temporally resolved measurements of electric fields, excitation dynamics, voltage, current and other plasma parameters such as electron density, electron mean energy and EEDF in a strongly asymmetric CCRF discharge under various conditions.

![Figure 9](image_url)

**Figure 9.** Comparison between measured (blue line) and modeled (black line) current in the plasma at 45 Pa and 8 W. The measured RF voltage (red line) is also shown.

In this paper 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 it starts to be observable and at 1 Pa it is clearly pronounced. At higher pressures no high frequency oscillations of the RF current are observed. Figure 9 shows a plot of the measured current to the plasma wall at 45 Pa and 8 W in a pure krypton discharge. Although the current is not sinusoidal and no high frequency oscillations exist, it is well reproduced by the analytical PSR model using the parameters $\Omega$ and $\kappa$ shown in the figure (see chapter 3).

5.1. Results at 10 Pa

5.1.1. Current- and voltage measurements
At low pressures the current starts to deviate from the form shown in figure 9. Figure 10 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.

![Figure 10](image_url)

**Figure 10.** Comparison between measured (blue line) and modeled (black line) current in the plasma at 10 Pa and 8 W. The measured RF voltage (red line) is also shown.

The figure also shows a comparison to an analytical PSR model [26] presented in chapter 3. The agreement between theory and experiment is good. At lower pressure the PSR oscillations are much more pronounced (see chapter 5.2.1).

### 5.1.2. Langmuir probe measurements

Figures 11 and 11 show 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 \( n_e \approx 7 \cdot 10^9 \text{ cm}^{-3} \) and the electron mean energy is of the order of \( <\varepsilon> \approx 3 \text{ eV} \). Both profiles show shapes typical for CCRF discharges.

Figure 12 shows the EEDF, in the radial centre of the discharge. It has a Bi-Maxwellian shape typical for CCRF discharges at low pressures [31]. The enhancement of the high energy tail can be interpreted as influence of stochastic heating, that 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 \( T_c \) 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 center those temperatures are \( T_c \approx 1 \text{ eV} \) and \( T_h \approx 5 \text{ eV} \).
Figure 11. Radially resolved measurement of the electron density (left graph) and electron mean energy (right graph) in a pure Kr discharge (10 Pa, 8W) 2.5 cm above the electrode.

Figure 12. EEDF in the radial centre of a pure Kr discharge (10 Pa, 8W) 2.5 cm above the electrode.

5.1.3. Electric field measurements

Figure 13 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. 4 different phases were investigated. The electric fields in the sheath shortly after the collapse (5 ns), at the phase $\Phi = \frac{\pi}{2}$ (19 ns), $\Phi = \pi$ (37 ns, maximum sheath expansion) and $\Phi = \frac{3\pi}{2}$ (55 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 5 ns and 19 ns. Furthermore, there is an asymmetry between $\Phi = \frac{\pi}{2}$ (sheath expansion, 19 ns) and $\Phi = \frac{3\pi}{2}$ (sheath collapse, 55 ns) in terms of the electric field distribution. This asymmetry might be explained by the amplitude of the current at different phases, that is shown in figure 10. As a very rough estimate the ion density in the sheath is assumed to be constant (matrix sheath model). Then
Figure 13. Spatial and temporal evolution of the electric field in the sheath of an asymmetric CCRF discharge at 10 Pa and 8 W in krypton

\[ s \propto Q \]  \hspace{1cm} (7)

where \( s \) is the sheath width and \( Q \) is the charge in the sheath. Consequently

\[ \dot{s} = v_s \propto \dot{Q} = I \]  \hspace{1cm} (8)

where \( v_s \) is the sheath velocity and \( I \) the current. The amplitude of the current is much higher at the beginning of the RF cycle than at the end. Therefore, following equation 8 the sheath velocity is much higher at the beginning of each RF cycle compared to its end. Consequently, there is an asymmetry between sheath expansion and collapse, that is also observed in the electric field measurement in terms of different sheath expansions at \( \Phi = \frac{\pi}{2} \) and \( \Phi = \frac{3\pi}{2} \). Furthermore, the FDS measurements are time averaged over one laser pulse (5 ns). This naturally leads to a slight asymmetry at these phases.

