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NUMERICAL STUDY ON ION ENERGY CONTROL IN A SEPARATE-ELECTRODE DF-CCP DISCHARGE

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Abstract. The effect of driving dual frequencies in a separate-Electrode dual-frequency capacitively coupled argon plasma discharge is investigated using a Particle-in-Cell with Monte Carlo Collisions (PIC-MCC) model. The frequency (13.56 MHz) is applied on one electrode, while the opposite electrode is powered by a variable frequency ranging from 27.12 MHz to 67.8 MHz. Our numerical results demonstrate that the combined electrical and geometric asymmetry effects enable quasi-independent control of ion bombardment energies at each electrode. Specifically, increasing the high frequency on the large-area electrode leads to a marked rise in the ion energy at the opposite, low-frequency powered small electrode, even as the DC self-bias voltage decreases. Furthermore, we find that the high-frequency powered electrode consistently sustains the highest ion energies, irrespective of its surface area.

Keywords: Capacitively coupled Plasma discharge, electrical asymmetry effect, Particle-in-Cell.

1. Introduction

Dual-frequency capacitively coupled plasma systems (DF-CCP) have established themselves as an essential technology in plasma processing, offering greater control and flexibility in a variety of applications [1]. These systems use two separate radio-frequency (RF) energy sources operating at different frequencies, typically low frequency (LF) and high frequency (HF), to generate and maintain the plasma. The dual-frequency approach allows independent control of ion energy and flux, enabling precise manipulation of plasma characteristics. This unique capability makes DF-CCP systems particularly useful in semiconductor manufacturing processes, such as etching and deposition, where precise control of plasma parameters is essential to achieve the desired material properties and dimensions. In capacitively coupled plasma (CCP) discharges, the plasma potential is typically lower than in other plasma sources such as inductively coupled or microwave plasmas. This relatively low plasma potential reduces the kinetic energy of ions impinging on the substrate, thereby minimizing surface damage and uncontrolled sputtering, and enabling more controlled material interactions [2].

The motivations for employing asymmetric geometry in radio-frequency capacitively coupled plasma (RF-CCP) arise from the advantages it offers, including control over ion energy distribution and flux for specific applications. It is to address a major challenge in etching and deposition processes that dual-frequency capacitively coupled plasma (DF-CCP) discharges have emerged, providing an approach for independently controlling ion flux and ion bombardment energy on substrates. Traditionally, this control relies on two main asymmetry mechanisms: geometric asymmetry [3], where the difference in electrode areas influences potential distribution, and electrical asymmetry [4], generated by applying multiple frequencies.

This paper presents two primary objectives. The first investigates the Electrical Asymmetry Effect (EAE) generated by the coupling of a fundamental frequency RF voltage waveform with its harmonics (n=2-5) in a Capacitively Coupled Plasma (CCP) discharge. This study specifically examines the application of distinct RF voltage waveforms to each electrode and analyzes how varying the high frequency waveform on one electrode affects the ion bombardment energy on the other, fixed low-frequency electrode. The second objective evaluates the influence of reactor geometry on the overall behavior of the DF-CCP discharge, exploring its interaction with the applied frequencies.

2. Physical Model

In this section, we briefly describe the electrical asymmetry effect (EAE), focusing on waveforms that include even or odd harmonics of the fundamental frequency and their role in the formation of a DC self-bias.

The discharge operates between two non-identical, parallel plate electrodes, as presented in figure 1. The driving voltage waveforms on the powered electrodes used in this work possess an identical voltage amplitude V_0 and are defined by :

$$V_l(t) = V_0 e^{i\omega t} \quad \text{and} \quad V_r(t) = V_0 e^{in\omega t} \tag{1}$$

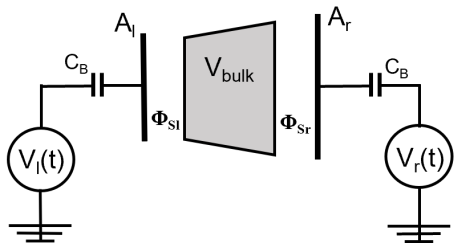

Figure 1. Schematic of the simulated DF-CCP discharge.

