Striations in dual-frequency capacitively coupled CF$_4$ plasmas: the role of the high-frequency voltage amplitude

Yong-Xin Liu$^1$, Zoltán Donkó$^2$, Ihor Korolov$^3$, Edmund Schüngel$^4$, You-Nian Wang$^1$ and Julian Schulze$^{3,5}$

$^1$Key Laboratory of Materials Modification by Laser, Ion, and Electron Beams (Ministry of Education), School of Physics, Dalian University of Technology, Dalian 116024, People’s Republic of China
$^2$Institute for Solid State Physics and Optics, Wigner Research Centre for Physics, Hungarian Academy of Sciences, H-1121 Budapest, Konkoly-Thege Miklós str. 29-33, Hungary
$^3$Institute for Electrical Engineering, Ruhr-University Bochum, D-44801 Bochum, Germany
$^4$Evatec AG, Switzerland
$^5$Department of Physics, West Virginia University, Morgantown, WV 26506-6315, United States of America

E-mail: yxliu129@dlut.edu.cn

Received 19 January 2019, revised 24 April 2019
Accepted for publication 6 June 2019
Published 25 July 2019

Abstract

Striations in dual-frequency (DF, 8/40 MHz) capacitively coupled CF$_4$ plasmas have been investigated by phase resolved optical emission spectroscopy and via Particle-in-Cell/Monte Carlo collision simulations. The properties of striated structures of various plasma parameters in a DF discharge and the effect of the high-frequency voltage amplitude, $\phi_{H}$, on the striated structures and charged species densities were studied at a gas pressure of 100 Pa. The measured spatiotemporal electronic excitation rates at different $\phi_{H}$ are in good agreement with the simulation results. It was found that the excitation/ionization patterns are modulated not only in space, but also in time by two frequencies. As $\phi_{H}$ increases, the width of a single ion density peak, $d_{\text{peak}}$, generally increases, leading to a decrease of the number of striations and finally to the disappearance of striations at higher $\phi_{H}$. $d_{\text{peak}}$ is believed to be determined by a local balance between the generation (via electron-impact ionization and dissociative attachment) and losses (primarily via recombination of the positive and negative ions, and detachment) of ions. We observed a hysteresis of the axial profiles of the measured plasma emission intensity and the simulated electron-impact excitation rate induced by increasing and decreasing $\phi_{H}$ semi-continuously. The dependence of $d_{\text{peak}}$ on $\phi_{H}$ was found to play a key role in the appearance of the hysteresis.

Keywords: striations, electronegative plasma, low pressure, capacitively coupled RF discharge

1. Introduction

Capacitively coupled radio-frequency (CCRF) discharges operated in electronegative gases, such as O$_2$, CF$_4$, SF$_6$, etc, have been widely used in material processing industries, such as thin film deposition, dielectric etching, etc [1–3]. Therefore, electronegative discharges have attracted tremendous attention from researchers in both academia and industry [4, 5]. Due to the presence of negative ions, electronegative CCRF discharges behave in a different and more complex way as compared to electropositive discharges [6–9]. They often exhibit much lower electron densities [10, 11], a high electric field in the bulk region [12], and double layers [13–16], which greatly alter the discharge structures. Besides $\alpha$- and/or $\gamma$-mode, that commonly occur in electropositive discharges [17, 18], another electron power absorption mode, termed as ‘drift-ambipolar (DA)’ mode, analyzed in details in, e.g. [19–21], is dominant at high electronegativity, $\mu$, which
is defined as the ratio of the negative ion density, \(n_n\), to the electron density, \(n_e\), averaged over the electrode gap, i.e. 
\[ \mu = \frac{n_n}{n_e}. \]
In this mode, the electrons gain energy from a drift field throughout the plasma bulk and from a steep ambipolar field at the edge of the collapsing sheath. A transition between \(\alpha\)-, \(\gamma\)-, and DA-modes can occur when changing external parameters, such as the working pressure, the RF voltage, the driving frequency, etc [22–29]. The electronegativity, \(\mu\), is believed to play a key role in these mode transitions. At a high electronegativity, i.e. \(\mu \gg 1\), the discharge is generally operated in DA-mode, while it turns into \(\alpha\)- or \(\gamma\)-mode at a low electronegativity, i.e. \(\mu \sim 1\) [21, 23].

Generally, in these plasma operating modes ions are considered to be at rest on the time scale associated with the radio frequency, i.e. non-responsive to the drift field inside the plasma bulk, due to their large mass. At the same time, electrons can be accelerated by the instantaneous drift field and can obtain substantial energy, leading to excitation/ionization of neutrals. At conditions when the ion plasma frequency becomes comparable to or higher than the driving frequency, positive and negative ions may respond to the RF electric field with an oscillating motion, generating space charges wherever there are ion density gradients [30–32]. The electric field caused by the space charges enhances or attenuates the local drift electric field in the bulk, resulting in a spatially modulated electric field profile. The total field reinforces the response of the positive and negative ions to the alternating RF electric field by pushing them towards the ion density maxima. Consequently, the space charges, as well as the striated electric field are enhanced due to a positive feedback. The effect is self-amplified until a periodic steady state is established. The striated electric field results in the spatial modulation of the electron power absorption rate and, consequently, of the electron-impact excitation/ionization rate.

We have reported experimental observations of self-organized striated structures of the plasma emission in CCRF CF<sub>4</sub> plasmas driven at a frequency of 8 MHz by phase resolved optical emission spectroscopy (PROES) [30]. These observations were reproduced and analyzed by Particle-in-Cell/Monte Carlo collision (PIC/MCC) simulations. Moreover, an analytical model indicated that the resonance between the eigenfrequency of the ion–ion system and the driving frequency is the basis for the formation of the striated structures. The simulation results showed that the plasma eigenfrequency, \(\omega_{eigen}\), calculated based on the \((\text{CF}_4^+ + \text{F}^-)\) ion density minima in the bulk region is near the driving frequency, \(\omega_{RF}\), i.e. 
\[ \omega_{eigen} = \sqrt{\omega_d^2 + \omega_e^2} \approx \omega_{RF}, \]
which confirmed the prediction given by the analytical model [31]. The striation gap (defined as the distance between two ion density peaks) was found to be approximately inversely proportional to the driving frequency and the working pressure, while it exhibited a weak dependence on the RF voltage and the electrode gap [31, 32]. A transition between the ‘striated’ and ‘non-striated’ modes was observed by changing either the pressure or the RF voltage; for 13.56 and 18 MHz driving frequencies, a phase diagram as a function of the pressure and voltage amplitude was constructed to present the parametric window for the presence of the striations. It was found that the striations are generally present at higher pressures or RF voltages, where the ion density exceeds a ‘critical value’ so that they can respond to the RF electric field inside the plasma bulk. It should be noted that all the work mentioned above has been done for single-frequency (SF) CCRF discharges [30–32].