Following Poisson’s equation:

\[ \frac{dE}{dx} = -\frac{e}{\varepsilon_0} (n_i - n_e) \]  \hspace{1cm} (9)

a steeper decrease of the electric field in the sheath \( (n_e = 0 \text{ cm}^{-3}) \) corresponds to a higher 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 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 13.

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. Following

\[ E = -\nabla \Phi \]  \hspace{1cm} (10)

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 14 and very good agreement is found.

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

\[
\frac{dE}{dx} = \frac{e}{\varepsilon_0} n_i \tag{11}
\]

where \(e\) is the elementary charge, \(\varepsilon_0\) the dielectric constant and \(n_i\) the ion-density. Integration of equation 11 from 0 to the momentary sheath edge \(s(t)\) yields:

\[
E_{el} = \frac{e}{\varepsilon_0} \int_0^{s(t)} n_i \, dx \tag{12}
\]

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

\[
\sigma = e \int_0^{s(t)} n_i \, dx \tag{13}
\]

Substitution of equation 13 into equation 12 yields a direct relation between the electric field at the electrode and the surface charge density:

\[
\sigma = \varepsilon_0 E_{el} \tag{14}
\]

Using equation 14 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 14. Obviously

\[
U_s \propto \sigma^2 \tag{15}
\]

This relation is an important fundamental assumption for a one dimensional Plasma Series Resonance (PSR) model [26] and has now been verified experimentally.
5.1.4. Excitation dynamics

Figure 15 shows the spatio-temporal excitation into $Kr_2p_5$ close to the powered electrode under the same conditions as before. The excitation was calculated out of the measured temporally and spatially resolved emission using equation 6. An absolute phase calibration between the FDS and PROES measurements was performed, so that the time scales in figures 13 and 15 are identical. In figure 15 two RF cycles are shown and the sheath width can be estimated to be 5 mm corresponding to the position where the excitation starts to increase. This agrees very well with the maximum sheath width $s_{10Pa} \approx 5 \text{ mm}$ determined from the electric field measurements.

![Figure 15. Spatio-temporal plot of the excitation into $Kr_2p_5$ close to the powered electrode in an asymmetric CCRF discharge at 10 Pa and 8 W in krypton. The line indicates the trajectory of a beam of high energetic electrons, that is accelerated by the expanding sheath.](image)

During the phase of fastest sheath expansion (5 ns - 19 ns, see figure 13) the most intense excitation is observed. At this phase a beam of high energetic electrons is generated by the expanding sheath, that penetrates into the plasma bulk (straight line in figure 15). During the time interval from 5 ns to 19 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 high energetic directed beam electrons loose energy and their directed character through collisions. Similar phenomena have been observed in H [14, 2, 3] before. In case of the investigations performed in H, electron beams could not directly be 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 $v_{d,10Pa} \approx 7 \cdot 10^5 \text{ m/s}$. 

15
5.2. Results at 1 Pa

5.2.1. Current and voltage measurements

Figure 16 shows a comparison between the measured current to the plasma wall (SEERS sensor implemented into the side wall of the GEC chamber) and the modeled current using equation 3 at 1 Pa and 8 W. The measured RF voltage is also shown.

![Figure 16](image-url)

Figure 16. Comparison between measured (blue line) and modeled (black line) current in the plasma at 1 Pa and 8 W. The measured RF voltage (red line) is also shown.

At high pressures (45 Pa) the current is nearly sinusoidal as it is usually assumed and the agreement with the PSR model is good (see figure 9). At an intermediate pressure of 10 Pa small high frequency oscillations superimposed on the sinusoidal shape become observable (see figure 10). At low pressures (1 Pa) the temporal characteristics of the current are completely different compared to 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 [25, 26]. The current oscillations are well reproduced by the analytical PSR model (equation 3) under the corresponding conditions.