In the DF-CCP operating at low pressure, the sheath thickness is large making the sheath capacitance substantially smaller than C_B ($C_{\text{sheath}} \ll C_B$). As a result, this large disparity in impedance justifies the common modeling assumption that, under low-pressure conditions, the blocking capacitors can be treated as effective short circuits for the RF signal. Such an approximation simplifies circuit analysis without compromising accuracy in the determination of whether even or odd harmonics in the applied signal are responsible for the electrical asymmetry. Nevertheless, when C_B is relatively small, a non-negligible fraction of the RF voltage can still appear across the blocking capacitors, which may become relevant in precision voltage balance analyses [5], particularly in low-pressure electropositive discharges where neglecting V_{bulk} remains a realistic assumption [6].

Then the effective voltage across the plasma, $V(t)$, is the difference between the sheath potential drops, ϕ_{Sl} and ϕ_{Sr} :

$$V(t) = V_r(t) - V_l(t) \approx \phi_{Sr}(t) + \phi_{Sl}(t) \qquad (2)$$

If the sheaths are to be electrically symmetric, it is obvious that their individual voltage waveforms will exhibit a half-period symmetry, i.e., $V(t) = -V\left(t + \frac{T}{2}\right)$. As a result, the polarity of one of the voltages needs to be reversed.

$$V_0[e^{in\omega t} - e^{i\omega t}] = -V_0[e^{in(\omega t+\pi)} - e^{i(\omega t+\pi)}] \qquad (3)$$

The relation 3 is valid only when n is odd ($e^{in\pi} = -1$). This implies that no DC self-bias is generated by the waveform shape, and the sheaths remain temporally symmetric.

In a Capacitively Coupled Plasma (CCP) discharge, the development of a DC self-bias (η) is directly related to the need for current balance at the electrodes. At two distinct times within the RF period, one sheath voltage reaches its maximum $\phi_{\max}$ and the other its minimum $\phi_{\min}$. The DC self-bias arises from this voltage difference [4]:

$$\eta = -\frac{\phi_{\max} + \varepsilon\phi_{\min}}{1+\varepsilon} \qquad (4)$$

Where ε is the symmetry parameter given by the maximum sheaths voltage ratio:

$$\varepsilon = \left|\frac{\hat{\phi}_{Sl}}{\hat{\phi}_{Sr}}\right| \qquad (5)$$

Geometric asymmetry is a a fundamental mechanism in the formation of DC self-bias. In CCPs, if the electrodes possess unequal surface areas, a natural DC self-bias develops [1, 6]. This basic principle of maintaining current balance across plasma-electrode interfaces remains fundamental. However, in an asymmetric separate-electrode Dual-Frequency Capacitively Coupled Plasma, the interaction of the two applied frequencies with distinct power delivery to each electrode introduces significant modifications to this classical behavior. While geometric asymmetry remains a fundamental characteristic of the reactor, the specific electrical conditions can intricately influence, or even fundamentally reverse, the classically expected effect of geometric asymmetry on individual sheath voltage drops. With two independent power supplies, an overall DC self-bias (η) will form, whose sign and amplitude will be a complex combination of the geometric and electrical asymmetry induced by the two frequencies and their independent coupling to the plasma. Nevertheless, from the numerical results obtained, the DC self-bias can be evaluated using the following relation:

$$\eta = |\phi_{Sr}| - |\phi_{Sl}| \qquad (6)$$

3. Simulation

The discharge operates between two non-identical, parallel plate electrodes, as presented in figure 1. The left electrode, A_l, is positioned at $x = 0$ cm, and the right electrode, A_r, at $x = 3$ cm; both are powered. Their respective surface areas are 10 cm^2 and 16 cm^2. The plasma is generated in a 20 mTorr pure argon gas, with a neutral gas temperature of 0.03 eV. To model the discharge we use 'XPDP2,' a bounded electrostatic Particle-in-Cell Monte Carlo Collisions (PIC-MCC) code, originally developed by Verboncoeur et al [7] and Vahedi et al [8]. The code simulates the collisional processes of an argon discharge using a Monte Carlo routine. These processes include electronic excitation, ionization, and elastic scattering for electron-atom collisions, as well as charge exchange and isotropic scattering for ion-atom collisions. The computational domain is spatially discretized using 180×600 cells, ensuring that the distance between successive nodes remains below $0.8\,\lambda_D$. Macro-particle statistical weights are adjusted to assure over 80 macro-particles per cell within the plasma bulk. The simulation time step was set to $dt = 3.6\ 10^{-11}$ s, corresponding to ~ 1000 times smaller than the RF period at 27.12 MHz and ~ 400 times at 67 MHz.

Under the low-pressure conditions considered, where electron-neutral collision times far exceed both the RF and plasma periods, this temporal resolution ensures accurate resolution of RF modulation. Gas density is considered uniform throughout the domain and is fixed based on the operating pressure and ambient temperature. Charged particle densities are determined by integrating the self-consistently simulated

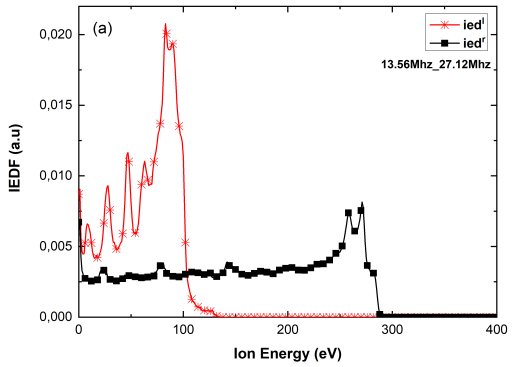

Figure 2. Time averaged Ion Energy Distribution Function (IEDF) at 13.56 MHz and its second harmonic.

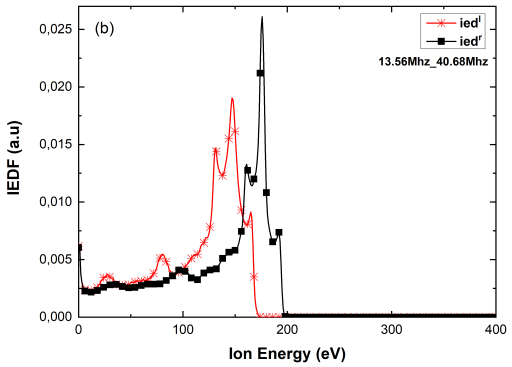

Figure 3. Time averaged Ion Energy Distribution Function (IEDF) at 13.56 MHz and its third harmonic.

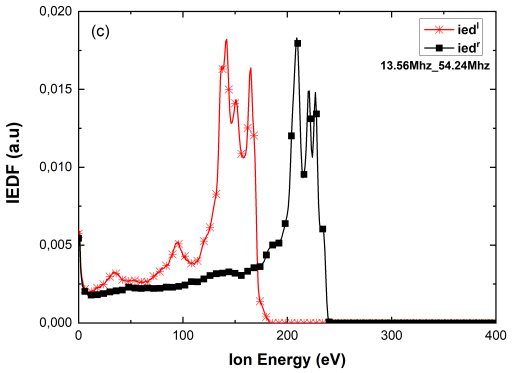

Figure 4. Time averaged Ion Energy Distribution Function (IEDF) at 13.56MHz and its fourth harmonic.

ion and electron trajectories, which are computed via the PIC/MCC method. Both ions and electrons are tracked from their point of creation through ionization until they reach any boundary of the simulation volume. It is assumed that complete neutralization of the charged particle flux occurs upon reaching these surfaces. The plasma potential is derived by numerically solving Poisson's equation, utilizing Dirichlet boundary conditions. Electrode ion bombardment induces an emission of secondary electron. To simulate this flux of electrons, we consider a secondary electrons emission coefficient of $\gamma = 0.2$. Nevertheless, the impact of the γ value on the discharge simulation proves to be negligible because the majority of power delivered to the plasma is dissipated through ohmic and stochastic heating. These latter heating mechanisms are self-consistently simulated in the PIC/MCC method. All results presented in this paper are averaged over one RF period after the simulation reaches the steady state.