Dual- or multi-frequency CCRF discharges, however, have been extensively used as well as SF driven discharges, in particular, for some important applications, such as plasma enhanced chemical vapor deposition and plasma etching in the semiconductor industry, due to their ability of independent control of the energy and flux of ions bombarding the electrode [33–41]. In classical dual-frequency (DF) CCRF plasmas, one frequency is high (\(\geq 27.12\) MHz) and, thus, the high-frequency (HF) voltage/power determines the plasma density, while the other frequency is typically low (\(< 13.56\) MHz), so the low-frequency (LF) voltage/power controls the sheath potential drop as well as the ion energy. In DF capacitive discharges, the charged species dynamics becomes more complex, due to coupling effect of the HF and LF oscillations. Based on the PROES technique, Gans et al observed a strong coupling of two frequencies in the emission profiles in a He–O<sub>2</sub> discharge, i.e. the electron ionization dynamics is determined jointly by both frequencies and the plasma density determined by the HF source is also influenced by the LF source [42]. The frequency coupling effect was found to be influenced by the relative voltage amplitudes and phase of the two sources with different frequencies [43]. Dissimilar from those characteristics of electropositive discharges, the electron dynamics was found to exhibit frequency coupling effects due to constructive/destructive interaction of drift electric fields originating from the HF and LF sources in electronegative plasmas [20, 21]. So, it is expected that the striations in DF CCRF discharges exhibit some new behaviors, due to the presence of the HF source.

In this work, we first investigate the basic properties of the striations, i.e. the spatially modulated electron power absorption and ionization/excitation dynamics, as well as the ion dynamics, in a DF electronegative capacitive discharge operated at the same LF voltage amplitude of 300 V and pressure of 100 Pa as in [30], while the HF source frequency is chosen to be \(f_H = 40\) MHz, and the HF voltage amplitude is set to \(\phi_H = 80\) V. This serves as the basis for an analysis of the effect of the HF voltage amplitude on plasma parameters. Subsequently, we change \(\phi_H\) and study its effects on the striated discharge structures. Finally, a hysteresis of various plasma parameters induced by increasing/decreasing \(\phi_H\) is presented and analyzed in details.

The paper is organized in the following way: in section 2, the experimental setup including diagnosticks, and the simulation method are described. In section 3, the results are given and discussed in three parts, as described above. Finally, conclusions are drawn in section 4.
2. Experiment and PIC/MCC simulations

2.1. Experimental setup

The plasma reactor together with the diagnostic tools is schematically shown in figure 1 (also described in [22]). The plasma is produced in CF$_4$ with a 5% admixture of Ne as a probe gas for PROES, between two parallel circular stainless-steel electrodes. Both electrodes are 10 cm in diameter, separated by 1.5 cm, and surrounded by Teflon. The gas mixture is first introduced through a central tube along the axis of the upper electrode and then spreads outwards through 8 outlets. The total gas flow rate was fixed at 25 sccm via a mass flow controller. The chamber is made of stainless steel with an inner diameter of 28 cm. A two-channel function generator (RIGOL, DG4162) is used to generate two sinusoidal signals (8 MHz/40 MHz) with a locked phase ($\varphi = 0^\circ$), and then these two signals are amplified by two power amplifiers (AR, Model 1000A225) and then applied to the lower electrode via two matching networks. The upper electrode and the chamber wall are grounded. A high-voltage probe (Tektronix P6015A) connected to a digital oscilloscope (LeCroy, Waverunner) was used to monitor the voltage waveforms at the powered electrode. The amplitudes of the HF and LF sources were recorded by a home-made program based on the MATLAB GUI. The target voltage amplitudes of HF and LF sources were fixed at 100 Pa, the LF voltage amplitude at 300 V, and the HF voltage amplitude is adjusted in the range of 0–160 V.

An intensified charge-coupled device (ICCD) camera (Andor iStar DH734) equipped with an objective lens is used to detect the light emission from the plasma. An interference filter (center wavelength: 589 nm, full width-half maximum: 10 nm) is placed behind the objective lens to select the Ne 2p$_1$ – 1s$_2$ 585.5 nm transition. The ICCD camera is fixed at a distance of one metre away from the view port of the chamber. The camera gate is controlled in a synchronized manner by a pulse delay generator (SRS INC., Model DG645), which is triggered by a signal with the same frequency as the LF source frequency (8 MHz) provided by the same function generator. The time step and the gate width of the camera are both set to 4 ns in this work. From the light emission measurements, that are performed in a sequence through the entire LF period (~125 ns), the spatio-temporal distribution of the electron-impact excitation rate from the ground state into Ne 2p$_1$-state is calculated based on a collisional-radiative model (for more details see [44, 45]).

2.2. PIC/MCC simulations

Our numerical studies are based on a bounded 1D3V PIC simulation code, complemented with a Monte Carlo type treatment of collision processes (PIC/MCC) [46, 47], which has been described in [21] in details. The code considers one spatial (axial) coordinate and is three-dimensional in the velocity space. The cross sections of electron-CF$_4$ collision processes are taken from [48], with the exception of the electron attachment processes (producing CF$_3$ and F$^-$ ions), for which we use data from [49]. We use an extensive set of electron-impact collision processes, however, disregard many of the products created in these reactions. The only products considered are CF$_3^+$, CF$_3^-$, and F$^-$ ions, which play the most important role in CF$_4$ discharge plasmas. These ions can participate in various ion–molecule reactive processes, as well as in elastic collisions [50–52]. The ion–ion recombination rate coefficients are taken from [27], while the rate for the electron-CF$_3^+$ recombination process is from [26]. In the simulations, we assume a gas temperature of 350 K. We neglect the ion-induced emission of secondary electrons from the electrodes in order to simplify the analysis. We assume that electrons reaching the electrodes are reflected with a
probability of 0.2 [53]. More details of our model, tables, and graphical representations of the cross sections can be found in previous publications, e.g. [54]. The simulations make it possible to determine in a self-consistent manner the spatio-temporal distributions of several discharge characteristics, such as the electric potential and field strength, charged particle densities, fluxes, velocities, as well as reaction (e.g. excitation, ionization, attachment, etc) rates, which provide a deep insight into the physics of the plasma considered.