5.2.2. Langmuir probe measurements

Figure 17 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, that is explained in paragraph 5.2.5. The agreement is good and a Bi-Maxwellian shape is observed in both experiment and theory. At 1 Pa the electron mean energy is \( < \varepsilon > \approx 5.1 \text{ eV} \), 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 \( n_e \approx 2 \cdot 10^9 \text{ cm}^{-3} \).
Figure 17. Comparison between an experimentally obtained EEDF (points) measured close to the radial centre of a pure Kr discharge (1 Pa, 8W) 2.5 cm above the electrode and a theoretically calculated EEDF (solid line) using a simple analytical model with input parameters obtained independently from the experiment (see chapter 5.2.5)

5.2.3. Electric field measurements

Figure 18 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. Similar to the situation at 10 Pa several different phases were investigated.

Figure 18. Spatial and temporal evolution of the electric field in the sheath of an asymmetric CCRF discharge at 1 Pa and 8 W in krypton

Due to the lower pressure the maximum sheath width is bigger than in the 10 Pa case (see figure 13) and the electric field in the sheath is lower. Based on figure 18 the maximum sheath width can be estimated to be $s_1 \approx 1\, \text{cm}$. Similar to the case of higher pressure the fast sheath expansion between 5 ns and 19 ns as well as the sheath collapse are clearly observable. Furthermore, the same asymmetry as before between sheath expansion and collapse with respect
to the electric field distribution is found. This asymmetry can again be explained by the different amplitudes of the current (see figure 16) during sheath expansion and collapse.

![Graph showing electric field distribution with measurement and high V-probe data.](image1)

**Figure 19.** 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.

At 1 Pa and 8 W the voltage measured by a high voltage probe close to the electrode can again be compared to the momentary sheath voltage resulting from an integration of the measured electric fields over the sheath. This comparison is shown in figure 19 and very good agreement is found.

Furthermore, the proportionality of the sheath voltage $U_s$ to the surface charge density $\sigma$ can be investigated. The surface charge density is again calculated out of the electric field at the powered electrode using equation 14. The result is the same as before. The sheath voltage is proportional to the square of the surface charge density also at higher pressures.

Based on the time resolved measurements of the electric field shown in figure 18 the time averaged electric field in the sheath can be calculated by averaging the measured electric fields at different positions in the sheath over one RF cycle. The result is shown in figure 20. Integration of the time averaged field yields the time averaged potential $\Phi$ in the sheath at different positions (see figure 20). As the ions in the discharge are much heavier than the electrons, the ions can only react to this time averaged potential.

In the sheath energy and flux conservation apply for the ions:

$$\frac{1}{2} M v_i^2(x) + e\Phi(x) = \frac{1}{2} M v_0^2$$  \hspace{1cm} (16)

$$n_i(x) v_i(x) = n_0 v_0$$  \hspace{1cm} (17)

Here $M$ is the ion mass, $v_i$ the ion velocity in the sheath, $n_0$ 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. Substituting $v_i$ in equation 16 using equation 17 the ion density $n_i(x)$ can be derived in dependence of the time averaged potential $\Phi$ in the sheath:
Figure 20. 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

\[ n_i(x) = n_0 \left(1 - \frac{2e\Phi(x)}{Mv_0^2}\right)^{-\frac{1}{2}} \]  

(18)

Figure 21. Time averaged ion density in the sheath at 1 Pa and 8 W in a pure Kr discharge

The result in terms of the space resolved ion density in the sheath is shown in figure 21. The ion density is \(2 \cdot 10^8 \, \text{cm}^{-3}\) 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 \(n_i \approx 2 \cdot 10^9 \, \text{cm}^{-3}\) agrees very well with the electron density in the bulk \((n_e \approx 2 \cdot 10^9 \, \text{cm}^{-3})\) measured by a Langmuir probe (see chapter 5.2.2).

