Frequency coupling in low-pressure dual-frequency capacitively coupled plasmas revisited based on the Boltzmann term analysis

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Received 6 May 2022, revised 26 September 2022
Accepted for publication 4 October 2022
Published 7 November 2022

Abstract

Electron power absorption dynamics is investigated in radio-frequency (RF) argon capacitively coupled plasmas (CCPs) at low pressure (4–70 Pa) excited by a dual-frequency waveform with frequencies of 27.12 MHz and 1.937 MHz. Based on the spatio-temporal dynamics of the ambipolar electric field a novel interpretation of the mechanism of frequency coupling is given, which is not based on the hard wall model, as in previous explanations. Within this framework, frequency coupling arises due to the decreased size of the ambipolar region outside the sheath when the low-frequency sheath is close to its full expansion, which leads to decreased ionization in this region. It is shown, under the circumstances considered here, ohmic power absorption is dominant. The spatio-temporally averaged ambipolar power absorption shows nonmonotonic behaviour as a function of pressure, first increasing, then, after reaching a local maximum, decreasing as the pressure is increased. It is shown, that the reason for this nonmonotonic behaviour is ultimately connected to the frequency coupling mechanism.

Keywords: electron power absorption, capacitively coupled plasma, frequency coupling, Boltzmann term analysis, particle-in-cell simulation, dual frequency discharges

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

Low-pressure capacitively coupled radio-frequency plasmas (RF CCPs) are prevalent in various industrial applications, ranging from surface manufacturing to biomedicine [1–5]. In particular, for etching processes, CCPs with two simultaneous frequency components, i.e. dual-frequency CCPs have been widely used, as they allow, at least to a certain extent, separate control of the plasma density and the ion impact energy on the substrate [6–8]. There has been a vast amount of research conducted on the topic of dual-frequency discharges, investigating electron power absorption (heating) mechanisms in dual-frequency (DF) discharges, such as collisionless heating [9–11], nonlinear electron resonance heating [12–15], bounce resonance heating [16–18], heating mode transitions [19–22], the separate control of ion energy and flux and the electrical asymmetry effect [23–35]. An important mechanism, that makes this separate control limited, is the frequency coupling [36–38]. Ideally, the low-frequency voltage amplitude is responsible for the mean energy of the ions, while the high-frequency voltage amplitude determines the ionization rate and, with this, the flux of the ions. However, it was found that, in the absence of secondary electrons, increasing the low-frequency amplitude will decrease the flux. An explanation for this was given based on the hard wall model: when the low-frequency sheath is close to its full expansion, the instantaneous sheath edge reaches a region with a high ion density. Consequently, the speed at which the local high-frequency sheath width can increase will be smaller than in regions with a low ion density [19]. Thus, within the framework of the hard wall model, where the sheath expansion velocity determines the electron power absorption, stochastic heating is attenuated and the local ionization is diminished when the low-frequency sheath is expanded compared to times when the local low-frequency sheath is short. It is crucial that this explanation assumes that the sheath edge resembles a hard wall, from where electrons can ‘bounce back’, as no electric field is assumed to exist outside the sheath. However, this has recently been demonstrated to not be a good assumption [39].

In this paper the electron power absorption dynamics in low-pressure DF CCPs are investigated using the Boltzmann term method, a spatio-temporal analysis of electron power absorption [40–42] and, based on this, a novel interpretation of frequency coupling is given, which does not use the assumptions of the hard wall model. The Boltzmann term method was first proposed in the seminal paper of Surendra and Dalvie [40] and has since been used to investigate electron power absorption in CCPs at low pressure for inert [41–44] as well as reactive gases [45–47], in low-pressure CCPs under the effect of a magnetic field [48–50], in atmospheric microplasma jets [51] and in Hall thrusters [52–54]. Schulze et al [42] showed that, in low-pressure argon discharges excited by a single-frequency waveform, the dominant power absorption mechanism on space and time average is pressure heating, which is fundamentally related to the ambipolar electric field. The reason for this was found to be a temporal asymmetry in the parallel electron temperature between the expanding and collapsing phases of the sheath, which is a consequence of the following self-amplifying mechanism: the ambipolar electric field is proportional to the normalized electron density gradient, i.e. \( E_{\nabla n} \propto -/(1/n_e)(\partial n_e/\partial x) \). Thus, for an inert gas such as argon, where the electron density monotonically increases from the electrode towards the center of the discharge, the ambipolar field accelerates electrons towards the center of the discharge. Thus, this electric field ‘pushes away’ electrons from the vicinity of the instantaneous sheath towards the plasma bulk. Thus, in one half of the RF cycle, electrons will be accelerated by this electric field while, in the other half, they will be decelerated.