In the simulations we use 600 grid points to resolve properly the inter-electrode gap and 7500 time steps within a LF period (8 MHz) to resolve properly the temporal dynamics of all plasma species and to fulfill the relevant stability criteria of the numerical method. The PIC/MCC simulations are carried out for parameter sets matching the experimental conditions. It should be mentioned, however, that due to the asymmetry of the experimental electrode configuration, the measured voltage waveforms exhibit a significant negative self-bias for some conditions, which cannot be accounted for by the simulations. In all cases, the HF and LF amplitudes of the measured voltage waveforms are used as input for the PIC/MCC simulations. In section 3.2, the simulation result of a single LF (8 MHz) discharge is used as the initial condition for all the simulations of DF discharges operated at various HF voltages. In the investigation of the hysteresis effect in section 3.3 with increasing and decreasing HF voltage, each simulation run is initialized with the previously converged result, i.e. after one simulation run is converged, a new simulation with an ‘adjacent’ HF voltage amplitude (±4 V with respect to the previous case) is started.

3. Results and discussion

In this section, three parts are included: (i) various plasma parameters, such as the spatially modulated electron-impact ionization/excitation rate, the electric field, the charged species densities, the electron power absorption rate, etc. at the base case (LF voltage amplitude \( \phi_L = 300 \text{ V} \), HF voltage amplitude \( \phi_H = 80 \text{ V} \), electrode gap \( L = 1.5 \text{ cm} \) and pressure \( p = 100 \text{ Pa} \) are discussed in section 3.1. (ii) The effects of increasing the HF voltage amplitude, \( \phi_H \), on the striated structures are studied in section 3.2. (iii) In section 3.3, the hysteresis of various plasma parameters induced by increasing/decreasing \( \phi_H \) is shown and analyzed.

3.1. Properties of the striations at the base case (\( \phi_H = 80 \text{ V} \))

Figures 2(a) and (b) show the spatio-temporal distributions of the measured electron-impact excitation rate from the ground state into the Ne 2p\(_1\) state and the simulated electron-impact ionization rate, respectively, under the conditions mentioned above. One can see from figure 2(a) that at each electrode excitation maxima occur at three distinct times (indicated by the three white arrows) within each half of the LF cycle, due to the modulation of the HF source. Particularly, the measured excitation rate exhibits its maximum (is constructively enhanced) when the oscillation of the HF and LF sheaths is in phase (i.e. the HF and LF sheaths are expanding or collapsing at the same time), while it is nearly invisible, when the motion of the HF and LF sheaths is out of phase (this will be analyzed in more detail later in figure 3). The three enhanced excitation maxima at each electrode within each half of the LF period are caused by the constructive interaction of the drift electric fields originating from the HF and LF sources. It can be seen that the most significant excitation/ionization occurs at the time, \( t_1 \), which is indicated by the left vertical dashed line in the panels of figure 2. This is because both the LF and HF sheaths expand/contract at their fastest speed at \( t_1 \), leading to the maximum RF current within one LF period flowing through the region between the two electrodes and, consequently, the maximum electric field within one LF period inside the bulk region has to build up to drive electrons flowing through the plasma bulk region, due to the low conductivity (low electron density) there. Besides, the measured excitation patterns exhibit a remarkable spatially modulated structure, and the excitation maxima at a certain time occur at fixed axial positions (actually at the edges of high electric field regions), which are determined by the ‘comb-like’ ion density profile (see figure 3(a)). This ion density profile is almost unaffected by the HF oscillation, as the ion plasma frequency is generally lower than the HF source frequency. These experimentally observed patterns are in very good qualitative agreement with the striated spatio-temporal distribution of the ionization/excitation rate obtained from the simulations (figures 2(b) and (c)). This allows us to further explore the details of the striations in electronnegative DF discharges operating in CF\(_4\) based on the additional PIC/MCC simulation results, such as the electron-impact excitation rate, the dissociative attachment rate (corresponding to the formation of F\(^-\) ions), the net charge density, the electric field, the electron power absorption rate and the mean electron energy, which are presented in figures 2(c)–(h).

It has already been found in our previous work that the spatially modulated excitation/ionization rate is caused by the spatially modulated electric field profile (figure 2(f)), which leads to a spatially modulated electron power absorption rate (figure 2(g)), as well as mean electron energy (figure 2(h)) [30–32]. It can be seen in figure 2(f) that, due to the presence of the HF source, in a DF discharge the electric field is not only modulated in space, but also modulated in time by the HF source during each half of the LF period. The total electric field inside the plasma bulk becomes a superposition of the drift field generated by the externally applied DF voltages and the electric field caused by the space charge (figure 2(e)). However, the net charge density exhibits a similar spatio-temporal distribution as that in a single 8 MHz discharge, because the positive and negative ions do not react to the HF electric field, but solely to the LF electric field inside the bulk region.

In addition, one can see that the calculated electron-impact excitation rate (figure 2(c)) is higher than the ionization rate (figure 2(b)), and the excitation patterns are broader in time than those of the ionization, due to the much lower threshold energy of electron-impact excitation. For the measured excitation rate in figure 2(a), the asymmetric spatio-temporal distribution for
Each half of the LF period is attributed to the fact that a negative DC self-bias is formed in the experiment (which can be seen in the waveform in the bottom panels of figure 2), due to the larger effective area of the grounded electrode compared to the powered electrode.

In order to further understand the underlying physics behind the striated excitation/ionization patterns, the spatial profiles of various plasma parameters are shown in figure 3 at the times \( t_1 = 0.28 T_L \) and \( t_2 = 0.38 T_L \), indicated by the two vertical dashed lines in figures 2 and 4(a1), (b1), and (c1). One can see from figures 3(a) and (b) that the \( \text{CF}^+ \) and \( F^- \) ion densities exhibit ‘comb-like’ profiles, which are similar to those observed in a SF 8 MHz discharge [30–32]. At the times \( t_1 \) and \( t_2 \), the positive ions move towards the powered electrode, while the negative ions move towards the grounded electrode. As a consequence, spatially alternating net charge profiles are produced at the positions, where ion density gradients are present. By comparing the net charge density profiles at times \( t_1 \) and \( t_2 \) (see the pink curves in figures 3(a) and (b)), one can find that they are quite similar, suggesting that the positive and negative ions barely respond to the oscillation of the HF electric field. So, the electric field caused by the space charge does not change significantly from time \( t_1 \) to \( t_2 \). By contrast, the absolute value of the minima of the electric field, which are a superposition of the HF and LF drift electric fields and the space charge field, decreases from \( t_1 \) to \( t_2 \). Therefore, the excitation/ionization rate reaches its maximum at time \( t_1 \), while it is largely weakened at time \( t_2 \). This can also be further understood by comparing the profiles of the electron heating rate at times \( t_1 \) and \( t_2 \) in figures 3(e) and (f). It can be seen that the profile of the electron power absorption rate ‘mirrors’ that of the electric field at time \( t_1 \), as the electron flux is uniform along the bulk region, while the electron power absorption rate exhibits a similar profile as the electric field at time \( t_2 \), due to the fact that electrons now move towards the opposite direction with respect to that at time \( t_1 \) (see figure 4(c2)).

