Suppression of nonlinear standing wave excitation via the electrical asymmetry effect

Kai Zhao¹, Zi-Xuan Su¹, Jia-Rui Liu¹, Yong-Xin Liu¹,* Ö, Yu-Ru Zhang¹ Ö, Julian Schulze¹,² Ö, Yuan-Hong Song¹ Ö and You-Nian Wang¹ Ö

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 Electrical Engineering, Ruhr-University Bochum, 44801 Bochum, Germany

E-mail: yxliu129@dlut.edu.cn

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Abstract

The electrical asymmetry effect (EAE) enables separate control of the ion flux and the mean ion energy in capacitively coupled plasmas (CCP). While a variety of plasma processing applications benefit from this, large-area, very-high-frequency CCPs still suffer from lateral nonuniformities caused by electromagnetic standing wave effects (SWE). Many of such plasma sources are geometrically asymmetric and are operated at low pressure so that high frequency nonlinear plasma series resonance (PSR) oscillations of the RF current are self-excited. These PSR oscillations lead to the presence of short wavelength electromagnetic waves and a more pronounced SWE. In this work, we investigate the influence of the EAE on the nonlinear standing wave excitation in a geometrically asymmetric, low pressure capacitively coupled argon plasma driven by two consecutive harmonics (30 MHz and 60 MHz) with an adjustable phase shift, $\theta$. We use a hairpin probe to determine the radial distribution of the electron density in combination with a high-frequency $B$-dot probe to measure the radial distribution of the harmonic magnetic field, which in turn is used to calculate the harmonic current density based on Ampere’s law. Our experimental results show that the asymmetry of the discharge can be reduced electrically via the EAE. In this way the self-excitation of high frequency PSR oscillations can be attenuated. By tuning $\theta$, it is, therefore, possible to switch on and off the nonlinear standing wave excitation caused by the PSR and, accordingly, the plasma uniformity can be optimized.

Keywords: capacitively coupled plasmas, electrical asymmetry effect, plasma series resonance, nonlinear standing wave, plasma uniformity

(Some figures may appear in colour only in the online journal)

1. Introduction

A variety of resonance modes exist in bounded low-temperature plasmas [1–3]. The self-excitation of nonlinear plasma series resonance (PSR) oscillations, as one of the most important resonances, has been extensively studied in capacitively coupled radio-frequency discharges [4–18]. This type of
resonance generally features a series of pronounced higher harmonics superimposed on the fundamental driving frequency in the current and electric field in the discharge. Physically, the PSR originates from a periodic exchange between the kinetic electron energy in the plasma bulk and the electric field energy in the plasma sheath, which is analogous to an RLC series resonance in an electrical circuit [6]. The bulk inductance arises from the electron inertia and the sheaths act as two non-linear capacitors, whereas the electron-neutral collisions behave as a resistance and, consequently, the PSR occurs. The presence of the PSR allows the discharge to be excited with a quite small voltage while providing a sufficiently high current for sustaining the plasma. Although the fundamental driving frequency, $\omega$, is normally below the PSR frequency, $\omega_{PSR}$, the nonlinear charge–voltage characteristic of the sheath generates harmonics that can be strongly strengthened at or near the series resonance frequencies, drastically enhancing the local electron power deposition [10, 11]. The amplitude of the PSR oscillations is limited by the electron-neutral collisions in the plasma bulk.

It has been recognized that the discharge asymmetry (either geometrical or electrical) is a necessary condition for the efficient self-excitation of the PSR oscillations. Taking a single-frequency capacitively coupled plasma (CCP) for example, significant PSR oscillations were observed mostly in geometrically asymmetric discharges, where the effective area of the grounded electrode is much larger than that of the powered electrode, since in geometrically symmetric discharges the non-linearities of the sheath charge–voltage relations cancel out due to two nearly identical sheaths [17, 18]. A way to control the discharge asymmetry is to excite a plasma via the electrical asymmetry effect (EAE) [19–39]. It has been shown that by utilizing the EAE, the PSR oscillations can be self-excited even in a geometrically symmetric discharge [14, 17]. More recently, it was revealed that the magnetic asymmetry effect (MAE) provides the opportunity to control the discharge symmetry and consequently the PSR by adjusting the magnetic flux density at the powered electrode [40–43]. Moreover, the presence of different surface materials at the powered and grounded electrode was found to affect the discharge symmetry [44–46].

