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The electrical asymmetry effect in multi-frequency capacitively coupled radio frequency discharges

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Abstract

The electrical asymmetry effect (EAE) in geometrically symmetric capacitively coupled radio frequency discharges operated at multiple consecutive harmonics is investigated by a particle-in-cell (PIC) simulation and an analytical model. The model is based on the original EAE model, which is extended by taking into account the floating potentials, the voltage drop across the plasma bulk, and the symmetry parameter resulting from the PIC simulation. Compared with electrically asymmetric dual-frequency discharges we find that (i) a significantly stronger dc self-bias can be generated electrically and that (ii) the mean ion energies at the electrodes can be controlled separately from the ion flux over a broader range by tuning the phase shifts between the individual voltage harmonics. A recipe for the optimization of the applied voltage waveform to generate the strongest possible dc self-bias electrically and to obtain maximum control of the ion energy via the EAE is presented.

(Some figures in this article are in colour only in the electronic version)

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<td>$L$</td>
<td>electrode gap</td>
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<tr>
<td>$s$</td>
<td>sheath width</td>
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<tr>
<td>$f = 1/T_{\text{RF}}$</td>
<td>fundamental driving frequency</td>
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<tr>
<td>⟨..⟩</td>
<td>time average over $T_{\text{RF}}$</td>
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<td>$k$</td>
<td>total number of applied consecutive harmonics</td>
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<tr>
<td>$n$</td>
<td>index for the applied consecutive voltage harmonics</td>
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<td>$\tilde{\phi}(t)$</td>
<td>driving voltage waveform (sum of $k$ consecutive harmonics)</td>
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<td>$\tilde{\phi}_n(t)$</td>
<td>amplitude of the $n$th of $k$ consecutive driving harmonics</td>
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<td>$\tilde{\phi}_n^{(k)}(t)$</td>
<td>$\tilde{\phi}(t)/\sum_{n=1}^{k} \tilde{\phi}_n^{(k)}$ (normalized driving voltage waveform)</td>
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<td>$\theta_n$</td>
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<td>global maximum, minimum of $\tilde{\phi}(t)$</td>
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<td>$\phi_{\text{sp,sg}}$</td>
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<td>maximum sheath voltage at the powered, grounded electrode</td>
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<td>minimum, maximum dc self-bias</td>
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<td>$\varepsilon$</td>
<td>symmetry parameter</td>
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<td>$A_{p.g}$</td>
<td>surface area of the powered, grounded electrode</td>
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<tr>
<td>$n_{i,e}$</td>
<td>ion, electron density</td>
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1. Introduction

Capacitively coupled radio frequency (CCRF) discharges play a major role in various applications such as semiconductor manufacturing and the production of solar cells. In the context of etching and deposition processes on microscopic scales the ion energy and flux at the substrate surface are crucial parameters. Separate control of these two parameters at the electrodes is technologically most important [1, 2].

The classical approach for realizing this separate control is the use of dual-frequency (df) discharges operated at substantially different frequencies, e.g. 2 and 27 MHz [3–7]. The quality of the separate control of ion energy and flux in these discharges is, however, limited due to the coupling of both frequencies [8–18] and the effect of secondary electrons [14, 19, 20]. In order to obtain better control of the ion energy distribution function at the substrate surface, triple-frequency CCRF discharges operated at substantially different frequencies [21, 22] and hybrid discharges [23–29] have also been studied.

Recently, the electrical asymmetry effect (EAE) was proposed as a novel approach for realizing this separate control in an almost ideal way in df discharges driven by a fundamental frequency and its second harmonic, i.e. the voltage waveform [8, 30–40]

$$
\tilde{\phi}^{(2)}(t) = \tilde{\phi}_1^{(2)} \cos (2\pi ft + \theta_1) + \tilde{\phi}_2^{(2)} \cos (4\pi ft).
$$

(1)

Here, $\tilde{\phi}_1^{(2)}$ and $\tilde{\phi}_2^{(2)}$ are the amplitudes of the applied fundamental frequency and its second harmonic, respectively. $\theta_1$ is the fixed, but adjustable phase shift between the two driving voltage waveforms. By experiments, simulations, and modeling it has been demonstrated that a variable dc self-bias, $\eta$, can be generated via the EAE by tuning $\theta_1$ from 0° to 90°. $\eta$ was found to change almost linearly as a function of $\theta_1$ for 0° $\leq \theta_1 \leq 90°$ [32–34]. Therefore, the mean ion energy at both electrodes can be controlled separately from the ion flux by tuning $\theta_1$. The optimum choice of voltage amplitudes for the generation of the strongest possible dc self-bias and to obtain maximum control of the ion energy in df discharges was found to be $\tilde{\phi}_1^{(2)}/\tilde{\phi}_2^{(2)} \approx 1/2$ [37].

Until now the investigations of the EAE have been restricted to df discharges driven by a voltage waveform defined by equation (1). In such df discharges the mean ion energy at the electrodes can be changed by a factor of about 2 by tuning $\theta_1$ [32–34]. A broader range of ion energy control (separately from the ion flux) would clearly be beneficial for applications. Thus, a method to maximize the range of ion energy control without affecting the ion flux is required.

Therefore, in this work the investigations of the EAE are extended from dual- to multi-frequency discharges. The EAE in geometrically symmetric discharges driven by multiple consecutive harmonics is investigated by a one dimensional electrostatic particle-in-cell (PIC)-simulation [32] and an extended version of the analytical model of the EAE [31]. It is demonstrated that compared with df discharges (i) a significantly stronger dc self-bias can be generated electrically if the discharge is driven by multiple consecutive harmonics with particularly chosen individual amplitudes and (ii) that the mean ion energies at the electrodes can be controlled separately from the ion flux over a broader range.