In a symmetric discharge, the conduction current density will change sign in the two halves of the RF cycle, but remains identical in magnitude. Thus, unless there is a temporal asymmetry in the ambipolar power absorption, it will necessarily be zero on space and time average. It was found that the parallel electron temperature, \( T_\parallel \) to which the ambipolar electric field is also proportional, is temporally asymmetric. The reason for this is that during sheath expansion, where electrons gain energy from the ambipolar field, the temperature locally increases (as it is determined by the electrons themselves). As the electric field depends on the temperature, if it increases, so does the electric field, which can accelerate electrons even more. This self-amplifying mechanism stops when the sheath is fully expanded. During sheath collapse, such a local temperature increase does not happen, as the incoming electrons are decelerated. Thus, due to this temporal asymmetry, the ambipolar power absorption and, subsequently, pressure heating, is nonzero on space and time average in a low-temperature CCP excited by a single-frequency waveform.

In this work, dual-frequency discharges are investigated. Here, as the electron conduction current originating from the low-frequency component is superimposed to that of the high-frequency component, the above temporal asymmetry argument is no longer tenable, at least locally, as the magnitudes of the electron conduction current densities will not be the same in the two halves of the (high-frequency) RF cycle. We show that although the temporal asymmetry argument holds in this case as well, at least in a global sense, the presence of a low-frequency component changes the spatio-temporal dynamics of the ambipolar electric field, which is intimately related to the frequency coupling mechanism, for which, thus, a novel interpretation can be given within the framework of the Boltzmann term analysis. This interpretation is not related to the local sheath velocity, but is entirely based on the dynamics of the ambipolar electric field, and the decrease in the size of the ambipolar region outside the sheath, when the low-frequency sheath is close to its full expansion. We show that this leads to an overall decrease in ambipolar power absorption on space and time average and to nonmonotonic behaviour as a function of pressure.

The paper is structured as follows: section 2 describes the Boltzmann term analysis and provides details about the simulation method applied in this paper. In section 3, the results are presented and discussed. Finally, in section 4, conclusions are drawn.
2. Computational method and theoretical background

Our numerical studies are based on a 1d3v electrostatic PIC/MCC code [55, 56]. We investigate argon discharges with a fixed electrode gap of $L = 25$ mm in the pressure range between 4 and 70 Pa. The plasma is excited by a dual-frequency waveform, $\phi(t)$, given by

$$\phi(t) = \phi_1 \cos(2\pi f_1 t) + \phi_2 \cos(2\pi f_2 t),$$

with $\phi_1 = 250$ V, $\phi_2 = 500$ V, $f_1 = 27.12$ MHz, $f_2 = f_1/14 = 1.937$ MHz. For the surface model of the electrode a constant electron reflection coefficient of 0.2 is assumed [57]. As this is the first attempt to investigate the frequency coupling effect based on the Boltzmann term analysis, we simplify the investigated scenarios by disregarding secondary electron emission from the electrodes. The temperature of the background gas is held at $T_g = 350$ K. In the simulation electrons and positive argon ions are traced. The spatial resolution of $N_x = 1200$ grid points and a temporal resolution of $N_t = 70000$ timesteps are used to ensure the stability of the simulation and good numerical accuracy of the results. To obtain good statistics, the superparticle number is kept at $\approx 2 \times 10^5$. The collision cross sections for electron–atom collisions are taken from [58] while, for the ion–atom collisions, they are taken from [59]. The collision model is identical to that described in, e.g. [56]. The code has been validated against experiments [60]. The results are obtained by accumulating data for 3000 RF cycles in the converged state of the system.

To investigate the spatio-temporal dynamics of the electric field and the electron power absorption, the Boltzmann term analysis approach is used [42, 43, 47], for which the input parameters are provided by the PIC/MCC simulations. This method has been thoroughly described in, e.g. [43, 44]; here, we only mention the most important concepts. The method is based on the spatially one-dimensional momentum balance equation for the electrons

$$m_e \frac{\partial}{\partial t} (n_e u_e) + m_e \frac{\partial}{\partial x} (n_e u_e^2) = -n_e e E_{\text{tot}} - \frac{\partial}{\partial x} p_l - \Pi_e. \quad (2)$$