Over the past decades, the PSR phenomenon in a CCP driven at or below the frequency of the conventional 13.56 MHz has been investigated extensively [4–18]. In these studies, the PSR behavior at different radial positions was expected to be identical and the plasma parameters (e.g. the sheath thickness, the plasma density, the harmonic excitations, etc) in the interelectrode space were assumed to be independent of the radial position. This is justified, only if the half-wavelength of the electromagnetic waves in a plasma is much larger than the reactor dimension, i.e., the standing wave effect (SWE) is negligible. Recently, there is a tendency to power CCP reactors at much higher frequencies, aiming to achieve higher ion fluxes on the substrate (improving throughput), with a concomitant reduction in the ion bombardment energy (minimizing substrate damage) [47]. In this scenario, plasma parameters can no longer be assumed to be radially uniform, because the SWE will come into play due to a shortened wavelength. A 2D model and a spatially resolved measurement of the plasma parameters are, thus, necessary to uncover the underlying physics. Such nonuniformities caused by the SWE are a major problem for large area plasma processing applications. Existing methods to prevent such nonuniformities are limited and technically involved. For instance, dielectric lenses [48] and segmented [49] or graded conductivity electrodes [50] can be used. However, such techniques are expensive and might not be appropriate for dual-frequency excitation. Overall, more flexible and cheaper methods are needed.

Motivated by this, some efforts have been made in understanding the SWE, and a strong relationship between the SWE and the plasma nonuniformities has been demonstrated [51–65]. In published studies [51–65], researchers emphasized the role of the fundamental driving frequency. However, nonlinear effects, such as the higher harmonics excited by the PSR oscillations, have not received much attention. Recently, a correlation between the higher harmonic excitations and the center-peaked plasma density profiles was made by Miller et al. [66], Lane et al. [67], and Sawada et al. [68]. Higher harmonic excitations and the coupling mechanism between the PSR and the SWE were later predicted by Lieberman et al. in theory and simulation [69, 70], and observed by Zhao et al. experimentally [71]. It was shown that in a very-high-frequency (VHF) CCP, the higher harmonics excited by the PSR can induce spatial wave resonances (SWRs), with voltage and current maxima on axis, resulting in a center-high plasma density profile. Until now, experimental investigations on the nonlinear standing wave excitation have mostly been limited to single-frequency CCPs. Relevant studies of dual-frequency or multi-frequency excitation remain sparse, to the best of our knowledge.

It is well known that the frequently used, industrial CCP sources are often geometrically asymmetric. Naturally, such an asymmetry can induce significant PSR oscillations and nonlinear SWEs in a discharge excited by a high frequency and operated at low pressure, which in turn will cause severe plasma nonuniformities. Optimization of the plasma uniformity has, thus, become a major challenge in both academia and industry. Considering the technological difficulties of changing the geometric symmetry of a plasma reactor, the EAE can be considered as an alternative method to control the discharge symmetry and the self-excitation of PSR oscillations. As a consequence, it should be possible to manipulate the nonlinear standing wave excitation and to optimize the plasma uniformity. Illuminating work by Schüngel et al. [72] experimentally demonstrated the possibility to improve the plasma uniformity via the EAE in a CCP operated in the electromagnetic regime. However, the important roles of the self-excitation of the PSR and the nonlinear standing waves were not taken into account.

In this work, we experimentally investigate the influence of the EAE on the nonlinear standing waves induced by the PSR in a geometrically asymmetric, low pressure CCP reactor driven by two high frequency sources (30 MHz and 60 MHz) with an adjustable relative phase shift. To bring the effects of nonlinear harmonic excitations on the plasma uniformity
into clearer focus, we use a hairpin probe to determine the radial distribution of the electron density in combination with a high-frequency B-dot probe to measure the radial distribution of the harmonic magnetic field, which in turn is used to calculate the harmonic current density based on Ampère’s law. This paper is structured in the following way. In section 2, the experimental setup and diagnostics are described. Detailed experimental results regarding the influence of the EAE on the dc self-bias, the nonlinear standing wave excitation and the radial distribution of the plasma density are presented and discussed in section 3. Finally, conclusions are drawn in section 4.

2. Experimental setup

The experimental apparatus equipped with the diagnostic tools is schematically illustrated in figure 1. The cylindrical CCP reactor has an inner diameter of 40 cm, containing two stainless steel parallel disk electrodes, 30 cm in diameter, separated by 4 cm. Highly purified (99.999%) argon gas is injected evenly into the discharge region through the showerhead-like top electrode. The chamber is evacuated to a base pressure of $\leq 10^{-3}$ Pa by a turbomolecular pump backed by a roughing pump. The gas flow rate is set to 30 sccm via a mass flow controller, and the gas pressure is fixed at 4 Pa for all measurements. A 3 mm-thickness Teflon liner is attached to the sidewall of the chamber in order to confine the plasma.