The paper is structured in the following way: in section two the basics of the PIC simulation are outlined. In section three the extended version of the analytical model of the EAE is introduced. The results are presented in section four. This section is divided into three parts. First, a recipe for customizing the applied voltage waveform to generate the strongest possible variable dc self-bias electrically in geometrically symmetric multi-frequency discharges is presented based on the EAE model. Second, the electrical generation of a stronger variable bias in such discharges is discussed at different neutral gas pressures based on simulation and model results. Third, the effect of the optimized voltage waveform on the range of ion energy control is outlined. Finally, conclusions are drawn in section five.

2. PIC simulation

The simulations presented here are based on a one dimensional (1d3v) bounded plasma PIC code complemented with a Monte Carlo treatment of collision processes (PIC/MCC). The neutral gas temperature is taken to be $T_g = 400$ K and the electrode gap is $L = 2$ cm for all conditions investigated. The discharge is operated in argon. The cross sections for electron–neutral and ion–neutral collision processes are taken from [41–43]. At the planar, parallel and infinite electrodes, electrons are reflected with a probability of 20% [44]. Secondary electron emission from the electrodes is neglected. Discharges at neutral gas pressures of 5, 10 and 100 Pa are investigated.

A voltage waveform corresponding to the sum of $k$ consecutive harmonics with individual amplitudes of harmonics, $\tilde{\phi}_n^{(k)}$, is applied to one electrode:

$$
\tilde{\phi}^{(k)}(t) = \sum_{n=1}^{k} \tilde{\phi}_n^{(k)} \cos (2\pi nf t + \theta_n),
$$

(2)

where $n$ is an integer number and $f = 13.56$ MHz is the applied fundamental rf. $\theta_n$ is the fixed, but adjustable phase
shift of the \(n\)th harmonic. The dc self-bias, \(\eta\), is determined in an iterative way to ensure that the charged particle fluxes to each of the two electrodes, averaged over one period of the fundamental frequency, balance. Details of the PIC simulation can be found elsewhere [32, 45–47].

3. Model

The original model of the EAE has been designed for df discharges and is described in detail in [31]. It is based on three assumptions: (i) each sheath is assumed to collapse completely at least once per RF period, i.e. the floating potentials are neglected, (ii) the voltage drop across the bulk is neglected and (iii) the maximum charges in both sheaths are assumed to be identical. Here, we extend the original version of the model by dropping these assumptions.

The model is based on the voltage balance in the context of a global model of a CCRF discharge:

\[
\tilde{\phi}^{(k)}(t) + \eta = \phi_{sp}(t) + \phi_{sg}(t) + \phi_b(t). 
\]

Here, \(\phi_{sp}(t)\) and \(\phi_{sg}(t)\) are the time-dependent voltage drops across the sheath adjacent to the powered and grounded electrode, respectively (\(\phi_{sp}(t) < 0, \phi_{sg}(t) > 0 \forall t\)). \(\phi_b(t)\) is the voltage drop across the plasma bulk.

At the times of maximum applied voltage, \(\tilde{\phi}_{sp}^{(k)}\), and minimum applied voltage, \(\tilde{\phi}_{sg}^{(k)}\), within one period of the fundamental applied frequency the voltage balance is

\[
\tilde{\phi}_{sp}^{(k)} + \eta = \phi_{sp}^{(k)} + \phi_{sg}^{(k)} + \phi_b^{(k)} .
\]

\[
\tilde{\phi}_{sg}^{(k)} + \eta = \phi_{sp}^{(k)} + \phi_{sg}^{(k)} - \phi_b^{(k)} .
\]

Here, \(\phi_{sp}^{(k)}\) and \(\phi_{sg}^{(k)}\) are the floating potentials at the powered and grounded electrodes, respectively. \(-\phi_{sp}^{(k)}\) and \(-\phi_{sg}^{(k)}\) are the maximum respective sheath voltages. Generally, the dynamics of both sheaths are 180° out of phase [30], i.e. when the absolute value of one sheath voltage is maximum, the absolute value of the other sheath voltage is minimum. This minimum corresponds to the floating potential. At the times of maximum and minimum applied voltage the bulk voltages differ by their sign, whereas their absolute values are assumed to be identical for symmetry reasons.

By introducing the symmetry parameter \(\varepsilon\) [31],

\[
\varepsilon = \frac{\phi_{sg}}{\phi_{sp}} \approx \left(\frac{A_p}{A_g}\right)^2 \frac{\bar{n}_{sp}}{\bar{n}_{sg}} \left(\frac{Q_{mg}}{Q_{mp}}\right)^2 ,
\]

an analytical expression for \(\eta\) can be found. In equation (6), \(A_p\) and \(A_g\) are the surface areas of the powered and grounded electrodes, respectively, and \(\bar{n}_{sp}\) and \(\bar{n}_{sg}\) are the mean ion densities in the respective sheath. \(Q_{mp}\) and \(Q_{mg}\) are the maximum charges in the sheath at the powered and grounded electrodes, respectively [40]. These maximum charges may be different in the extended model.

Combining equations (4), (5) and (6) yields an analytical expression for the dc self-bias:

\[
\eta = -\frac{\tilde{\phi}_{sp}^{(k)} + \tilde{\phi}_{sg}^{(k)}}{1 + \varepsilon} + \frac{\phi_{sp}^{(k)} + \varepsilon \phi_{sg}^{(k)}}{1 + \varepsilon} + \frac{1 - \varepsilon}{1 + \varepsilon} \phi_b^{(k)}. 
\]

The original version of the EAE model yields only the first term in equation (7) [31]. In the extended version of the model \(\varepsilon\) is taken from the PIC simulation. Thus, assumption (iii) is no longer made. Taking into account the floating potentials (assumption (ii)) yields the second term in equation (7). The third term results from the consideration of the bulk voltage (assumption (ii)). Both terms are also affected by the symmetry parameter and, therefore, by the elimination of assumption (iii).