Similar to the results pertaining a SF discharge, one can see from figure 3(c) that the excitation/ionization maxima basically occur at the right edges of the high electric field regions inside the bulk (indicated by the vertical dashed lines in figure 3(c)), and they concentrate at the side of the collapsing LF sheath when both the HF and the LF sheaths are at their contracting phase, as it takes some distance for the
electrons to be accelerated to the ionization/excitation threshold energy (towards the ground electrode). By contrast, the excitation/ionization is quite weak after the time \( t_2 \) when the HF and LF sheaths are out of phase (see figure 3(d)), as the electric field changes its sign in the axial direction (see figure 3(f)).

Due to their small mass, electrons can respond to the electric field instantaneously, so that the axial profile of the electron density is determined by the axial profile of the transient electric field. At the time \( t_1 \), there is no significant electron density peak inside the bulk region, as the electric field is not spatially alternating, while at time \( t_2 \) the electron density profile exhibits peaks, due to the spatially alternating electric field profile inside the bulk region. Although the electrons do not contribute much to the net space charge due to their much lower density compared to the ion densities (the electron density is \( \sim 100 \) times lower than the densities of the \( \text{CF}_3^+ \) and \( \text{F}^- \) ions), they play the key role in creating the striated excitation/ionization pattern after gaining substantial energy from the spatially modulated electric field.

Figure 4 shows the spatio-temporal distributions of the densities (left column), fluxes (middle column), and mean axial velocities (right column) of the main charged species: \( \text{CF}_3^+ \) ions (upper row), \( \text{F}^- \) ions (middle row), and electrons (bottom row) for the spatial range \( 0.2 < x/L < 0.8 \) under the same conditions as in figure 2. The densities of the \( \text{CF}_3^+ \) and \( \text{F}^- \) ions are not modulated by the DF oscillating electric field (see figures 4(a1) and (b1)), but the fluxes and the mean axial velocities exhibit complex spatial and temporal structures modulated by the two frequencies (see figures 4(a2), (b2), (a3) and (b3)). However, it should be noted that the spatio-temporal profiles of the ion fluxes and mean velocities exhibit quite different behaviors. The ion flux profile is modulated not only by the LF source, but also by the HF source, while the mean ion velocity profiles (see figures 4(c1) and (c2)) highly resemble that of the electric field inside the bulk region. This is because the ion flux is a product
of the ion density and the mean axial velocity. We can see from the middle column of figure 4 that at the positions of the ion density maxima, the positive and negative ions are accelerated into the opposite directions, and lag behind the HF drift electric field by a phase of $\pi/2$, suggesting that they respond to the HF drift electric field, while, it is hard to see from the right column of figure 4 that the axial ion velocity is affected by the HF drift electric field. The mean ion velocities, whose profiles look quite different from the ion flux profiles, are approximately proportional to the local electric field strength, because the ions move a short distance per half RF period, which is significantly smaller than the width of the high electric field regions, so that they only experience the local electric field.

The response of the ions to a DF electric field can be analyzed in figure 5, which displays the transient electric field and the mean axial velocity of the $\text{CF}_2^+$ ions at the positions $x_1 = 0.5L$ and $x_2 = 0.53L$ (which correspond to the positions of the ion density minimum and maximum, respectively) as a function of time within one LF period under the same conditions as in figure 2. At $x_1$, the electric field is a superposition of a LF and a HF electric fields, which are enhanced due to the presence of the space charge and, thus, the total electric field exhibits a much larger amplitude, compared to that at $x_2$, where the electric field is characterized by a single HF oscillation. Unlike the DF electric field, the $\text{CF}_2^+$ ion axial velocity at $x_1$ (see red dashed curve in figure 4(a)) shows a SF oscillation, and lags behind the electric field by a phase of $\pi/2$, suggesting that the motion of ions is dominated by the LF electric field. Please note that the red dashed sine curve in figure 5(a), however, is somewhat distorted, due to the influence of the HF electric field. The influence can be clearly seen from figure 5(b) in the sense that the variation of the $\text{CF}_2^+$ ion axial velocity with time behaves like the HF electric field at $x_2$, but with the same phase lag of $\pi/2$.

The different spatial scales of the response of the ions to the LF and HF electric fields can be quantitatively analyzed by calculating the relative displacement of the positive and negative ions in a DF electric field. Based on our previous

![Figure 4. PIC/MCC simulation results: spatio-temporal plots of the $\text{CF}_2^+$ ion density, flux, and mean axial velocity (first row), $\text{F}^-$ ion density, flux, and mean axial velocity (second row), and electron density, flux, and mean axial velocity (third row) for the spatial region $0.2 < x/L < 0.8$ under the same conditions as in figure 2. Note that in the first column the vertical dashed lines indicate the times, at which the electron, $\text{CF}_2^+$, and $\text{F}^-$ ion density profiles are shown in figure 2(a).]
work in [31], the motion of the positive and negative ions immersed in a DF electric field can be analyzed by solving analytically a one dimensional uniform slab ion–ion plasma model

\[
\ddot{x}_- + \nu \dot{x}_- - \omega_-^2 (x_- - x_0) = -\frac{e}{m_-} [E_L \cos(\omega_L t) + E_H \cos(\omega_H t)]
\]

for the negative ions and

\[
\ddot{x}_+ + \nu \dot{x}_+ + \omega_+^2 (x_+ - x_-) = \frac{e}{m_+} [E_L \cos(\omega_L t) + E_H \cos(\omega_H t)]
\]

for the positive ions. Here, \(x_\pm, m_\pm, \nu, \omega_\pm\) are the displacements, the masses, and plasma angular frequencies of the negative and positive ions, respectively, \(\nu\) is the ion-neutral collision frequency, and \(e\) is the elementary charge. \(E_L/\omega_L\) and \(E_H/\omega_H\) are the amplitudes/angular frequencies of the LF and HF electric fields, respectively. Please note that the phase shift \(\varphi\) between the LF and HF electric fields is set to zero, which is the case in this work, as the relative phase of the externally applied LF and HF voltages is set to zero.