A controllable electrical asymmetry is realized by applying a specific voltage waveform to the bottom electrode, with the top electrode and the chamber wall being grounded. The voltage waveform is a superposition of a fundamental frequency, $\omega = 2\pi f$, and its second harmonic, $2\omega$, with an adjustable relative phase shift, $\theta$, 

$$\phi(t) = \phi_0 [\cos(\omega t + \theta) + \cos(2\omega t)], \quad (1)$$

where $\phi_0 = 30$ V and $f = 30$ MHz. In the experiment, a signal in the form of equation (1) generated by a function generator (Tektronix AFG31252) is amplified by a broadband power amplifier (AR, Model 1000/A225) and then delivered through a matching network to the bottom electrode. Note that the power source is connected capacitively to the electrode through a blocking capacitor, thus establishing a negative dc self-bias. The voltage waveform at the powered (bottom) electrode is measured by a high-frequency, high-voltage probe (Tektronix P5100A, 500 MHz bandwidth) and acquired via a digital oscilloscope (Tektronix MSO56).

In this work, special attention is paid to the measurement of the voltage waveform at the electrode. A specific electric terminal, comprising a central conductor and a thick outer insulating barrier, was installed at one flange on the sidewall, in order to electrically contact the voltage probe (see figure 1). Inside the chamber, the central conductor of the terminal is connected to the powered (bottom) electrode via a 10 mm-long, semi-rigid coaxial cable (RG 405, OD = 2.2 mm). In this way, the distance between the measurement point and the powered electrode is minimized and, thereby, the accuracy of the waveform measurement is improved. Indeed, a comparative measurement at atmospheric pressure indicated that the difference in the measured waveforms (including the amplitude and the phase) for two cases, (i) as the voltage probe is connected to the electric terminal and (ii) to the electrode center in the vented chamber, is reasonably small for frequencies below 60 MHz. The variations of the voltage amplitude and phase on the powered electrode versus radius are expected to be quite small, because the half-wavelengths of both driving frequencies are much larger than the electrode diameter.

Moreover, an absolute calibration of the voltage probe has been carried out. A sample signal, produced by the function generator, is output to one channel of the oscilloscope via a BNC cable. The voltage signal at the output end of the inner conductor of the BNC cable is simultaneously monitored by the voltage probe and recorded by another channel of the oscilloscope. A comparison of the amplitudes and the phases of the waveforms acquired by these two channels yields the amplitude calibration factor, $\alpha$, and the phase calibration factor, $\beta$, of the voltage probe. Given the amplitude $V_{\text{meas}}$ and phase $\theta_{\text{meas}}$ of the measured voltage waveform, their corrected values, $V_{\text{corr}}$ and $\theta_{\text{corr}}$, are given by, $V_{\text{corr}} = \alpha V_{\text{meas}}$ and $\theta_{\text{corr}} = \theta_{\text{meas}} + \beta$, respectively. Here, the calibration factors are $\alpha = 0.91$ and $\beta = 101.1^\circ$ at 30 MHz, and $\alpha = 0.85$ and $\beta = 210.3^\circ$ at 60 MHz.

To ensure the presence of the correct voltage waveform shape at the powered electrode, an automated feedback control program based on a MATLAB instrument control toolbox (ICT) is developed. Two primary functions are integrated into the program; one is reading waveforms from an oscilloscope and the other is creating and downloading an arbitrary waveform to a function generator. The feedback procedures to produce a target waveform is as follows. (i) An initial waveform (defined by equation (1)) with an amplitude $\phi_{\text{in},m}$ and a phase $\theta_{\text{in},m}$ (the subscript $m = 1, 2$ denotes the harmonic order), comprising 10,000 points per cycle, is created on a computer and then downloaded to the function generator. Subsequently, it is amplified and applied to the bottom electrode. (ii) The voltage waveform applied to the powered electrode is recorded by the oscilloscope, downloaded to the computer, and is analyzed online via the fast Fourier transform (FFT) algorithm, yielding its amplitude $\phi_{\text{out},m}$ and phase $\theta_{\text{out},m}$. (iii) $\phi_{\text{out},m}$ and $\theta_{\text{out},m}$ are, respectively, compared with the amplitude $\phi_{\text{ targ},m}$ and the phase $\theta_{\text{ targ},m}$ of the target waveform, and then $\phi_{\text{ in},m}$ and $\theta_{\text{ in},m}$ are updated individually, in order to bring $\phi_{\text{out},m}$ and $\theta_{\text{out},m}$ closer to $\phi_{\text{ targ},m}$ and $\theta_{\text{ targ},m}$, respectively. (iv) Iterate the procedures above until the constraint conditions $0.98 \leq \phi_{\text{out},m}/\phi_{\text{ targ},m} \leq 1.02$ and $|\theta_{\text{out},m} - \theta_{\text{ targ},m}| \leq 2^\circ$ are satisfied.