In the following, \(\varepsilon, \phi_{sp}^{(i)}, \phi_{sg}^{(i)}\) and \(\phi_b\) are taken from the PIC simulation and are used as input parameters for the extended version of the EAE model. The effect of taking into account the floating potentials, the correct symmetry parameter and the bulk voltage on the analytical calculation of the dc self-bias in multi-frequency discharges will be discussed in section 4.2.

4. Results

4.1. A recipe for optimizing the applied voltage waveform for the electrical generation of a variable dc self-bias

At high pressures, typically above 100 Pa in argon (collisional sheaths), and in geometrically symmetric discharges \(A_p = A_g\) the original version of the EAE model, i.e. only the first term of equation (7) with \(\varepsilon = 1\), yields the following expression for the dc self-bias [31]:

\[
\eta = -\frac{\phi_{sp}^{(k)} + \phi_{sg}^{(k)}}{2}.
\]

The effect of the simplifications made in the context of the model’s derivation and the situation at lower pressures will be discussed in detail in section 4.2.

According to equation (8) the dc self-bias depends on the difference between the absolute values of the global extrema of the applied voltage waveform, i.e. the bigger this difference the stronger \(\eta\) will be. A stronger \(\eta\) with maximum \(n_{max}\) and minimum \(n_{min} = -n_{max}\) that can still be tuned from its minimum to its maximum by adjusting the phase shifts between the driving frequencies, \(\theta_{n}\), will lead to a broader range of ion energy control at the electrodes.

Compared with df discharges operated at a fundamental frequency and its second harmonic (equation (1)), the difference between the absolute values of the global extrema of the applied voltage waveform can be increased by driving one electrode with the sum of multiple consecutive harmonics, i.e. a voltage waveform corresponding to a finite Fourier series with components corresponding to \(\phi_{sp}^{(k)}\) (equation (2)). The effect of adding more consecutive harmonics to the applied voltage waveform is shown in figure 1. In this figure the individual amplitudes of harmonics, \(\phi_n^{(k)}\), are chosen to be identical and the voltage waveform is normalized by \(\sum_{n=1}^{N} \phi_n^{(k)}\)

\[
\tilde{\phi}_{N}^{(k)}(t) = \frac{\tilde{\phi}_{sp}^{(k)}(t) + \tilde{\phi}_{sg}^{(k)}(t)}{\sum_{n=1}^{N} \phi_n^{(k)}}.
\]

Note that \(N\) indicates normalized waveforms, while \(n\) is the harmonics index. All phase shifts, \(\theta_n\), are chosen to be \(0^\circ\), which is the phase of strongest dc self-bias. \(k = 2\) corresponds
to the electrically asymmetric df discharges investigated before [8, 30–40]. Obviously, adding more consecutive harmonics increases the difference between the absolute values of the global extrema of $\tilde{\phi}_N^{(k)}(t)$ and, therefore, leads to the generation of a stronger dc self-bias and finally to a broader range of ion energy control (see sections 4.2 and 4.3).

If $k$ consecutive harmonics are used, $\tilde{\phi}_N^{(k)}(t)$ will have $k$ local minima and one global maximum within one period of the fundamental frequency. If identical amplitudes of harmonics and more than 2 consecutive harmonics are used ($k \geq 3$ in figure 1), the value of the minima will not be the same. Similar to the phenomenon of Gibbs ringing in Fourier theory [48, 49], the value of the first and last minimum within one period of the fundamental frequency will be more negative compared with all other minima. This situation is clearly not ideal for the generation of the strongest possible dc self-bias by maximizing the difference between the absolute values of the global extrema of $\tilde{\phi}_N^{(k)}(t)$. A particular choice of amplitudes of harmonics however, will solve this problem. If these are chosen according to the following criterion, the values of all minima of $\tilde{\phi}_N^{(k)}(t)$ will be identical (see appendix A). This criterion for the selection of amplitudes of harmonics is similar to the use of the Lanczos $\sigma$-factor in Fourier theory to reduce Gibbs ringing [50]:

$$\tilde{\phi}_0^{(k)} = \phi_0 \frac{k - n + 1}{k}.$$  \hspace{1cm} (10)

The constant $\phi_0$ determines the ion flux and the absolute values of the mean ion energies at the electrodes. If the individual amplitudes of harmonics are chosen according to this criterion, the $k$ minima of $\tilde{\phi}_N^{(k)}(t)$ will occur at times $t_m$ within one period of the fundamental frequency $f$ with $f t_m = 2\pi m / (k + 1)$ ($m = 1, 2, \ldots, k$, see appendix A). For a normalized voltage waveform the absolute value of the minima equals $k^{-1}$, while the only maximum is 1 independent of $k$ and is always located at $t = 0$ (see appendix A).

As an example, according to equation (10), in a discharge driven by 3 consecutive harmonics the amplitudes normalized by $\phi_0$ of the fundamental, second and third harmonic should be chosen as 3/3, 2/3 and 1/3, respectively. In a previous work the optimum choice of the normalized amplitudes of harmonics in df discharges driven by two consecutive harmonics was found to be 2/2 and 1/2 for the fundamental frequency and the second harmonic, respectively [37]. Generally, the df scenario is a special case of equation (10) ($k = 2$).

Figure 2 shows the effect of this particular choice of the individual amplitudes of harmonics on the applied voltage waveform. Obviously, compared with identical amplitudes of harmonics (see figure 1) the difference between the absolute values of the global extrema of $\tilde{\phi}_N^{(k)}(t)$ is increased and, therefore, a stronger dc self-bias will be generated and the range of ion energy control will be increased (see sections 4.2 and 4.3).