These two coupled equations describe the motion of both ion species in the LF and HF electric fields. By subtracting equation (1) from (2), we obtain

\[
(m_+ + m_-) \ddot{x}_+ + m_- \ddot{x}_- - \omega_+^2 (x_+ - x_-) = \frac{e}{m_-} [E_L \cos(\omega_L t) + E_H \cos(\omega_H t)].
\]

We define \(x = x_+ - x_-\), \(\omega_+^2 = \omega_-^2 + \omega_0^2\), \(\omega_0^2 = \frac{e^2 n}{\varepsilon_{0} m\nu}\), \(\beta_L = \frac{eE_L}{\mu}\), and \(\beta_H = \frac{eE_H}{\mu}\), where \(x\) is the relative displacement of the positive and negative ions, \(\omega_0\) is the eigenfrequency of the positive and negative ion plasma, and \(\mu\) is the reduced mass of the positive and negative ions.

The coupled motion of positive and negative ions can be described with

\[
\ddot{x} + \nu \dot{x} + \omega_0^2 x = \beta_L \cos(\omega_L t) + \beta_H \cos(\omega_H t).
\]

In order to obtain an analytical solution, the same assumptions as in [31] are adopted here. When the system reaches steady state \((t \to \infty)\), the analytical solution of equation (4) is:

\[
x(t) = \frac{\beta_L}{\sqrt{(\omega_L^2 - \omega_0^2)^2 + \nu^2 \omega_0^2}} \sin(\omega_L t + \varphi_L) + \frac{\beta_H}{\sqrt{(\omega_H^2 - \omega_0^2)^2 + \nu^2 \omega_0^2}} \sin(\omega_H t + \varphi_H)
\]

with \(\varphi_L = \frac{\omega}{\sqrt{(\omega_L^2 - \omega_0^2)^2 + \nu^2 \omega_0^2}}\) and \(\varphi_H = \frac{\omega}{\sqrt{(\omega_H^2 - \omega_0^2)^2 + \nu^2 \omega_0^2}}\).

It can be seen from equation (5) that the relative displacement of the positive and negative ions in a DF electric field is actually a linear superposition of their relative displacements in a LF and a HF electric field, i.e. \(x(t) = x_L(t) + x_H(t)\), where \(x_L(t) = A_L \sin(\omega_L t + \varphi_L)\), and \(x_H(t) = A_H \sin(\omega_H t + \varphi_H)\) are the relative displacements of the positive and negative ions in a single LF and a single HF electric field, respectively, and \(A_L = \sqrt{\omega_L^2 - \omega_0^2 + \nu^2 \omega_0^2}\) and \(A_H = \sqrt{\omega_H^2 - \omega_0^2 + \nu^2 \omega_0^2}\) are the amplitude of \(x_L(t)\) and \(x_H(t)\), respectively.

More specifically, we consider the ion motion at the position \(x_0\), where the ion density is minimal and the electric field exhibits a DF oscillation, i.e. the scenario in figure 5(a). At this position, the ion–ion plasma eigenfrequency, \(\omega_0\), is close to the LF frequency, \(\omega_L\), i.e. the ion–ion plasma system is in resonance with the LF electric field, and the amplitude of the ions’ displacement reaches the maximum. For simplicity, we assume \(\omega = \omega_L\), so equation (5) reduces to

\[
x(t) = \frac{eE_L}{\mu \nu \omega_L} \sin(\omega_L t) + \frac{eE_H}{\mu \sqrt{(\omega_H^2 - \omega_L^2)^2 + \nu^2 \omega_0^2}} \sin(\omega_H t + \varphi_H).
\]

At the base case of this study [31], \(\nu = 1.1 \times 10^3\) m s\(^{-1}\), we have
which has exactly the same form as in the SF case [31]. Therefore, it is concluded that the motion of the positive and negative ions is dominated by the LF electric field when the HF and LF is significantly different. However, the case will be more complex if the the HF and LF are close to each other, which is worth to study in the future.

Due to the small mass, electrons can respond instantaneously to the HF electric field, so that the electron flux is almost in phase with the HF electric field inside the bulk. Its absolute value exhibits a maximum, when the LF and HF sheaths are in phase, while it reaches a minimum when the LF and HF sheaths are out of phase. The mean electron absorption rate (figure 4(c3)) is observed to be more significantly modulated in space than the mean electron energy (figure 2(h)), with the maxima occurring at the right edges of the high-field regions during the first half of the LF period.

### 3.2. Effects of increasing the HF voltage

The effects of increasing the HF voltage amplitude, $\phi_H$, on the striated structure of different plasma parameters are investigated in this section. Please note that the simulations of DF discharges operated at various $\phi_H$ are all initialized by the converged result of a single LF (8 MHz) discharge. Similarly, in the experiment, each DF discharge is also ignited as a single 8 MHz discharge. Figure 6 shows the dependence of the spatio-temporal distributions of the measured electron-impact excitation rate (first column), the calculated ionization rate (second column), electric field (third column), net charge density (forth column) and electron power absorption rate (fifth column) on the HF voltage amplitudes under otherwise the same conditions as in figure 2. Generally, we find good agreement between the spatio-temporal distributions of the electron-impact excitation rates measured experimentally and the electron-ionization rate obtained from the simulation. As $\phi_H$ increases, the distance between two adjacent excitation/ionization maxima increases both in the experiment and in the simulation, and the striated structures of the excitation or ionization vanish at $\phi_H = 120$ V. This can also be observed based on the spatio-temporal plots of the electric field, the net charge density and the electron power absorption rate in the three right columns of figure 6. By increasing $\phi_H$, the distance between two adjacent high field (or high electron power absorption) regions is enlarged, while the width of the high field (or high electron power absorption) regions do not exhibit a significant dependence on $\phi_H$. This is linked to the axial profiles of the ion density at various $\phi_H$ in figure 7.

To be more specific, for the single LF case the excitation/ionization patterns occur at the edge of the collapsing sheath within each half of the LF period, which is exactly the same as found in our previous work [30]. With the addition of the HF voltage, e.g. at $\phi_H = 20$ V, the excitation/ionization patterns that occur at the collapsing phase of the sheath begin to split into two maxima at two distinct times, and the left one is much stronger in intensity than the right one (see figures 6(a2) and (b2)). As $\phi_H$ increases to 60 V, one can clearly observe three excitation/ionization maxima per half of each LF period, that occur at the times when the HF and LF sheaths are simultaneously in their collapsing phase. It is apparent that the patterns in the middle are much stronger, because both the HF and LF sheaths contract at their highest speed (see figure 6(c3)). With the further increase of $\phi_H$, the spatio-temporal distributions of the excitation/ionization rate, electric field, electron power absorption rate and other plasma parameters become much stronger modulated by the HF source, so that the intensities of the three excitation/ionization patterns per each half LF period become similar. At $\phi_H = 120$ V, the striated discharge structures vanish and the excitation/ionization patterns caused by the expanding sheath are observed, due to enhanced HF sheath heating. Meanwhile, the drift electric field inside the bulk region caused by the HF current is enhanced as $\phi_H$ increases, leading to an enhanced coupling effect between the two frequencies. It should be noted from figure 6(e4) that a net charge density can still be observed close to the edge of the plasma bulk due to the fact that the positive and negative ions move into opposite directions, although the striations are not present. This suggests that the positive and negative ions still respond to the RF electric field inside the bulk region at $\phi_H = 120$ V, as the densities of positive and negative ions are much higher than their ‘critical densities’, that are determined by the resonance condition [30, 31].