To reveal the effects of nonlinear harmonic excitations on the plasma, we use a high-frequency B-dot probe to measure the radial distribution of the harmonic magnetic field. A detailed description of the structure, calibration and experimental validation of the B-dot probe has been given in reference [71]. The B-dot probe is combined with a real time numerical technique to suppress the capacitive pickup, thus extracting a relatively accurate inductive signal. An in-house program based on MATLAB ICT is used to communicate with...
In the case of the current experiment, the discharge current is dominated by all the magnetic field Fourier components. The frequency domain via the FFT, yielding the amplitudes of the frequency range 1–300 MHz, thus yielding the magnetic field strength to the output voltage of the oscilloscope, enabling us to collect and analyze data online.

An absolute calibration of the B-dot probe was carried out with a standard magnetic field produced by a Helmholtz coil, over the frequency range 1–300 MHz, thus yielding the magnetic sensitivity (i.e. the ratio of the magnetic field strength to the output voltage of the B-dot probe) at each frequency. The time-domain signal detected by the B-dot probe is transformed into the frequency domain via the FFT, yielding the amplitudes of all the magnetic field Fourier components.

The relationship between the total current density \(J\) and the magnetic field \(H\) in a CCP follows Ampere’s law,

\[
\nabla \times H = J, \tag{2}
\]

where \(J\) is the sum of the conduction current \(J_c\) and the displacement current density \(J_d = \partial \mathbf{D}/\partial t\). Considering a transverse magnetic mode in a cylindrical CCP reactor and assuming an azimuthal symmetry of the discharge, the magnetic field, \(H_r\), is purely azimuthal and independent of \(\varphi\), whereas the electric current density has two components, i.e. an axial (capacitive) current density \(J_z\) and a radial (inductive) current density \(J_r\). Note that for a lower density plasma, i.e. the case of the current experiment, the discharge current is dominated by \(J_z\). Thus, taking the axial component of equation (2) yields,

\[
1 \frac{\partial (rH_r)}{\partial r} = J_z. \tag{3}
\]

The magnetic field has \(N\) harmonic components, i.e. \(H_{r,n}(r) = \sum_{n=1}^{N} H_{r,n}(r) = \sum_{n=1}^{N} H_{r,n}(r)e^{i\phi_n+\phi_n(r)}\), with \(n\) denoting the harmonic order, and \(H_{r,n}\) and \(\phi_n\) being the amplitude and phase of the harmonic magnetic field, respectively. Thus, given the radial distribution of the harmonic magnetic field \(H_{r,n}\), we can determine the amplitude and phase distributions of the harmonic current density \(J_{z,n}\) at different radial positions.

A hairpin probe, described in detail elsewhere [65], is employed to measure the radial distribution of the electron (plasma) density. The electron density, \(n_e\), is determined by the shift of the resonant frequency in vacuum with respect to the resonance frequency in the plasma. The measured electron density is corrected numerically using a step-front sheath model [73]. The hairpin probe and the B-dot probe are mounted on different translational stages so that they can be moved radially along the mid-plane of the electrodes (see figure 1). Furthermore, an intensified charge-coupled device camera is employed to measure the spatiotemporal distribution of the optical emission intensity within one fundamental frequency period, from which we can calculate the spatiotemporal distribution of the electron-impact excitation rate from the ground state of Ar into the Ar 2p1 state based on a collisional radiative model [65, 74–76].

3. Results and discussion

The dc self-bias, \(\eta\), is a measure of the discharge symmetry that can be induced either geometrically, electrically, magnetically, or by a combination of these mechanisms. Previous work [19, 20, 28] has demonstrated that \(\eta\) depends on the symmetry parameter, \(\varepsilon\), which corresponds to the absolute value of the ratio of the maximum voltage drops across the powered and grounded electrode sheaths,

\[
\varepsilon \approx \left(\frac{A_p}{A_g}\right)^2 \frac{n_{sp}}{n_{sg}}, \tag{4}
\]

which is proportional to the squared ratio of the area of the powered electrode \((A_p)\) and the grounded electrode \((A_g)\) as well as to the ratio of the mean ion density in the sheath at the powered electrode \((n_{sp})\) and at the grounded electrode \((n_{sg})\). For a single-frequency CCP, the maximum of the applied voltage waveform, \(\left|\phi_{\max}\right|\), always equals the absolute value of the minimum, \(\left|\phi_{\min}\right|\). A geometrical reactor asymmetry \((A_p \neq A_g)\) is then one prominent way to induce a discharge asymmetry.

Figure 1. Sketch of the experimental setup of a dual-frequency capacitive argon discharge driven by a fundamental frequency of 30 MHz and its second harmonic of 60 MHz with an adjustable relative phase shift. Both driving frequencies are applied to the bottom electrode.
and to generate a dc self-bias. However, for dual- or multi-frequency CCPs a dc self-bias can arise even in a geometrically symmetric \((A_p = A_g)\) discharge as long as \(\left| \tilde{\phi}_{\text{max}} \right| \neq \left| \tilde{\phi}_{\text{min}} \right|\), which is the concept of the EAE. Previous work has also demonstrated that adjusting the relative phase between two consecutive harmonics used as the driving voltage waveform allows to control the symmetry parameter, \(\varepsilon\), which, in turn, controls the self-excitation of the PSR [17]. For \(\varepsilon = 1\) the self-excitation of the PSR is minimized due to the symmetry of the discharge and the compensation of the sheath-nonlinearities at both electrodes. The stronger \(\varepsilon\) deviates from unity, i.e., the more asymmetric the discharge is, the more strongly the PSR is self-excited, since the sheath nonlinearities no longer cancel out.