4.2. The effect of the optimized voltage waveform on the electrical generation of a variable dc self-bias

Figure 3 shows the normalized dc self-bias, $\tilde{\eta} = \eta / \sum_{k=1}^{4} \tilde{\phi}_n^{(k)}$, as a function of the number of applied consecutive voltage harmonics, $k$, at $\theta_n = 0^\circ \forall n$ at a high pressure of 100 Pa (collisional sheaths). The discharge is geometrically symmetric and a voltage waveform according to equation (2) with optimized amplitudes of harmonics (equation (10)) is applied to one electrode.

In a single frequency discharge ($k = 1$) no dc self-bias can be generated electrically via the EAE ($\tilde{\phi}_n^{(k_1)} = -\tilde{\phi}_n^{(k_2)}$), $k = 2$ corresponds to the electrically asymmetric df discharge with optimized amplitudes that has been investigated before [8, 30–40]. In agreement with previous results the strongest possible bias at high pressures ($\varepsilon \approx 1$) in df discharges is 25%, i.e. the normalized bias can be tuned from $-25\%$ to $+25\%$ by adjusting the phase shift between the two driving voltages. This allows us to change the mean ion energies separately from the ion flux by a factor of about 2 at the electrodes [32–34]. According to figure 3, adding more consecutive harmonics to the driving voltage waveform increases the absolute value
of the strongest possible bias substantially: for example, in a triple-frequency discharge the absolute value of the strongest possible bias is 33% of the total voltage amplitude. This corresponds to an increase of more than 30% compared with the df case. Adding more consecutive harmonics increases the absolute value of the bias further. For \( k > 10 \) we observe a saturation of the normalized bias at \( |\tilde{\eta}| \approx 40\% \).

Figure 3 also shows that the simulation results cannot be reproduced by the original version of the EAE model for \( k > 2 \) due to the assumptions made in the frame of its derivation (see section 3). Using the symmetry parameter \( \epsilon \) from the PIC simulation and taking into account the floating potentials as well as the bulk voltage, which are both neglected in the original EAE model, yields good agreement between the modeling and simulation results.

Based on equation (7), the corrections to the bias resulting from the original EAE model, \( \Delta \tilde{\eta}_b \), due to taking into account the floating potentials as well as the bulk voltage, \( \Delta \tilde{\eta}_b \), can be calculated. Both terms are also affected by the correct symmetry parameter from the PIC simulation:

\[
\Delta \tilde{\eta}_b = \frac{1 - \epsilon}{1 + \epsilon} \tilde{\eta}_b - \sum_{n=1}^{k} \theta_n \phi_n^{(k)},
\]

\[
\Delta \tilde{\eta}_b = \tilde{\eta}_b - \tilde{\eta}_o - \Delta \tilde{\eta}_b.
\]

Here \( \tilde{\eta} \) is the normalized bias resulting from the extended EAE model (equation (7)).

Figure 4 shows these corrections to the original EAE model as a function of the number of applied consecutive harmonics, \( k \), at 100 Pa and \( \sum_{n=1}^{k} \phi_n^{(k)} = 150 \text{ V} \). The bulk voltage is approximately constant independently of \( k \) and the correction to the original EAE model due to taking into account the bulk voltage, \( \Delta \tilde{\eta}_b \) (third term in equation (8)), is negligible.

Taking into account the floating potentials and using the symmetry parameter from the PIC simulation as an input parameter for the extended model of the EAE affects the first and second term in equation (7) and results in the correction \( \Delta \tilde{\eta}_{b,c} \) shown in figure 4. The effect of \( \epsilon \) on the third term is neglected, since \( \Delta \tilde{\eta}_b \approx 0 \). \( \Delta \tilde{\eta}_{b,c} \) increases as a function of \( k \) from 0% at \( k = 2 \) to 3.5% at \( k = 10 \).

As shown in the left plot of figure 5, the floating potentials at both electrodes change as a function of \( k \). The floating potential at the grounded electrode, \( \phi_{bg}^{(k)} \), increases from 3.6 V at \( k = 2 \) to 6 V at \( k = 10 \). Physically, this is caused by the change of the applied voltage waveform: if 10 consecutive harmonics are applied, \( \phi_{bg}^{(k)}(t) \) will be minimum during most of the period of the applied fundamental frequency, \( T_{RF} \). This is similar to the voltage waveform applied in [26, 27]. As shown in the left plot of figure 6, this causes the sheath at the grounded electrode to be collapsed during most of \( T_{RF} \), while the sheath at the powered electrode is fully expanded almost all the time. The situation is significantly different for lower \( k \), e.g. \( k = 2 \) (right plot in figure 6), for which the sheath adjacent to the grounded electrode collapses only at two distinct times within \( T_{RF} \). Therefore, since the ion and electron fluxes to each electrode must compensate within \( T_{RF} \), the floating potential at the grounded electrode must be higher for \( k = 10 \) compared with the df scenario to prevent too high a continuous electron loss to the grounded electrode. The absolute value of the floating potential at the powered electrode, \( |\phi_{sp}^{(k)}(t)| \), decreases as a function of \( k \) from 1.6 V at \( k = 2 \) to 0.3 V at \( k = 10 \). This is caused by an increase of the first derivative of \( \phi_{sp}^{(k)}(t) \) around \( t = 0 \) (sheath collapse at the powered electrode), as a function of \( k \). This reduces the time when the sheath potential is lower than the electron temperature, i.e. it reduces the time during which electrons can reach the powered electrode to compensate the ion flux.

These characteristics of the floating potentials cause the maximum changes in both sheaths, \( Q_{mg} \) and \( Q_{mp} \), to become different at high \( k \). The high floating potential at ground causes a substantial fraction of the total uncompensated positive charge in the discharge, \( Q_{tot} \), to be located inside the sheath at ground, even when the sheath adjacent to the powered electrode
Figure 5. Left: floating potentials at the grounded (left scale, solid black line and triangles) and powered electrode (right scale, dashed blue line and squares) as a function of \( k \). Right: the symmetry parameter, \( \varepsilon \), as a function of \( k \). All of the data result from PIC simulations at 100 Pa (\( \sum_{n=1}^{\infty} \phi_n^{(k)} = 150 \text{ V}, \theta_n = 0^\circ \forall n \)).