The axial profiles of the time-averaged $C_F^+$, $F^-$, $C_F^-$ ion and electron densities obtained from the PIC/MCC simulations at different $\phi_H$ are illustrated in figure 7 under the same conditions as in figure 6. At $\phi_H \leq 100$ V, the $C_F^+$, $F^-$ ion densities exhibit ‘comb-like’ profiles. Particularly, as $\phi_H$ increases, the width of a single ion density peak, $d_{\text{peak}}$ (defined as the full width at half maximum of a single ion density peak) is gradually increased, so that the ion density peaks vanish one by one until one peak is left at $\phi_H = 120$ V. Similar to the finding in our previous work [30–32], for a given $\phi_H$ when the striations are present, the minima of the densities of the major ion species ($C_F^+$ and $F^-$) are observed to be comparable in the bulk region, as the minimum densities of the different ions are determined by the requirement to fulfill the resonance condition. The enlarged $d_{\text{peak}}$ at higher $\phi_H$ can be understood by a local balance between the ion generation, the net ion inflow and the ion losses for a single ion density peak. The positive and negative ion generation are, respectively, determined by the electron-impact ionization and dissociative attachment, the net ion inflow is primarily dominated by the focusing effect of the spatially modulated electric field, i.e. the positive and negative ions are constantly pushed towards the positions of the ion density maxima by the spatially modulated electric field in between two adjacent ion density maxima [31], and the ion losses are mainly dominated by the recombination of the positive and negative ions and the detachment of the negative ions. With the increase of $\phi_H$, the ionization and dissociative attachment rates are increased and, furthermore, more positive and negative ions generated in regions of high electric field...
between two ion density maxima) are focused into the positions of the ion density maxima. In order to ensure a balance between the enhanced ion generation and the losses, the ion density peak must broaden in space to enhance the ion losses via, e.g. recombination within a specific ion density peak. This explains why a single ion density peak widens and its peak value increases simultaneously.

The calculated maxima \((n_{\text{CF}_3,\text{max}}, n_{\text{F},\text{max}})\) and minima \((n_{\text{CF}_3,\text{min}}, n_{\text{F},\text{min}})\) of the \text{CF}_3^+ and \text{F}^- ion density inside the bulk region, the space-averaged electron density, \(n_e\), (multiplied by...
Figure 7. PIC/MCC simulation results: axial profiles of the time-averaged CF$_3^+$, F$^-$, CF$_3$ ion and electron densities in the range of $\phi_H = 0$–120 V. The electron density is multiplied by a factor of 10. The other conditions are the same as in figure 6.

Figure 8. PIC/MCC simulation results: maxima and minima of the CF$_3^+$ (red squares) and F$^-$ (blue triangles) ion densities, the space-averaged electron density (black circles) inside the bulk region (multiplied by a factor of 100), electronegativity, $\mu$, (magenta diamonds), and space averaged densities of CF$_3^+$ (red plus sign) and F$^-$ (blue crosses) ions as a function of $\phi_H$. The electronegativity is defined as the ratio of the space-averaged F$^-$ ion density to the electron density. The discharge conditions are the same as in figure 6.

In this section, we discuss a hysteresis of various plasma parameters, including the plasma emission intensity/electron excitation rate, and charged species densities and their spatial profiles, induced by increasing and decreasing $\phi_H$ quasi-continuously under the conditions: $f_L = 8$ MHz, $f_H = 40$ MHz, $\phi_L = 300$ V. The experimentally measured axial profiles of the plasma emission intensity as a function of $\phi_H$ are compared to the electron-impact excitation rate obtained from the PIC/MCC simulations, and a good qualitative agreement is achieved. Then, the properties of the hysteresis are further described and analyzed based on additional quantities obtained from the simulation, including the CF$_3^+$ ion density maximum, $n_{\text{CF}_3^+\text{max}}$, and minimum, $n_{\text{CF}_3^+\text{min}}$; the space-averaged electron density, $n_e$, the widths of a single ion density peak and the plasma bulk, and the charged species profiles at various $\phi_H$.

3.3. Hysteresis induced by the HF voltage

In this section, we discuss a hysteresis of various plasma parameters, including the plasma emission intensity/electron excitation rate, and charged species densities and their spatial profiles, induced by increasing and decreasing $\phi_H$ quasi-continuously under the conditions: $f_L = 8$ MHz, $f_H = 40$ MHz, $\phi_L = 300$ V. The experimentally measured axial profiles of the plasma emission intensity as a function of $\phi_H$ are compared to the electron-impact excitation rate obtained from the PIC/MCC simulations, and a good qualitative agreement is achieved. Then, the properties of the hysteresis are further described and analyzed based on additional quantities obtained from the simulation, including the CF$_3^+$ ion density maximum, $n_{\text{CF}_3^+\text{max}}$, and minimum, $n_{\text{CF}_3^+\text{min}}$; the space-averaged electron density, $n_e$, the widths of a single ion density peak and the plasma bulk, and the charged species profiles at various $\phi_H$. 