For the discharge conditions studied in this work (Ar, 30 MHz + 60 MHz, \(\phi_0 = 30\) V, 4 Pa, 4 cm electrode gap), the variation of the dc self-biases, \(\eta\), with the phase shift, \(\theta\), is shown in figure 2(a). \(\eta\) increases monotonically from \(-10.9\) V at \(\theta = 0^\circ\) to \(5.4\) V at \(\theta = 90^\circ\), followed by a reverse trend within the range of \(90^\circ \leq \theta \leq 180^\circ\). The absolute value of the minimum dc self-bias (at \(\theta = 0^\circ\)) is much larger than that of the maximum self-bias (at \(\theta = 90^\circ\)), which is attributed to the geometrical asymmetry of the discharge. Although two equal-sized electrodes are employed, the discharge is not perfectly geometrically symmetric, due to the capacitive coupling between the oscillating plasma potential and the grounded sidewall. While a Teflon liner is attached to the sidewall of the chamber, its thickness (~3 mm) might be comparable or even smaller than the sheath thickness, resulting in a significant displacement current between the plasma and the sidewall that strongly increases the geometrical asymmetry. Thus, a negative dc self-bias develops under electrically symmetric conditions \((\left| \tilde{\phi}_{\text{max}} \right| = \left| \tilde{\phi}_{\text{min}} \right|)\) at \(\theta = 45^\circ\), in order to compensate electron and ion fluxes to each electrode within one RF period.

It has been found that the dc self-bias shifts towards a more negative value as the geometrical asymmetry increases [21, 31, 77]. Nevertheless, our result indicates that the asymmetry of the discharge can be reduced electrically via the EAE in a VHF driven CCP and an adjustable dc self-bias is achieved. This is crucial for controlling the ion energy at the electrode for surface modifications.

Furthermore, our experiments reveal a dependence of the plasma density at the discharge center on \(\theta\), as shown in figure 2(b). The plasma density is maximum at \(\theta = 0^\circ\) and decreases monotonically with \(\theta\), reaching a minimum at \(\theta = 90^\circ\), a trend that is opposed to the variation of \(\eta\). The period averaged optical emission intensity at the reactor center exhibits a similar dependence as the plasma density on \(\theta\), shown in figure 2(c).

To gain a better understanding of the dependence of the plasma density on \(\theta\), radially resolved measurements of the plasma density have been performed and the results are shown in figure 3. At \(\theta = 0^\circ\), when \(\eta\) is most negative, the plasma density exhibits a center-high distribution, characterized by a central narrow peak superimposed on a much broader peak and an abrupt drop at the electrode edge. Similar to previous observations [68], such a plasma density profile indicates the importance of the higher harmonics, which induce short-wavelength standing waves along the sheath-bulk plasma interface, leading to enhanced electron heating at the electrode center. With the increase of \(\theta\), there is an overall decline of the plasma density,
which is attributed to suppressed nonlinear PSR oscillations at a higher $\theta$, as will be discussed in more detail later. The radial profile of the plasma density becomes relatively uniform at $\theta = 45^\circ$, and evolves into an edge-high distribution at $\theta = 90^\circ$, when $\eta$ is most positive. This result qualitatively agrees with previous experimental results in reference [72], where two equal-sized square electrodes and two much higher driving frequencies of 40.68 MHz and 81.36 MHz were used. Overall, our results demonstrate that the radial uniformity of the plasma density can be controlled by tuning the phase shift between these two consecutive harmonics. The EAE, thereby, provides an opportunity to optimize the plasma uniformity without changing the reactor configuration.

To shed light on the effects of higher harmonics on the plasma uniformity, we measured the magnetic field at the electrode edge ($r = 15$ cm) and close to the electrode center ($r = 4$ cm) for various $\theta$ ($0^\circ$, $45^\circ$, and $90^\circ$), with their time-domain signals shown in the first row of figure 4. The second and third rows in figure 4 show the FFT spectra of the magnetic field signals, which allow us to capture more details about the higher harmonics. For all phases, one can identify distinct differences of the magnetic field waveforms at the electrode center and at the edge (figures 4(a)–(c)). At $\theta = 0^\circ$, the magnetic field close to the electrode center (figures 4(d)–(f)) exhibits more prominent high-frequency oscillations compared to the electrode edge (figures 4(g)–(i)) due to the nonlinear standing wave excitation. With the increase of $\theta$, however, these high-order harmonics close to the electrode center tend to be damped, as shown in figures 4(d)–(f).