5.2.4. Excitation dynamics

Analog to the investigations performed at 10 Pa and presented in the previous paragraph,
the effect of electron heating in term of the spatio-temporal excitation, probed through the Kr\textsubscript{2}\textsuperscript{p}\textsubscript{5}-state, is also investigated at 1 Pa. The result is shown in figure 22.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure22.png}
\caption{Spatio-temporal plot of the excitation into Kr\textsubscript{2}\textsuperscript{p}\textsubscript{5} close to the powered electrode in an asymmetric CCRF discharge at 1 Pa and 8 W in krypton}
\end{figure}

The maximum sheath width obtained from the spatio-temporal contour plot of the excitation is \(s_{1\text{ Pa}} \approx 1\text{ cm}\). This is again in good agreement with the electric field measurements (see figure 18). Similar to the situation at a higher pressure of 10 Pa (see figure 15) the generation of a high energetic electron beam is observed. From the slope of the corresponding trajectory the propagation velocity of the beam-electrons can be estimated to be \(v_{d,1\text{ Pa}} \approx 2 \times 10^6 \frac{\text{m}}{\text{s}}\). In comparison to the observed drift velocity at higher pressures \(v_{d,10\text{ Pa}} \approx 7 \times 10^5 \frac{\text{m}}{\text{s}}\), the beam electrons are substantially faster at lower pressures. This is caused by the bigger sheath at lower pressure, that expands during the same time interval (5 ns - 19 ns), but over a longer distance \(s_{10\text{ Pa}} = 0.5\text{ cm}, s_{1\text{ Pa}} = 1\text{ cm}\). The corresponding drift energy at 10 Pa is \(E_{d,10\text{ Pa}} \approx 1.5\text{ eV}\), whereas the drift energy at 1 Pa is \(E_{d,1\text{ Pa}} \approx 11\text{ eV}\). Because of this difference in energy, that is deposited in the plasma by beam electrons, it is well understandable, that stochastic heating does only play a significant role at low pressures.

5.2.5. Simple analytical model

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

\[ f = f_0 + f_1 \cos \theta + f_{beam} \]  
(19)

\[ \cos \theta = \frac{v_z}{v} \]  
(20)

The beam part is assumed to be a shifted version of the time independent distribution function:
\[ f_{\text{beam}}(\vec{v}) = \alpha f_0 \left[ (\vec{v} - \vec{u})^2 \right] \Theta(\vec{v} \cdot \vec{u}) \]  

The factor \( \alpha = \frac{n_s}{n_0} \) takes into account that the electron density in the collapsed sheath \( n_s \), which represents the beam density, is much smaller than the density in the plasma bulk \( n_0 \).

According to the hard wall model [34, 35] the velocity \( u \approx 2 \cdot w \) corresponds to twice the sheath expansion velocity \( w \). The Heaviside function \( \Theta \) takes into account that only electrons with positive velocities in the direction of \( u \), i.e. electrons flowing out of the sheath, can originate from the sheath.

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:

\[ g_{\text{beam}}(v) = \alpha \int_0^1 f_0(v^2 + u^2 - 2vu y) dy \]  

Here all velocities are normalized to the thermal velocity \( \sqrt{\frac{2kT}{m}} \). 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:

\[ g_{\text{beam}}(v) = \alpha f_0(v^2)e^{-u^2}e^{\frac{2vu}{v u}} - 1 \]  

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 [26], 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 \( g_{\text{bulk}} = < f > \) finally has the following form:

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

\[ h(v) = 1 + \frac{\tau}{T} \left( e^{-u_0^2}e^{\frac{2vu u}{2vu_0}} - 1 \right) \]  

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 12. Alternatively \( n_s \) is given by the average value of the ion density in the sheath, that was calculated in chapter 5.2.3 (see figure 21). Both ways yield \( n_s \approx 4 \cdot 10^8 \text{ cm}^{-3} \). The density in the bulk is \( n_0 \approx 2 \cdot 10^9 \text{ cm}^{-3} \) (see chapter 5.2.2). Therefore \( \alpha \approx 0.2 \) is assumed. \( \tau \) can be estimated either from the PROES (figure 22) or current measurements (see figure 16). In both cases an electron beam is observed during 15 % of the RF cycle. Therefore, \( \tau \approx 0.15 \) is assumed. The sheath expansion velocity \( w \) can be determined out of the excitation (see figure 22) to be \( 1.1 \cdot 10^6 \text{ cm s}^{-1} \). The electron temperature of the low energetic part of the EEDF, that determines the thermal velocity, is \( kT_e = 2.3 \text{ eV} \) (see chapter 5.2.2). Normalizing \( u = 2 \cdot w \) by the resulting thermal velocity yields \( u_0 \approx 2.5 \).