4. Results

Asymmetric discharges, which can result from geometric asymmetry [1, 9], electrical asymmetry [10, 11], or a combination of the two [10], inherently lead to the generation of a DC self-bias. This work investigates the Electrical Asymmetry Effect (EAE) in a geometrically asymmetric Separate-Electrode Dual-Frequency Capacitively Coupled Plasma (DF-CCP) discharge, which is composed of a fundamental frequency (13.56 MHz) and its nth harmonic.

In the Figures 2 and 4, we can see that when a fundamental frequency is combined with an even harmonic, the resulting voltage waveform is temporally asymmetric, leading to the formation of a DC self-bias voltage across the electrodes. This self-bias arises from the combined electrical and geometrical asymmetry. In these two figures, we should expect a rather significant DC self-bias. However, the higher the harmonic frequency, the less significant the DC self-bias becomes. Comparing Figure 2 (13.56 – 27.12 MHz) to Figure 4 (13.56 – 54.24 MHz), as the high frequency increases; The ion energy on the 13.56 MHz-left-powered electrode (ied^l) increases significantly (peaks shift from ∼90 eV to ∼150 eV). Conversely, the ion energy on the high-frequency-right-powered electrode (ied^r) decreases (peaks shift from ∼270 eV to ∼220 eV). This trend concerning the ion energy reaching the two electrodes shows that the choice of the high frequency enables a certain control over the ion bombardment on the surface of both electrodes, allowing for tailored ion bombardment for specific processing needs. Furthermore, the increase of the harmonic frequency on the right electrode (i.e. 54.24 MHz) leads to a denser and more intensely heated electrons via stochastic and collisional heating mechanisms. This significant modification of the plasma leads to thinner plasma sheaths on both electrodes (see Figure 6) and has two major effects: First, it shifts the charge balance such that a less negative DC self-bias voltage η is required. Second, it increases the average plasma bulk (See Figure 7). This is because, to effectively confine the more mobile electrons, the averaged plasma potential must become more positive. It is this increase in the average bulk that, even with a less negative DC Self-bias

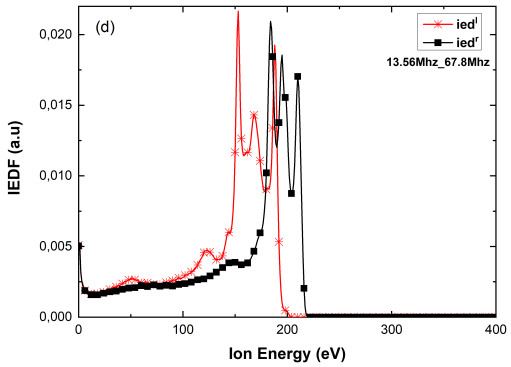

Figure 5. Time averaged Ion Energy Distribution Function (IEDF) at 13.56MHz and its fifth harmonic.

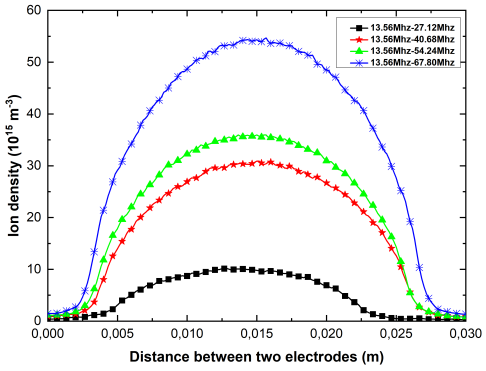

Figure 6. Spatial distribution of the ion density for different dual-frequency combinations

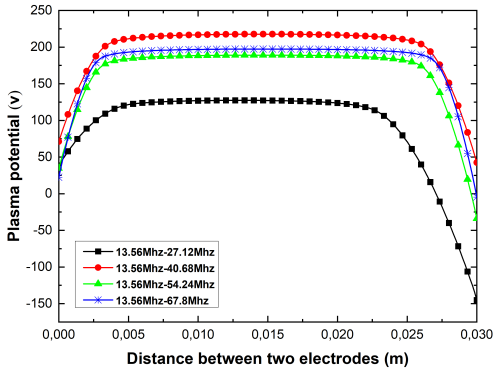

Figure 7. Average spatial distribution of the plasma potential for different dual-frequency combinations.