The nonlinear standing wave excitation originates from the nonlinear coupling between the PSR and the SWR [69–71]. During the expansion and collapse of a sheath, the PSR frequency $\omega_{\text{PSR}} = (s/l) \omega_p$ sweeps across multiples of the fundamental frequency $\omega$, thus exciting a series of higher harmonics, $\omega_{\text{PSR}} = N \omega$, where $s$ is the sheath thickness, $l$ is the length of the plasma bulk, and $\omega_p$ is the electron plasma frequency. In parallel, as the half-wavelength of the electromagnetic surface wave in a plasma, $\lambda_p/2 = \pi/k$, becomes comparable to or smaller than the electrode diameter, $2R$, the SWR frequency $\omega_{\text{SWR}} = (s/l) \chi_0 c/R$ will also sweep across multiples of the fundamental frequency $\omega$, i.e., $\omega_{\text{SWR}} = M \omega$, where $k$ is the wave number of radially propagating surface waves, $\chi_0$ is the $m$th zero of the zero-order Bessel function of the first kind, $J_0(\chi)$, and $c$ is the speed of light in vacuum. Typically, a strong coupling between the PSR and the SWR can be observed at $N = M$ [71].

In a single-frequency excited, highly geometrically asymmetric discharge, there will be a larger voltage drop across the powered electrode sheath, where a much stronger sheath oscillation could cause a stronger PSR. In this scenario, it is reasonable to adopt a single-sheath model to investigate the nonlinear PSR behavior [69, 71], wherein, the sheath oscillations and all harmonic excitations are expected to be
dominated by the fundamental-frequency voltage at the powered electrode.

In contrast to this, the mechanism causing higher harmonic excitations in dual-frequency discharges is more involved. Particularly, all odd harmonics are excited by the fundamental frequency (30 MHz), whereas the even harmonics can be excited by both the fundamental frequency (30 MHz) and the second harmonic (60 MHz), or even by an intermediate-order harmonic that is excited by the fundamental and/or the second component. For example, both the fundamental frequency and the second harmonic can excite the 4th harmonic of the fundamental, which is combined with the fundamental and second components, giving rise to the excitation of the 8th harmonic. This complex coupling mechanism can lead to a non-monotonic variation in the amplitude of the higher harmonic as a function of $\theta$. As seen from figures 4(d)–(f), while most of the higher harmonics are damped with increasing $\theta$ from $0^\circ$ to $45^\circ$, an abnormal increase in the amplitude of harmonic magnetic field can be identified for $n = 2$ and $n = 4$. Furthermore, the suppression of the higher harmonics exhibits a nonsynchronous behavior with $\theta$ depending on the harmonic order $n$. Although there is a continuous suppression of the high-order ($n = 5–8$) harmonics as $\theta$ increases, a remarkable suppression of the low-order ($n = 3–4$) harmonics is only observed at $\theta > 45^\circ$, for which all high-order ($n = 5–8$) harmonics tend to be vanished almost completely. Altogether, it is clear that the excitation of higher harmonics can be controlled electrically by tuning $\theta$, via the EAE, since this relative phase shift controls the reactor symmetry and, therefore, the self-excitation of the PSR as well as the higher harmonics.

Figures 5(a)–(c) shows the radial profiles of the calibrated amplitude of the harmonic magnetic field $B_{\phi,n}$ ($n = 1–8$) at three typical values of $\theta$. At $\theta = 0^\circ$, for the fundamental frequency, $B_{\phi,1}$ is minimum at the center ($r = 0$) and increases almost linearly towards the edge ($r = 15$ cm). A slight decrease in $B_{\phi,1}$ versus radius can be observed around the electrode edge due to a rapid decline of the current density there. The growth rate of $B_{\phi,2}$ versus radius slightly declines with radius. For higher orders ($n = 3–8$), the maximum of $B_{\phi,n}$ appears inside the electrode region, with its position shifting towards the center with increasing $n$. The radial maxima of $B_{\phi,n}$ ($n = 1–8$), shown in figure 5(a), highlight the importance of the 5th and 6th harmonics, which are considerable as compared to the fundamental or the second component.