The resulting form of \( < f > \) 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 at high energies develops. This is clearly shown in figure 23 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 $N = \frac{1}{1 + \alpha (1 - \tau)}$. The form of the distribution function at different times is shown in figure 23 for the case of $u_0 = 2.5$. This Bi-Maxwellian agrees well with the measured EEDF at low pressures (see figure 12).

**Figure 23.** Left graph: Analytically calculated isotropic part of the time average of a Maxwellian distribution superimposed by a shifted Maxwellian distribution for two different beam velocities. Right graph: Distribution functions at two different time intervals and the average value ($u_0 = 2.5$).

5.3. Results at 0.5 Pa

**Figure 24.** Correlation between current, space and phase resolved excitation into $Kr2p_5$ and square of the current (deposited power) in a krypton discharge at 0.5 Pa and 8 W.
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 caused by the PSR effect is clearly pronounced. Figure 24 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. 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. Apparently electrons are accelerated by an electric field everywhere in the bulk at well defined phases. As the current oscillates between positive and negative values, electrons periodically move into different directions either into the bulk or towards the electrode. Similar measurements have been performed in hydrogen at 1.2 Pa and 60 W by our group before [36]. These results are in good agreement with PIC simulations of the power density performed by Vender [37].

6. Conclusion

Cause and effect of electron heating in an asymmetric CCRF discharges have been studied using the synergistic effect of various diagnostics with focus on the PSR effect. FDS in krypton was applied for the first time in order to measure electric fields in the sheath at high spatial and temporal resolution. PROES was used to measure the excitation into a specifically chosen krypton level spatially and temporally resolved within the RF cycle with one dimensional spatial resolution along the discharge axis. The cause of electron heating in terms of the spatio-temporal evolution of the electric field in the plasma boundary sheath and the effect of electron heating in terms of the excitation caused by high energetic electrons were, therefore, investigated. 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, electron mean energy and EEDF.

The measured EEDFs at 1 Pa and 10 Pa are Bi-Maxwellian showing an enhanced high energy tail, that can be attributed to stochastic heating, that is efficient at such low pressures. The measured electric fields at 1 Pa and 10 Pa show the fast sheath expansion and the sheath collapse. An asymmetry of the electric field distribution between sheath expansion and collapse is observed. As the density at the electrode is much lower than in the bulk, the sheath expands very fast at the beginning of each RF cycle. It expands faster than it collapses. This effect is also observed in regard to different amplitudes of the current at the beginning and at the end of each cycle. The observed asymmetry of the electric field is in agreement with this argumentation. During the sheath expansion the generation of beams of high energetic electrons by the expanding sheath could be observed by PROES. Due to an absolute phase calibration of FDS and PROES the effect of electron heating (excitation) can directly be linked to its cause (spatio-temporal evolution of the electric field in the sheath). A simple analytical model demonstrates, that these high 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 was observed both in the excitation and current to the chamber wall. In contrast to the tilted excitation structures caused by electron beams, that propagate at a finite velocity, the structures caused by the PSR effect are straight. Therefore, the excitation mechanisms are different and the straight excitation structures must be caused by an electric field in the bulk, that accelerates all electrons at the same time. The measured currents at high (45 Pa), intermediate (10 Pa) and low (1 Pa) pressures are well reproduced by an analytic PSR model [26]. The assumption, made in this model, that the sheath voltage is proportional to the square of the surface charge density at the electrode, has been verified experimentally.

Based on the time resolved electric field measurements in the sheath the spatially resolved ion density in the sheath was calculated showing good agreement with the electron density at the
sheath edge measured by a Langmuir probe.

Altogether the synergistic use of different diagnostic techniques provides a better understanding of electron 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 radio frequency of the applied voltage. As a consequence a much faster initial sheath expansion takes place, that strongly enhances the formation of an energetic electron beam. Further, the PSR effect leads to high frequency current oscillations, that contribute to the ohmic heating.

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