η, creates a larger average potential drop across the sheath of the left electrode (ϕ_{Sl}), resulting in higher ion bombardment energy on that electrode. This is a proof of the ability of DF-CCP systems to decouple or, at the very least, intricately control, key plasma parameters, where a modification on one electrode (HF) can influence the ion bombardment energy on the other electrode (LF). When a fundamental frequency is combined with an odd harmonic, the temporal symmetry of the voltage waveform is preserved, meaning the positive and negative half-cycles remain inverses of each other assuming no other asymmetries. Therefore, a DC self-bias is not generated by the tailored dual-frequency voltage waveform in such combinations, but rather by the inherent geometric asymmetry (as see in Figure 3 and 5) leading to distinct ion energy distributions on the two electrodes. This clear difference in ion bombardment energy between the two electrodes, despite the temporally symmetric voltage waveform, unequivocally demonstrates the influence of the geometric asymmetry in shaping the DC self-bias and thus controlling the ion energy distribution in such a dual-frequency system driven by an odd harmonic.

The observed increase in ion bombardment energies on both electrodes, as evident when comparing Figure 3 (13.56 – 40.68 MHz) to Figure 5 (13.56 – 67.8 MHz), is consistent with a tendency where the DC self-bias value decreases. The DC self-bias (η) is a complex function of the sheath voltage drops at both electrodes. In this separate-electrode configuration, each sheath responds primarily to its own applied frequency. Even if the DC self-bias becomes less negative, the average plasma potential may increase and still lead to larger potential drops across individual sheaths. Specifically, the time-averaged potential drop across the sheath of each driven electrode can increase in magnitude, thereby accelerating ions to higher energies. This happens because the overall potential shifts upwards with the increased plasma potential, compensating for the less negative global bias and ensuring efficient electron confinement, which ultimately results in higher ion bombardment energies.

Figure 7 provides quantitative insight into the DC self-bias. All potentials reported are time-averaged values. For the 13.56–40.68 MHz case the time-averaged potentials are approximately +70 V at the left electrode and +42 V at the right electrode, yielding a DC self-bias $\eta \approx -28$ V, defined here by using the equation 6. For the 13.56–67.8 MHz case the left and right electrode averages are approximatively +23 V and -5 V, respectively, giving the same $\eta \approx -28$ V. Although the self-bias remains almost unchanged between these odd-harmonic cases, the time-averaged sheath potential drops increase noticeably when the higher HF component is raised from 40.68 to 67.8 MHz. Specifically, the left powered electrode sheath drop increases from $\sim$ 145 V to $\sim$ 172 V, while the right powered electrode sheath increases from $\sim$ 173 V to $\sim$ 200 V. Because the mean ion impact energy is set primarily by the time-averaged sheath drop, these larger drops quantitatively explain the observed rise in ion bombardment energies in Figures 3 and 5. This behavior also underlines that geometric asymmetry, even in the absence of even harmonic waveform shaping, modifies the partitioning of voltage between the sheaths in

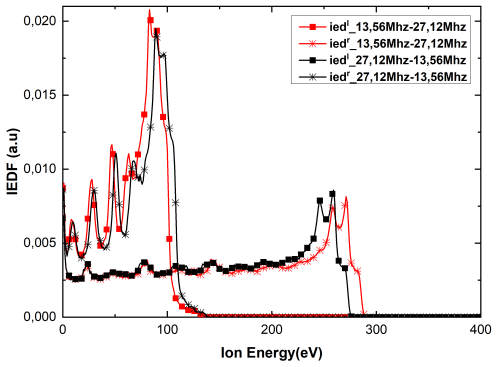

Figure 8. Time averaged Ion Energy Distribution Function (IEDF) for reversed dual-frequency combinations.