Here, $e$, $m_e$, $n_e$ and $u_e$ are the elementary charge, the electron mass, density and mean velocity, respectively. Meanwhile, $p_l$ denotes the diagonal element of the pressure tensor for the electrons (here, ‘parallel’ refers to the direction of the discharge axis, which coincides with the direction of the electric field and is perpendicular to the plane of the electrodes). $\Pi_e$ denotes the electron momentum loss, is given by $\Pi_e = m_e n_e n_f \langle \sigma_{\text{el}} v_f \rangle$, where $\sigma_{\text{el}}$ is the total momentum transfer cross section (including elastic as well as inelastic collisions, see [61]), and accounts for the effect of collisions between electrons and the atoms of the background gas. In the simulations, $\Pi_e$ is calculated by adding the quantity $n_e m_e \sigma_{\text{el}} v_f$ for each electron in each timestep to the respective spatio-temporal bin of $\Pi_e$, weighted by the electron weight. The $\Pi_e$ is then properly normalized at the end of the simulation. The electric field $E_{\text{tot}}$ can be written as a sum, $E_{\text{tot}} = E_{\text{in}} + E_{\text{CP}} + E_{\text{Ohm}}$, where the individual terms are given by

$$E_{\text{in}} = -\frac{m_e}{n_e} \left[ \frac{\partial}{\partial t} (n_e u_e) + \frac{\partial}{\partial x} (n_e u_e^2) \right],$$

$$E_{\text{CP}} = -\frac{1}{n_e} \frac{\partial}{\partial x} p_l, \quad (3)$$

$$E_{\text{Ohm}} = -\frac{\Pi_e}{n_e}.$$ 

The inertial electric-field term $E_{\text{in}}$ is the electric field needed to compensate for the change in the electrons’ momentum. The ohmic field, $E_{\text{Ohm}}$, describes the electric field needed to account for the collisional momentum loss of electrons as a result of collisions with the background gas atoms, while $E_{\text{CP}}$, the electric field corresponding to the spatial gradient of the electron pressure [62], is usually divided into two separate electric-field terms, according to $E_{\text{CP}} = E_{\text{Cn}} + E_{\text{CT}}$, where

$$E_{\text{Cn}} = \frac{T_i}{n_e} \frac{\partial n_e}{\partial x},$$

$$E_{\text{CT}} = -\frac{1}{e} \frac{\partial T ||}{\partial x}. \quad (4)$$

In these equations $T || = p_i/n_e = m_e \langle v_e^2 \rangle - u_e^2\rangle v = \frac{m_e}{n_e} \int d^3v (v_e - u_e)^2 f_e$ is the parallel electron temperature, which is a measure of the mean kinetic energy of the electron ensemble in its center of mass frame. Here, $E_{\text{CT}}$ is the electric-field term originating from the spatial variation of the temperature, while $E_{\text{Cn}}$ is, in quasineutral regions, the ‘classical ambipolar field’ [63].

After calculating the respective electric-field terms, the corresponding electron power absorption terms can be easily calculated by multiplying each electric-field term with the electron conduction current density, $j_e$.

3. Results

In this section, results are presented for a dual-frequency waveform with amplitudes $\phi_{1, 2} = (250, 500)$ V and corresponding frequencies $f_{1, 2} = (27.12, 1.937)$ MHz in the pressure range between 4 Pa and 70 Pa for argon gas.

Before discussing the results under these discharge conditions in detail, figure 1(a) shows a sketch that aids the understanding of the interpretation of frequency coupling within the framework of the Boltzmann term analysis, while figure 1(b) shows the applied voltage waveform. As will be shown later, the interpretation of frequency coupling rests on the spatio-temporal dynamics of the ambipolar electric field, $E_{\text{CP}} = -(T_i/n_e)(\partial n_e/\partial x)$. As shown in, e.g. [42], the electron density gradient in an inert electropositive gas such as argon, is significant between each electrode and the maximum of the corresponding sheath width, $x = s_{\text{max}}$. However, electrons, which are accelerated by the ambipolar electric field, can effectively gain energy only in the spatial region between the instantaneous sheath edge and $s_{\text{max}}$, where the electron density is significant. We note that under the conditions investigated here, the ion density was not found to be time modulated, and thus it is enough to concentrate on the electron dynamics.
Figure 1. A sketch for the illustration of the interpretation of the frequency coupling (a) and applied voltage waveform, $\phi(t)$ (b). The colormap in panel (a) shows the ambipolar electron power absorption, with red/blue corresponding to positive/negative values, respectively (without units, only for illustration purposes). The Roman numbers denote ambipolar regions in one RF cycle. The black line indicates the sheath edge. The powered electrode is situated at $x = 0$. In panel (a), the sheath region is indicated in the grey colour.

The importance of the ambipolar electric field necessitates the definition of the ambipolar region, i.e. a spatio-temporal region ranging between $s_{\text{max}}$ and the instantaneous sheath edge, where a nonzero ambipolar electric field can accelerate electrons.