Figure 6. Absolute values of the sheath voltages at the powered (dashed red line) and grounded electrode (solid black line) as a function of time within \( T_{RF} \) obtained from PIC simulations at 100 Pa, \( \sum_{n=1}^{\infty} \phi_n^{(k)} = 150 \text{ V}, \theta_n = 0^\circ \forall n \). Left: the discharge driven by 10 consecutive harmonics (\( k = 10 \)). Right: the discharge driven by 2 consecutive harmonics (\( k = 2 \)). The floating potentials at the grounded electrode, \( \phi_{sp} \), are indicated by the horizontal dotted lines.

is fully expanded. In contrast, there is almost no charge inside the sheath at the powered electrode when the sheath adjacent to the grounded electrode is fully expanded, since \( \phi_{sp} \) is very low. This reduces \( Q_{mp} \) relative to \( Q_{mg} \) and, therefore, also \( |\phi_{sp}| \propto Q_{mp} \) [51] relative to \( |\phi_{sp}| \propto Q_{mg} \) (see figure 6), which in turn causes \( \varepsilon \) to increase slightly as a function of \( k \) as shown in the right plot of figure 5.

Both the increase of \( \phi_{sp}^f \) and the decrease of \( |\phi_{sp}^f| (\phi_{sp}^f < 0) \) cause the second term in equation (7) to increase as a function of \( k \). Additionally, the floating potentials affect the symmetry parameter.

In retrospect, these results show that the original EAE model describes the EAE in df discharges accurately and for two frequencies the assumptions made in the context of its derivation are justified. However, if more than two frequencies are used, \( \Delta \eta \) will be significant and the extended version of the model must be used.

A comparison of the normalized self-bias values that can be reached at different pressures is shown in figure 7. The left plot shows the effect of choosing optimized individual amplitudes of harmonics according to equation (10) at high pressure compared with identical amplitudes of harmonics (PIC simulation: 100 Pa, \( \sum_{n=1}^{\infty} \phi_n^{(k)} = 150 \text{ V}, \theta_n = 0^\circ \forall n \)). For \( k = 2, 3, 4 \) the choice of optimized amplitudes increases the normalized dc self-bias by about 3.5%. The right plot in figure 7 shows \( \bar{\eta} \) as a function of \( k \) at 5 Pa (optimized amplitudes of harmonics, \( \sum_{n=1}^{\infty} \phi_n^{(k)} = 1500 \text{ V}, \theta_n = 0^\circ \forall n \)). At lower pressures a significantly stronger dc self-bias is generated electrically due to the self-amplification of the EAE [31]. At such low pressures the sheaths are almost collisionless and due to ion flux continuity inside the sheath a finite bias leads to different mean ion densities in both sheaths. This mechanism causes \( \varepsilon \) to deviate from unity in a way that leads to a stronger self-bias and self-amplifies the EAE [31, 32].

The results discussed so far clearly show that a significantly stronger dc self-bias can be generated electrically by driving the discharge with multiple consecutive harmonics and optimized amplitudes of harmonics. However, in order to control the ion energies at the electrodes via the EAE in these multi-frequency discharges, the self-bias must be adjustable. This can be realized by tuning the phase shifts, \( \theta_n \), between the driving voltage harmonics.
Figure 7. Normalized dc self-bias resulting from the PIC simulation as a function of \( k \) using identical amplitudes of harmonics and optimized amplitudes (equation (10)) at 100 Pa (left plot, \( \sum_{n=1}^{N} \phi_n = 150 \text{ V}, \theta_n = 0^\circ \ \forall \ n \)) and using optimized amplitudes at 5 Pa (right plot, \( \sum_{n=1}^{N} \phi_n = 1500 \text{ V}, \theta_n = 0^\circ \ \forall \ n \)).

Figure 8. Normalized dc self-bias in a triple-frequency discharge as a function of \( \theta_1 \) and \( \theta_2 \) (\( \theta_3 = 0^\circ \)) with optimized amplitudes of harmonics at high pressures. The bias is calculated by the original model of the EAE assuming \( \varepsilon = 1 \) (equation (8)).

For a discharge driven by three consecutive harmonics with optimized amplitudes of harmonics as an example, figure 8 shows \( \eta \) as a function of \( \theta_1 \) and \( \theta_2 \) (\( \theta_3 = 0^\circ \)) at high pressures. The data are calculated using the original model of the EAE assuming \( \varepsilon = 1 \) (equation (8)). In principle, the extended model could also have been used here. However, this would have required enormous computational effort to obtain the required input parameters from the PIC simulation for all relevant combinations of \( \theta_1 \) and \( \theta_2 \). Based on the results shown in figure 3, no big differences between the two model versions are expected for \( k = 3 \). Figure 8 shows multiple options to tune \( \eta \) from \( \eta_{\text{min}} \approx -33\% \) to \( \eta_{\text{max}} \approx +33\% \). As shown in figure 9 the most simple option is to tune only \( \theta_2 \) from \( 0^\circ \) to \( 180^\circ \) (\( \theta_1 = \theta_3 = 0^\circ \)). Generally, also for higher numbers of applied consecutive harmonics, the bias can always be tuned from its minimum to its maximum by setting the phase shifts of all odd harmonics to \( 0^\circ \) and only adjusting the phase shifts of the even harmonics (see appendix B).

Figure 9. Normalized dc self-bias in a triple frequency discharge as a function of \( \theta_2 \) (\( \theta_1 = \theta_3 = 0^\circ \)) with optimized amplitudes of harmonics at high pressures. The bias is calculated by the original model of the EAE assuming \( \varepsilon = 1 \) (equation (8)).