The corresponding radial profiles of the harmonic current density, $J_{z,n}$, derived from $B_{\phi,n}$, are illustrated in figures 5(d)–(f). One can observe a relatively uniform distribution for the first two harmonics. For $n = 3–5$, $J_{z,n}$ first experiences a slow decline with radius, followed by a moderate rise, going through a valley which corresponds to the first node of a standing wave current. In fact, there is an abrupt jump in the phase of the harmonic current in the vicinity of the node (not shown here), corresponding to a current reversal. For $n = 6–8$, a second node appears within the electrode radius, with the current phase reversing twice as a function of the radius. The node position gradually shifts towards the electrode center as $n$ increases, due to a shrinking wavelength at a higher harmonic. It is worth noting that for higher harmonic currents, the central peak is much higher than the second or the third peak, reasonably explaining the center-high plasma density at $\theta = 0^\circ$ (see figure 3). Consistent with $B_{\phi,n}$ (figures 5(a)–(c)), $J_{z,n}$ over the whole electrode region experiences a distinct decline as $\theta$ increases (figures 5(d)–(f)), giving a qualitative interpretation of the decline of the plasma density as a function of $\theta$ (figure 3). In particular, with the increase
of \( \theta \), the central peak of the plasma radial profile in figure 3 becomes less evident, which is attributed to the suppression of the higher harmonics (particularly of the 5th and 6th harmonics). The results presented above demonstrate the important role of higher harmonics, which produce peaks of the harmonic current densities at the electrode center, leading to significant plasma nonuniformities. It is, therefore, feasible to improve the plasma uniformity by manipulating the higher harmonic excitations via the EAE, by controlling the discharge symmetry and, thus, the self-excitation of higher harmonics via the PSR.

Based on the radial profiles of \( J_{z,n} \), the harmonic current, \( I_n \), flowing axially across the electrodes, can be estimated by integrating \( J_{z,n} \) over the whole electrode area. \( I_n = 2\pi \int_0^L J_{z,n}(r) dr \). For the fundamental and the second components, we obtain \( I_1 = 0.85 \) A and \( I_2 = 0.78 \) A, which are smaller than the values, \( I_1 = 0.96 \) A and \( I_2 = 2.58 \) A, measured by a current monitor (Pearson 6585) mounted at the RF feeding point at the rear of the powered electrode. The discrepancy is reasonably acceptable since the harmonic current measured by the current monitor includes not only the interelectrode current but also the shunted displacement/conduction current due to the parasitic capacitance/discharge between the powered electrode and the grounded sidewall. Note that the proportion of the latter will become more significant for higher harmonics.

Since the nonlinear sheath oscillation is an inherent origin of the excitation of higher harmonics, the suppression of the harmonic excitations with \( \theta \) can be explained by the dependence of the nonlinear PSR oscillations on the discharge symmetry. At \( \theta = 0^\circ \), a strong asymmetry of the applied voltage waveform, \(|\phi_{\max}| > |\phi_{\min}|\), produces a strong electrical asymmetry, which is enhanced by the geometrical asymmetry of the plasma reactor, giving rise to the most negative dc self-bias (figure 2(a)) and a strong deviation of the symmetry parameter from unity (\( \varepsilon < 1 \)). Such a strongly negative dc self-bias corresponds to a much larger time-averaged voltage drop across the powered electrode sheath compared to the grounded one. As a consequence of the strong asymmetry (\( \varepsilon < 1 \)), the sheath oscillations are maximized at the powered electrode, while they are minimized at the grounded side, thus inducing most significant PSR excitations, and consequently, the strongest harmonic and nonlinear standing wave excitations in the plasma.

With the increase of \( \theta \), the asymmetry of the applied waveform varies from \(|\phi_{\max}| > |\phi_{\min}| to |\phi_{\max}| < |\phi_{\min}|\), which suggests that the geometrical asymmetry is gradually compensated by the electrical asymmetry, resulting in a transition of the dc self-bias from negative to positive and a more symmetric discharge, i.e., \( \varepsilon \) approaches to unity. A positive dc self-bias occurring at \( \theta = 90^\circ \) implies that the time-averaged sheath voltage drop adjacent to the grounded electrode exceeds the one adjacent to the powered electrode. However, due to the strong geometrical asymmetry, i.e., \( A_g \ll A_p \), the symmetry parameter \( \varepsilon \) is close to one, but still less than unity. Therefore, the PSR oscillations and the higher harmonic excitations are still dominated by the powered electrode sheath. This can be confirmed by examining the spatiotemporal distributions of the electron-impact excitation rate at three typical values of \( \theta \) shown in figure 6. We can see that for all \( \theta \) the electron-impact excitation rate exhibits a similar asymmetric structure, i.e., the value adjacent to the powered (bottom) electrode is stronger than that adjacent to the grounded (top) electrode, indicating a stronger sheath oscillation adjacent to the powered electrode. At lower pressures, the PSR oscillations will lead to a rapid sheath expansion, with a speed much faster than that caused by the fundamental frequency, during which multiple energetic electron beams are generated and penetrate into the plasma bulk, causing enhanced excitation and ionization [17]. The self-excitation of the PSR can be initiated at the time of sheath collapse due to the sudden change in the sign of the current [6, 18]. Due to a relatively long lifetime (22.4 ns) of the excited state Ar 2p1, a fine electron-impact excitation structure caused by the multiple electron beams cannot be identified in figure 6. By increasing \( \theta \) from 0° to 90°, the time-averaged voltage drop and the corresponding PSR oscillations at the powered electrode are gradually weakened and, thereby, the nonlinear standing waves associated with higher harmonics excitations are suppressed, resulting in a decline in the plasma density and a transition of the plasma density profile from a ‘convex’ shape to a ‘concave’ shape (figure 3).
While the EAE concept provides an efficient way to suppress the nonlinear standing wave excitation and optimize the plasma uniformity in the frame of the current study, this method is expected to be valid only under specific discharge conditions. First, a relatively low pressure is required, since at higher pressures the PSR oscillations and the resulting self-excitations of higher harmonics tend to be highly damped so that the nonlinear standing wave excitation should be negligible [71]. Second, since the EAE method acts on higher harmonics only, it can only be applied to CCP reactors where the half-wavelength of the fundamental driving frequency in the plasma is much larger than the electrode diameter, i.e., the fundamental-frequency standing wave should not cause remarkable lateral nonuniformities. In such cases, alternative methods such as using special electrodes [48–50] should be employed.