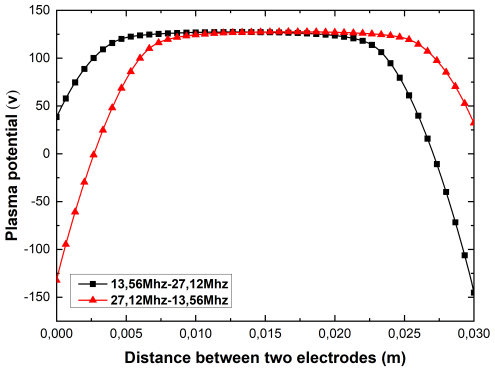

Figure 9. Average spatial distribution of the plasma potential between the two electrodes for reversed dual-frequency combinations (13.56–27.12 MHz and 27.12–13.56 MHz)

separate-electrode DF-CCP systems. The figure 8 displays the IEDFs for two frequency configurations. The first configuration is 13.56 MHz – 27.12 MHz: The low frequency (13.56 MHz) is applied to the left electrode (IED^l), and the high frequency (27.12 MHz) is applied to the right electrode (IED^r), The second configuration is 27.12 MHz - 13.56 MHz: The high frequency (27.12 MHz) is applied to the left electrode (IED^l), and the low frequency (13.56 MHz) is applied to the right electrode (IED^r). We can see that, in both cases, the electrode powered by the high frequency, regardless of its size, produces significantly higher ion energy. In the first configuration, the IED^r shows a maximum energy of around 260 eV, while the IED^l has a maximum energy of around 85 eV. When the frequencies are inverted, the roles are also reversed: the IED^l now has a maximum energy of around 275 eV, and the IED^r has a maximum energy of around 90 eV. Inverting the frequencies thus effectively inverts the electrical asymmetry effect, which controls the ion bombardment energy. We also note that the surface area of the electrode where the frequency is applied plays a significant role in the electrical asymmetry, as can be seen by comparing the DC Self-bias of each configuration.

The figure 9 confirms that the influence of the applied frequency and the surface electrode area are both critical to control the energy in plasma discharges. As demonstrated by the frequency inversion numerical experiment, the ion energy distribution is primarily dictated by the frequency applied to the electrode. For instance, the high-frequency (HF) powered electrode consistently experiences the highest ion bombardment energy, irrespective of whether it is the smaller or larger electrode. This clarifies that in this system, the electrical asymmetry introduced by the frequencies is the dominant factor controlling ion energy. While the frequency is the main determinant of the overall ion energy level, the electrode surface area still plays a significant modifying role. A comparison of IEDFs for the same frequency on different electrode areas reveals subtle but measurable differences in the shape and fine structure of the distributions.

5. Conclusions

The comprehensive comparison between odd and even harmonics in this geometrically asymmetric, separate-electrode DF-CCP highlights the distinct mechanisms governing ion energy control. While geometric asymmetry consistently induces a fundamental DC self-bias (η), the use of even harmonics provides an additional tool via the Electrical Asymmetry Effect (EAE). This effect allows for independent manipulation of ion bombardment energies on each electrode, offering enhanced control over processing conditions beyond what geometric asymmetry alone can provide. The interaction of these effects, along with the varying average plasma potential across different high frequencies, contributes to the complex and controllable ion dynamics observed. Our frequency inversion numerical experiment showed that the high-frequency (HF) powered electrode consistently develops the highest ion energy, regardless of its surface area. Although the EAE is dominant, geometric asymmetry is not without effect. We observed that the IEDF of a given frequency is not strictly identical when applied to electrodes of different surface areas. Geometric asymmetry acts as a modifying factor that adjusts the shape and fine structure of the ion energy distribution. Our results highlight that the behavior of the plasma is not governed by a single factor, but by a complex interaction between the EAE and geometric asymmetry. The EAE defines the overall energy scale, while geometric asymmetry fine-tunes the ion energy distribution. This interaction is a crucial aspect of DF-CCP systems with separately powered electrodes.

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