4.3. The effect of the optimized voltage waveform on the range of ion energy control

First, the effect of the optimized voltage waveform on the range of ion energy control in an argon discharge operated at 10 Pa and driven by three consecutive harmonics with optimized amplitudes of harmonics is discussed in detail as an example. Second, the range of ion energy control in discharges driven by more than three consecutive harmonics is analyzed.

Figure 10 shows the dc self-bias resulting from the PIC simulation as a function of \( \theta_2 \) in a triple-frequency discharge (fundamental frequency 13.56 MHz). The amplitudes of the harmonics are optimized according to equation (10) (\( \sum_{n=1}^{N} \phi_n = 300 \text{ V} \)). The horizontal dashed lines indicate the maximum and minimum bias that can be generated electrically in a df discharge driven at 13.56 and 27.12 MHz with \( \phi_1^{(2)} + \phi_2^{(2)} = 300 \text{ V} \) (optimized amplitudes) and otherwise identical discharge conditions. Compared with the df scenario the range of bias control via the EAE is enhanced by about 33\% by adding a third consecutive harmonic.
Figure 11 shows the ion flux-energy distribution functions at the powered (left plot) and grounded electrode (right plot) for different phase angles $\theta_2$ ($\theta_1 = \theta_3 = 0^\circ$) in the triple-frequency discharge (PIC simulation). The ion energy can be controlled by tuning $\theta_2$ from $0^\circ$ to $180^\circ$ and the role of both electrodes can be reversed. The peaks are caused by ion-neutral charge exchange collisions inside the sheaths in combination with the time modulated RF sheath voltage [52].

Figure 12 shows the mean ion energy and the ion flux at both electrodes as a function of $\theta_2$ in this customized triple-frequency discharge. The horizontal lines in the left plot indicate the maximum and minimum mean ion energy that can be realized in a df discharge driven at 13.56 and 27.12 MHz with $\phi_1^{(2)} + \phi_2^{(2)} = 300$ V and otherwise identical discharge conditions. By adding a third harmonic to the driving voltage waveform and choosing amplitudes of harmonics according to equation (10) the control range of the mean ion energy at both electrodes is enhanced from a factor of about 2 in df discharges to a factor of about 3 in triple-frequency discharges, while the ion flux remains constant within $\pm 15\%$.

The left plot in figure 13 shows the factor $\chi_i$ by which the mean ion energy at both electrodes can be changed via the EAE and the normalized dc self-bias as a function of the number of applied consecutive harmonics, $k$, in an argon discharge operated at 10 Pa (optimized amplitudes of harmonics, $\sum_{n=1}^{k} \phi_n^{(k)} = 300$ V):

$$\chi_i = \frac{\langle E \rangle_{\text{max}}}{\langle E \rangle_{\text{min}}}.$$  \hspace{1cm} (13)

Here $\langle E \rangle_{\text{max}}$ and $\langle E \rangle_{\text{min}}$ are the maximum and minimum mean ion energies at a given electrode that can be realized by tuning the phase shifts, $\theta_2$, between the driving harmonics. These results show that the range of mean ion energy control can be drastically enhanced up to a factor of about 9 by adding more consecutive harmonics to the applied voltage waveform. This substantial improvement of the range of ion energy control is expected to be most relevant for various technological applications of CCRF discharges.

While the absolute value of the dc self-bias basically does not increase as a function of $k$ at high numbers of applied consecutive harmonics, $\chi_i$ still increases. This is caused by an increase in $\langle E \rangle_{\text{max}}$ at almost constant $\langle E \rangle_{\text{min}}$. This in turn results from the following mechanism: as the total applied voltage, $\sum_{n=1}^{k} \phi_n^{(k)}$, is kept constant at 300 V, but higher frequencies are added, the plasma density increases from about $2.7 \times 10^9 \text{ cm}^{-3}$ at $k = 2$ to about $2.1 \times 10^{10} \text{ cm}^{-3}$ at $k = 16$ [53]. A similar increase in the ion fluxes to the electrodes is observed. This increase by almost one order of magnitude causes the maximum width of the sheaths to decrease. The left plot in figure 14 shows the maximum width of the sheath adjacent to the powered electrode, $s$, as a function of $k$. According to Brinkmann [54] $s$ is calculated by the following criterion:

$$\int_0^s n_e(x) \, dx = \int_{s/2}^{L/2} (n_i(x) - n_e(x)) \, dx.$$  \hspace{1cm} (14)

Here $n_e$ and $n_i$ are the electron and ion densities, respectively, $L$ is the electrode gap and $x$ is the distance from the powered electrode.

**Figure 10.** Dc self-bias resulting from the PIC simulations as a function of $\theta_2$ in a discharge driven by three consecutive harmonics at 10 Pa. The amplitudes of the harmonics are optimized according to equation (10) ($\sum_{n=1}^{k} \phi_n^{(k)} = 300$ V). The horizontal dashed lines indicate the maximum and minimum mean ion energy that can be realized in a df discharge driven at 13.56 and 27.12 MHz with $\phi_1^{(2)} + \phi_2^{(2)} = 300$ V (optimized amplitudes) and otherwise identical discharge conditions.

**Figure 11.** Ion flux-energy distribution functions at the powered (left plot) and grounded electrode (right plot) for different phase angles, $\theta_2$, ($\theta_1 = \theta_3 = 0^\circ$) in an argon discharge operated at 10 Pa and driven by three consecutive harmonics (PIC simulation). The amplitudes of the harmonics are optimized according to equation (10) ($\sum_{n=1}^{k} \phi_n^{(k)} = 300$ V).
Figure 12. Mean ion energy (left plot) and ion flux (right plot) at the powered (black line and squares) and grounded electrode (red line and circles) as a function of $\theta_2$ ($\theta_1 = \theta_3 = 0^\circ$) in an argon discharge operated at 10 Pa and driven by three consecutive harmonics with optimized amplitudes (PIC simulation, $\sum_k n \phi_n^{\text{tot}} = 300$ V). The horizontal lines in the left plot indicate the maximum and minimum mean ion energy that can be realized in a dc discharge driven at 13.56 and 27.12 MHz with $\phi_1^{(2)} + \phi_2^{(2)} = 300$ V (optimized amplitudes) and otherwise identical discharge conditions.