4. Conclusions

Experimental investigations on the dependence of the nonlinear standing wave excitation upon the EAE have been performed in a geometrically asymmetric, capacitive argon discharge driven by two consecutive harmonics (30 MHz and 60 MHz) with an adjustable phase shift, \( \theta \). To bring the effects of nonlinear harmonic excitations on the plasma uniformities into clearer focus, we employed a hairpin probe to determine the radial distribution of the electron density in combination with a high-frequency \( B \)-dot probe to measure the radial distribution of the harmonic magnetic field. With the measured harmonic magnetic field, the radial distribution of the harmonic current density was derived. Moreover, phase resolved optical emission spectroscopy was employed to determine the spatiotemporal distribution of the electron-impact excitation rate, which provided information on the discharge asymmetry and the PSR behavior at different \( \theta \).

It was found that the geometrical asymmetry of the discharge can be compensated electrically via the EAE (i.e. by tuning \( \theta \)) and, thereby, the dc self-bias, \( \eta \), and the plasma symmetry can be adjusted accordingly. Besides, our experiment revealed an evident dependence of the plasma density on \( \theta \). At \( \theta = 0^\circ \), when \( \eta \) is most negative and the discharge asymmetry is strongest, the plasma density exhibits a center-high distribution due to a strong self-excitation of the PSR and due to the presence of strong nonlinear standing wave excitation. With the increase of \( \theta \) the discharge becomes more symmetric and, thus, the PSR is self-excited less strongly. Consequently, the nonlinear standing wave excitation is suppressed and, while the plasma density decreases, its radial profile is much more homogeneous at \( \theta = 45^\circ \). At \( \theta = 90^\circ \) an edge-high distribution is even observed, when \( \eta \) is most positive. Overall, we find that the radial uniformity of the plasma density can be controlled and improved via the EAE.

The suppression of the harmonic excitations by tuning \( \theta \) can be explained by the dependence of the nonlinear PSR oscillations on the discharge symmetry. At \( \theta = 0^\circ \), the geometrical asymmetry is enhanced by the electrical asymmetry, giving rise to a strong asymmetry of the discharge (\( \epsilon \ll 1 \)). Such an asymmetry causes stronger PSR oscillations leading to significant nonlinear standing wave excitation in the plasma. With the increase of \( \theta \), the asymmetry of the discharge is reduced gradually. However, due to the geometrical asymmetry effect, the symmetry parameter remains to be below unity and PSR oscillations are always present, but their magnitude can be adjusted by phase control. Thus, most electron power absorption is always observed adjacent to the powered electrode. As the discharge symmetry is improved by phase control and the PSR self-excitation is attenuated, the harmonic magnetic field declines as a function of \( \theta \), the excitation of non-linear standing waves is reduced and the lateral uniformity of the plasma is enhanced. In conclusion, our results demonstrate that the nonlinear standing waves excited by the PSR oscillations can be switched on and off by tuning \( \theta \), so that the plasma uniformity can be improved. The EAE concept provides the opportunity to control the plasma uniformity without changing the reactor configuration, but by modifying only the external RF power supply system. Compared to existing and technically involving concepts to prevent plasma nonuniformities it should, therefore, be useful for the optimization of etching and thin deposition processes in the semiconductor industry.

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ORCID iDs

Kai Zhao https://orcid.org/0000-0002-9540-4643
Yong-Xin Liu https://orcid.org/0000-0002-6506-7148
Yu-Ru Zhang https://orcid.org/0000-0001-9863-2417
Julian Schulze https://orcid.org/0000-0001-7929-5734
Yuan-Hong Song https://orcid.org/0000-0001-5712-9241
You-Nian Wang https://orcid.org/0000-0003-4251-2453

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