high $\phi_H$, because the higher ion sources within every single ion density peak can be balanced by an enhanced recombination loss within a given and widened ion density peak, as explained in the last paragraph. Besides, as $\phi_H$ increases from 0 to 120 V, $n_e$ rises by a factor of 1.7, while the space averaged densities of CF$_3^+$ and F$^-$ ions increase by factors of 1.74 and 1.8, respectively, leading to a very slight increase of the electronegativity from $\mu = 42$ to 45. It, however, should be noted that over the $\phi_H$ range investigated here, $n_{\text{CF}_3^+\text{max}}$ and $n_{\text{F}^-\text{max}}$ are always much higher than their critical densities inside the bulk region, suggesting that the positive and negative ions can respond to the LF electric field and move toward opposite directions, although the striations are absent at $\phi_H = 120$ V.
Figure 9. Evolutions of the axial distribution of the measured emission intensity for the Ne emission line at 585 nm ((a) and (c)) and simulated electronic excitation rate ((b) and (d)), as a function of $\phi_H$. The top two panels indicate the results obtained when increasing $\phi_H$ to 160 V, while the bottom two panels show the results obtained when decreasing $\phi_H$ in panels (a), (b), and (d), respectively, indicate the critical $\phi_H$, at which the mode transition occurs and the two dashed lines in panel (c) indicate a voltage drop from $\phi_H = 88$ V to 80 V, that occurs in the experiment when the discharge switches from the ‘non-striated’ to the striated mode by decreasing $\phi_H$. The critical axial emission region is widened with a value of $\pm$ 4 V. In order to make a comparison with the experiment, in the PIC/MCC simulation changing $\phi_H$ by 4 V was implemented in the following way: (i) at a certain $\phi_H$ the simulation is converged, (ii) the simulation at $\phi_H \pm$ 4 V is initialized from these converged results. Figure 9(c) shows that at low $\phi_H$ in the experiment the striations are present, and the width of the intensive emission region does not change significantly with increasing $\phi_H$, until $\phi_H = 108$ V, at which the striations vanish, and then the intensive emission region widens with increasing $\phi_H$. Besides, we found from figure 9(a) that as $\phi_H$ increases the striation gap is enlarged, while the number of striations is reduced. By decreasing $\phi_H$ in the experiment, the intensive emission region narrows persistently until $\phi_H = 88$ V, indicating a shrinking plasma bulk length, as the axial span of the intensive emission (or electronic excitation) region in figure 9 could be considered as an indicator of the width of the plasma bulk. Subsequently, there is a slight voltage drop from $\phi_H = 88$ V to 80 V, that occurs when the plasma bulk broadens suddenly, meanwhile the striations emerge. This is a typical hysteresis induced by $\phi_H$, which has also been observed in the PIC/MCC simulations. It can be seen from figure 9(b) that when $\phi_H$ is increased the striations disappear at $\phi_H = 144$ V, which is higher than that in experiment. Consistent with the experimental results, we also see the widened striation gap and reduced number of striations at higher $\phi_H$. By decreasing $\phi_H$, again, similar to the experimental result (figure 9(c)), the plasma bulk is persistently shrinking until $\phi_H = 92$ V, at which the striations abruptly emerge, suddenly leading to a widened plasma bulk. It should be noted that a major difference between the experimental and the simulation results when decreasing $\phi_H$ in the striated mode is that in the simulation the axial profile of the excitation rate always exhibits 6 maxima inside the bulk region, while in the experiment the number of the excitation maxima.
is gradually increased. The difference between the experimental and the simulation results might be due to the absence of the secondary electron emission from the electrodes and the symmetrical discharge geometry in the PIC/MCC simulations. Nevertheless, the experimental results are clearly reproduced qualitatively by the simulations. Understanding the above differences is beyond the scope of this work.

Figure 10(a) displays the maximum, \(n_{\text{CF}_2\text{max}}\), and minimum, \(n_{\text{CF}_2\text{min}}\), of the \(\text{CF}_2^+\) ion density and the space-averaged electron density, \(\bar{n}_e\), as a function of \(\phi_H\) under the same conditions as in figure 9. Note here that the electron density has been multiplied by a factor of 100, and the digits indicate the number of the ion density peaks inside the bulk region. Figure 10(b) presents the widths of the plasma bulk and of a single ion density peak as a function of \(\phi_H\). The width of the plasma bulk, \(d_{\text{bulk}}\), herein, indicates the full width at half maximum of the entire ‘comb-like’ ion density profile. We can see from figure 10(a) that \(n_{\text{CF}_2\text{max}}\) exhibits a ‘sawtooth-like’ increase with increasing \(\phi_H\), i.e. a density drop occurs whenever one central density peak has vanished (meanwhile one sees abruptly widened single peaks). Again, this can be explained by the local balance between the ion generation and losses of the positive and negative ions. To ensure such a balance, an ion density maximum drop is able to compensate an abrupt broadening of ion density peaks, whenever one central ion density peak vanishes. At \(\phi_H \geq 144\) V, there is only one density peak left inside the bulk (see, e.g. the parabolic density profile at \(\phi_H = 152\) V in figure 11). It, however, should be noted that the ions can still react to the LF electric field. The reduced number of ion density peaks with increasing \(\phi_H\) can be clearly seen from the different panels of figure 11, which illustrates the axial profiles of the charged species densities for an increasing and a decreasing \(\phi_H\) at eight selected values. Please note in figure 11 that for each \(\phi_H\) the ion density peaks with the largest \(d_{\text{peak}}\) generally occur close to the edge of the plasma bulk, where the ionization rate exhibits its maximum. As explained before, a wide ion density peak generated at the edge of the plasma bulk can allow the enhancement of the recombination of the positive and negative ions to balance the high ionization there. With the increase of \(\phi_H\), the peripheral ion density peaks gradually widen, while the ion density peak vanishes consecutively from the center of the plasma bulk (see panels marked by black arrows in figure 11).

The dependence of \(d_{\text{peak}}\) and \(d_{\text{bulk}}\) on \(\phi_H\) can be clearly seen from figure 10(b) which shows that with the increase of \(\phi_H\) \(d_{\text{peak}}\) first grows gently, and then shows a ‘step-like’ increase at \(\phi_H > 92\) V, i.e. a jump in \(d_{\text{peak}}\) occurs whenever one central ion density peak vanishes. Finally, one sees \(d_{\text{bulk}} = d_{\text{p}}\) at \(\phi_H \geq 144\) V, where there is only one ion density peak inside the bulk region. Besides, we find that \(d_{\text{bulk}}\) exhibits a ‘sawtooth-like’ decrease with increasing \(\phi_H\) at \(\phi_H < 84\) V, after that it gradually widens, indicating that \(d_{\text{bulk}}\) exhibits a similar dependence on \(\phi_H\) as the width of the intensive emission region shown in figure 9(b).

By decreasing \(\phi_H\) from 160 V to 92 V, it can be seen from figures 10 and 11 that the \(\text{CF}_2^+\) ion density profile remains unimodal, with both its maximum and width decreasing linearly, due to continuously weakened ionization. At \(\phi_H = 92\) V, the ionization becomes so weak, that the plasma is hardly self-sustained. With a further decrease of \(\phi_H\), the plasma becomes unstable and the parabolic density profile collapses and, as a result, a new and more stable charged species density profile with four peaks is formed at \(\phi_H = 88\) V. During this process one sees in figure 10 an abrupt increases in \(n_{\text{CF}_2\text{max}}, \bar{n}_e, d_{\text{bulk}},\) and the excitation/ionization rate (see figure 9(d)). By turning \(\phi_H\) down to zero, the four-peak ion density profile remains, and \(n_{\text{CF}_2\text{max}}\) declines monotonously with decreasing \(\phi_H\). In the meantime, \(d_{\text{peak}}\) decreases slowly, while \(d_{\text{bulk}}\) remains the same, resulting in an enlarged gap between two adjacent ion density peaks, as can be seen in figures 11(a2), (b2), (c2), and (d). The reason for
the reduced $d_{\text{peak}}$ at lower $\phi_H$ can also be explained by the balance of ion generation and losses, as described before.