Figure 13. Left plot: control factor, $\chi_i$, by which the mean ion energy at both electrodes can be changed via the EAE (black triangles, left scale) and normalized dc self-bias (blue squares, right scale) as a function of $k$. Right plot: the ion flux-energy distribution functions at the powered electrode for different $k$ at $\theta_n = 0^\circ \forall n$. Discharge conditions: argon, 10 Pa, $\sum_k n \phi_n^{\text{tot}} = 300$ V (PIC simulation).

Figure 14. Left: PIC results for the maximum width of the sheath adjacent to the powered electrode, $s$, as a function of $k$. The horizontal dashed line indicates the ion mean free path, $\lambda_i \approx 1.4$ mm. Right plot: the ion distribution function at the powered electrode for different $k$ calculated from equation (15). Discharge conditions: argon, 10 Pa, $\sum_k n \phi_n^{\text{tot}} = 300$ V.

Since $s$ decreases as a function of $k$, but the ion mean free path, $\lambda_i$, remains constant, the ions undergo fewer collisions inside the sheath for higher $k$ and, therefore, a high energy peak such as observed in the right plot of figure 13 is formed with increasing $k$. This phenomenon can be described by the model of Lawler [55] for dc discharges. For high $k$ the voltage drop across the sheath adjacent to the powered electrode is similar to the situation in a dc discharge (see left plot in figure 6). Under the assumption of a current source at the sheath edge, a linearly increasing field, and charge exchange collisions as
the dominant collision type with an energy independent cross section, this model yields the following expression for the ion distribution function, \( f(E) \):

\[
f(E) = j \frac{M_i}{2E_0} e^{-\alpha} \left[ 2\delta(E - 1) + \frac{e^{\alpha\sqrt{1 - E}}}{\sqrt{1 - E}} \right].
\]

(15)

Here \( j \) is the ion flux into the sheath, i.e. the Bohm flux, \( M_i \) is the ion mass, \( \bar{E} = E/E_0, E = M_i v_i^2/2 \) is the ion energy, \( v_i \) is the ion velocity and \( E_0 \) is the maximum ion energy at the electrode. \( \phi_i \) is the temporally averaged sheath potential, \( q_i \) is the ion charge and \( \alpha = s/\lambda_i \). Equation (15) ensures that the ion flux within the sheath is constant. The second term is identical to the model of Davis and Vanderslice [56] for the ion flux within the sheath is constant. The second term is greatly reduced \([52]\). Particularly, the formation of the high energy peak with increasing \( k \) is reproduced and the model reveals its physical origin. The finite width of the high energy peak, however, is not reproduced. This width is most likely mainly caused by the temporal modulation of the sheath potential \([57, 58]\), which is not taken into account here.

Experimentally, adding one or more consecutive harmonics to the driving voltage waveform requires effort, which must be motivated by a substantial gain in process quality. The technological problem of matching in such multi-frequency discharges is not addressed in this work, but is clearly an experimental problem. However, based on our results driving a discharge at 3 or 4 consecutive harmonics will certainly be worth the effort, if a wide range of ion energy control (separate from the ion flux) is important for the process of interest.

5. Conclusions

The EAE in geometrically symmetric CCRF discharges operated at multiple consecutive harmonics has been investigated by a PIC simulation and an extended version of the original EAE model, which takes into account the floating potentials, the bulk voltage and the symmetry parameter from the PIC simulation to describe the EAE correctly in multi-frequency discharges.

Compared with electrically asymmetric rf discharges we have found that a significantly stronger variable dc self-bias can be generated electrically in discharges driven by multiple consecutive harmonics. For instance, by adding a third consecutive harmonic the strongest bias is increased by more than 30%. A recipe for customizing the applied voltage waveform to generate the strongest possible dc self-bias electrically has been presented. In analogy to Fourier theory the difference between the absolute values of the global extrema of the driving voltage waveform is maximized by choosing specific individual amplitudes of harmonics. In this way the bias can be tuned from its minimum, \( \eta_{min} \), to its maximum, \( \eta_{max} = -\eta_{min} \), by only adjusting the phase shifts of the even harmonics, while the phase shifts of the odd harmonics are set to 0°.

The corrections to the original EAE model made in the context of the extended model have been discussed individually and their effect on the correct prediction of the dc self-bias has been determined. It has been found that particularly the floating potentials must not be neglected at high numbers of applied consecutive harmonics.

The simulation results indicate that the range of mean ion energy control can be substantially increased by driving a discharge with multiple instead of only two consecutive harmonics with optimized amplitudes of harmonics. While the mean ion energy can only be changed by a factor of about 2 separately from the ion flux in an electrically asymmetric argon rf discharge operated at 10 Pa, it can be changed by a factor of about 3 in triple-frequency discharges and up to
a factor of about 9 if more consecutive harmonics are added. This substantial improvement to the range of ion energy control is expected to be most relevant for various technological applications of CCRF discharges.

The shape of the ion distribution function is found to change as a function of the number of applied consecutive harmonics, i.e. a high energy peak is formed at a high number of consecutive harmonics. This change of the shape of the IEDF is explained based on an analytical model originally developed by Lawler [55] for dc discharges.

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Appendix A. Characteristics of the normalized applied voltage waveform, \( \tilde{f}_N^{(k)}(t) \), with optimized amplitudes of harmonics

For \( \Theta_n = 0 \forall n \) the value of the global maximum of the normalized applied voltage waveform, \( \tilde{f}_N^{(k)}(t) \), (equation (9)) is located at \( t = 0 \) within one period of the fundamental frequency independent of \( k \). Its value is always unity, independent of \( k \). This can directly be understood by substituting \( t = 0 \) into equation (9).

By substituting \( ft_m = 2\pi m/(k + 1) \) \( m = 1, 2, \ldots, k \) into the first temporal derivative of \( \tilde{f}_N^{(k)}(t) \) for \( \Theta_n = 0 \forall n \), the \( k \) global minima of \( \tilde{f}_N^{(k)}(t) \) within one period of \( f \) are shown to be located at \( t_m \):

\[
\frac{\partial}{\partial t} \tilde{f}_N^{(k)}(t)|_{t=t_m} = 0
\] (A.1)

\[\rightarrow \sum_{n=1}^{k} (k-n+1)n \sin \frac{2\pi m}{k+1} = 0.\] (A.2)

Substituting

\[
\sin \left( \frac{2\pi m}{k+1} \right) = \frac{1}{2i} (a^n - a^{-n})
\] (A.3)

with \( a = \exp[2\pi i m/(k+1)] \) into equation (A.2) yields

\[
(k+1) \sum_{n=1}^{k} n (a^n - a^{-n}) = \sum_{n=1}^{k} n^2 (a^n - a^{-n}).
\] (A.4)

Using (i) \( a^{k+1} = 1 \), (ii) \( \sum_{n=1}^{k} na^n = a/(a-1)^2[1-(1+k)a^k + ka^{k+1}] \) and (iii) \( \sum_{n=1}^{k} n^2a^n = a/(a-1)^2[1-a+k^2a^{2k} + (1+k)^2a^k + (1-2k+2k^2)a^{2k}] \) yields

\[
(k+1) \sum_{n=1}^{k} n (a^n - a^{-n}) = \sum_{n=1}^{k} n^2 (a^n - a^{-n})
\] (A.5)

Using the second temporal derivative of \( \tilde{f}_N^{(k)}(t) \) it can be explicitly shown, e.g. via complete induction, that minima are located at \( t_m \) \( (\partial^2/\partial t^2)\tilde{f}_N^{(k)}(t_m) > 0 \).

Using \( \sum_{n=1}^{k} n = k(k+1)/2 \) and \( \cos(n ft_m) = 1/2(a^n + a^{-n}) \), the value of these minima is calculated by substituting \( t = t_m \) into \( \tilde{f}_N^{(k)}(t) \):

\[
\tilde{f}_N^{(k)}(t_m) = \frac{2}{k(k+1)} \sum_{n=1}^{k} (k+1-n) (a^n + a^{-n}).
\] (A.6)

With \( \sum_{n=1}^{k} a^n = \sum_{n=1}^{k} a^{-n} = -1 \) equation (A.6) yields

\[
\tilde{f}_N^{(k)}(t_m) = -1/k,
\] (A.7)

independent of \( m \), i.e. the values of all \( k \) minima are identical.

Appendix B. Tuning the dc self-bias by adjusting only the phase shifts of the even harmonics

Under the assumption of \( \varepsilon = 1 \) the most negative dc self-bias will be generated if all of the phase shifts of the harmonics are set to 0, i.e. \( \Theta_n = 0 \forall n \), since the sum of the global maximum at this particular choice of phase angles, \( \tilde{\phi}_{m1}^{(0)} \), and the global minimum, \( \tilde{\phi}_{m2}^{(0)} \), of \( \tilde{f}_N^{(k)}(t) \) is maximum positive (equation (8)). For a specific choice of phase shifts, \( \tilde{\eta}_n \), the sign of the bias can be reversed. For this choice of phase shifts the sum of the global maximum, \( \tilde{\phi}_{m1}^{(0)} \), and the global minimum, \( \tilde{\phi}_{m2}^{(0)} \), of the applied voltage waveform is maximum negative (equation (8)) and

\[
\tilde{\phi}_{m1}^{(0)} = -\tilde{\phi}_{m2}^{(0)},
\] (B.1)

Using equation (2), equation (B.1) yields

\[
\sum_{n=1}^{k} \tilde{\phi}_{m1}^{(k)} \cos(n x + \theta_n^{(0)}) = -\sum_{n=1}^{k} \tilde{\phi}_{m2}^{(k)} \cos(n x + \theta_n^{(0)}).
\] (B.2)

Here \( x = 2\pi ft \) and \( x_0 \) is a phase shift, which considers that \( \tilde{\phi}_{m1}^{(0)} \) and \( \tilde{\phi}_{m2}^{(0)} \) do not have to occur at the same time within one period of \( f \).

Equation (B.2) must be fulfilled for every summand individually:

\[
\cos(n x + x_0) + \cos(n x + \theta_n^{(0)}) = 0
\] (B.3)

\[\rightarrow \cos(n x)[\cos(x_0) + \cos(\theta_n^{(0)})]
\]

\[- \sin(n x)[\sin(x_0) + \sin(\theta_n^{(0)})] = 0.
\] (B.4)

Equation (B.4) is only valid for all \( x \), if

\[
\cos(x_0) + \cos(\theta_n^{(0)}) = 0 \land \sin(x_0) + \sin(\theta_n^{(0)}) = 0
\] (B.5)

\[\rightarrow \theta_n^{(0)} = nx_0 \pm \pi.
\] (B.6)

If \( x_0 = \pi \) is chosen, equation (B.6) proves that the dc self-bias can be tuned from its minimum, \( \eta_{\text{min}} \), to its maximum, \( \eta_{\text{max}} = -\eta_{\text{min}} \), by adjusting only the individual phase shifts of the even harmonics \( n \) (even) independent of the total number of applied consecutive harmonics \( k \).
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