It is worth mentioning that $n_{\text{CF}_3,\text{min}}$ inside the bulk region is almost independent of $\phi_H$, regardless of whether $\phi_H$ is increased or decreased, as $n_{\text{CF}_3,\text{max}}$ increases with $\phi_H$ in a similar way as $n_{\text{CF}_3,\text{max}}$, with the exception that an electron density jump occurs, whenever one central density peak vanishes. When decreasing $\phi_H$, $n_e$ exhibits the same dependence on $\phi_H$ as $n_{\text{CF}_3,\text{max}}$.

Figure 11. PIC/MCC simulation results: axial profiles of the time-averaged $\text{CF}_3^+$, $\text{F}^-$, $\text{CF}_3^-$ ion and electron densities (multiplied by a factor of 10) at 8 selected HF voltage amplitudes, i.e. $\phi_H = 0, 40, 60, 80, 88, 92, 132, \text{and} 152 \text{ V}$. Note that the panels marked by black arrows indicate the results obtained when $\phi_H$ is gradually increased from 0 V, while the ones marked by red arrows indicate the results obtained when $\phi_H$ is gradually decreased from 160 V. The profiles at $\phi_H = 80 \text{ V} \text{ and} 152 \text{ V}$ obtained when changing $\phi_H$ in opposite directions are combined into one panel, due to their identical profiles. The other conditions are the same as in figure 9.

4. Conclusion

Previously, we observed the self-organized striated structures of the plasma emission in SF capacitively coupled CF$_4$ plasmas via PROES and their formation was analyzed by PIC/MCC simulations [30–32]. In this work, we extended this topic to dual frequency (DF, 8/40 MHz) discharges under the same conditions as in the SF case ($\phi_H = 300 \text{ V}$, $p = 100 \text{ Pa}$, $L = 1.5 \text{ cm}$). The PROES technique was used to determine the spatiotemporal electron-impact excitation rate in the experiment and these experimental results were compared with the spatiotemporal ionization rate resulting from the PIC/MCC simulations. We first studied the properties of striated structures of various parameters in an electronegative DF capacitive discharge and then the effect of the HF voltage amplitude, $\phi_H$, on the...
striations. It was found that in DF discharges the striated excitation/ionization patterns are modulated by two frequencies. Temporally, the ionization/excitation exhibits maxima when the HF and LF sheaths are in phase, due to constructive interaction of the drift electric field originating from the HF and LF excitation. Spatially, the excitation/ionization maxima occur at fixed axial positions, which are determined by the ‘comb-like’ profile of $\text{CF}_2^+ / F^-$ ion density. Besides, the net charge density shows a similar spatio-temporal distribution as in a SF discharge, indicating that the positive and negative ions do not respond to the HF (40 MHz) electric field inside the bulk region.

The measured spatiotemporal electronic excitation distributions at different $\phi_H$ showed good agreement with the ionization patterns obtained from PIC/MCC simulations. This allowed us to explore the underlying physics behind the striations and to analyze the effect of $\phi_H$ on striations in DF capacitive discharges. It was found that as $\phi_H$ increases, the ion density peak widens, leading to a decrease of the number of striations and finally to a disappearance of the striations at a higher $\phi_H$. The ion density peak width is believed to be determined by a local balance between the ion generation that is dominated by the electron-impact ionization and dissociative attachment and losses that are dominated by recombination of the positive and negative ions. Specifically, at higher $\phi_H$ the ionization is enhanced. So, a single ion density peak has to widen, allowing the enhancement of the recombination to balance the higher ionization.

Particularly, when increasing and decreasing $\phi_H$ quasi-continuously, we observed a hysteresis of the axial profiles of the plasma emission intensity measured experimentally and of the electron excitation rate obtained from the PIC/MCC simulations. When increasing $\phi_H$, the ion density maximum exhibits a ‘sawtooth-like’ increase, i.e. a density drop occurs whenever one central ion density peak vanishes. Oppositely, by decreasing $\phi_H$ the ion density maximum declines monotonically, with a density jump occurring at $\phi_H = 92$ V. By contrast, the ion density minimum is almost independent of $\phi_H$, regardless of whether $\phi_H$ is increasing or decreasing, as it is primarily determined by the LF source frequency (8 MHz). The dependence of the width of the ion density peak on $\phi_H$ was found to play a key role for the appearance of the hysteresis. When increasing $\phi_H$, the ion density peak gradually widens and, as a consequence, central ion density peaks vanish one after another until one density peak is left inside the plasma bulk. When decreasing $\phi_H$ from 160 V the parabolic ion density profile narrows linearly due to continuously reduced ionization, until at $\phi_H = 92$ V the parabolic ion density profile becomes unstable and the plasma collapses, leading to the formation of a new and more stable charged species density profile, that exhibits four peaks inside the plasma bulk at $\phi_H = 88$ V.

Acknowledgments

This work has been financially supported by the National Natural Science Foundation of China (NSFC) (Grant Nos. 11875100, 11722541 and 11335004), by the Hungarian National Research, Development and Innovation Office, via Grant No. NKFIH 119357, by the German Research Foundation (DFG) within the frame of the collaborative research center SFBTR87 (project C1), and by the US National Science Foundation (Grant No. 1601080).

ORCID iDs

Yong-Xin Liu @ https://orcid.org/0000-0002-6506-7148
Zoltán Donkó @ https://orcid.org/0000-0003-1369-6150
Ihor Korolov https://orcid.org/0000-0003-2384-1243
You-Nian Wang @ https://orcid.org/0000-0002-6506-7148
Julian Schulze https://orcid.org/0000-0001-7929-5734

References


[40] Schulze J, Schüngel E, Donkó Z and Czarnetzki U 2010 Excitation dynamics in electrically asymmetric capacitively coupled radio frequency discharges: experiment, simulation, and model Plasma Sources Sci. Technol. 19 045028


[43] O’Connell D, Gans T, Semmler E and Awakowicz P 2008 The role of the relative voltage and phase for frequency coupling in a dual-frequency capacitively coupled plasma Appl. Phys. Lett. 93 081502


and dual frequency capacitively coupled plasma reactors

Phys. Rev. E 69 026406


IEEE Trans. Plasma Sci. 28 971