Thus, in figure 1(a), three ambipolar regions are listed: in regions I and III the low frequency (LF-) sheath width is small, and thus the available spatial region for an electron to be accelerated inside the ambipolar region is much greater than in region II, where the LF sheath is close to its full expansion, and thus electrons can only be accelerated in a limited spatial and temporal interval. The fact that the available ambipolar region for an electron is much smaller when the LF sheath is close to its full expansion, than when it is small, is one of the main reasons behind the mechanism of the frequency coupling.

Figure 2 shows the spatio-temporally averaged electron power absorption terms and their sum as a function of pressure. The total power absorption, $P_{\text{tot}}$, monotonically increases as a function of pressure due to the reason mentioned above, i.e. the increase in the number of collisions between electrons and background gas atoms. $P_{\text{Ohm}}$ is the dominant term throughout the whole pressure regime. The pressure gradient term, consisting of the ambipolar term, $P_{\nabla n}$, and the temperature gradient term, $P_{\nabla T}$, is smaller in amplitude than the ohmic term. Its value is close to zero at the lowest pressure, and increases monotonically until ≈ 7 Pa, where it reaches a maximum. Then, as the pressure is further increased, it decreases, acquiring a negative value for pressures above 20 Pa. This behaviour is closely related to the mechanism of frequency coupling, as will be discussed below.

Figure 2. Space- and time-averaged power absorption terms and their sum as a function of pressure. Discharge conditions: $L = 25$ mm, $(\phi_1, f_1) = (250 \text{ V}, 27.12 \text{ MHz})$, $(\phi_2, f_2) = (500 \text{ V}, 1.937 \text{ MHz})$. Comparing $P_{\nabla n}$ and $P_{\nabla T}$, one can see that while the former monotonically decreases as a function of pressure, the ambipolar term shows nonmonotonic behaviour.

Thus, under the conditions investigated, what needs to be explained is why the ambipolar power absorption shows this particular behaviour as a function of pressure, which leads to ohmic power absorption making the dominant contribution to the sustainment of the plasma. To investigate the electron power absorption dynamics in more detail, figure 3 shows the temporally averaged electron power absorption terms as a function of position for four different pressure values, 4 Pa (a), 7 Pa (b), 15 Pa (c) and 30 Pa (d), respectively. Figure 3(a) shows the power absorption terms for 4 Pa. Due to the low pressure the maximum sheath width, $s_{\text{max}}$, is large and, consequently, the bulk region is relatively narrow. The main contribution comes from ohmic power absorption, which is highest in the spatial region between the electrode and $s_{\text{max}}$, where the electron density is relatively low, and so is the conductivity of the plasma. The temperature gradient term, $P_{\nabla T}$, is positive in the vicinity of the electrodes, and decreases rapidly near the positions of the maximum sheath widths: a direct consequence of the spatio-temporal dynamics of the parallel electron temperature, as shown later. The ambipolar power absorption is negative in the vicinity of the electrodes, while it is positive near the positions of the maximum sheath widths, albeit with a smaller magnitude than the ohmic term.

This behaviour changes as the pressure is increased, as shown in figure 3(b), as here the ambipolar power absorption has a maximum near the positions of the maximum sheath
The vertical dashed grey lines indicate the positions of the maximum sheath width. Discharge conditions: $L = 25 \text{ mm}$, $(\phi_1, f_1) = (250 \text{ V}, 27.12 \text{ MHz})$, $(\phi_2, f_2) = (500 \text{ V}, 1.937 \text{ MHz})$. The density gradient leads to the local maximum of $P_\nabla n$. Similarly, $P_\nabla T$ shows a deep minimum around the position of $s_{\text{max}}$, as around this position the parallel temperature drastically decreases. Upon further increasing the pressure (figures 3(c) and (d)), the ambipolar power absorption decreases, as the marked maxima around $s_{\text{max}}$ disappear. At the same time, the ohmic power absorption increases monotonically, even in the bulk region. As $P_\nabla T$ increases in the bulk region as the pressure is increased, at 30 Pa (figure 3(d)) there is a significant power absorption in the bulk region, largely attributed to the ohmic term.

As shown in, e.g., [42], the positivity of the ambipolar power absorption on the space and time average in a single-frequency discharge rests on the temporal asymmetry of the electron temperature, which uses the fact that in one half of the RF cycle the electron conduction current density is permanently positive, while in the other half it is permanently negative. However, in a dual-frequency discharge this is no longer the case and thus, as a first step, it would be desirable to know how the electron current density is changed when a component with a second frequency is included in the RF driving-voltage waveform. Figure 4 shows the spatio-temporal distribution of the electron conduction current density, $j_c$, for the two pressure values, 4 Pa (a) and 30 Pa (b). In a geometrically symmetric single-frequency discharge the current is symmetric in time, i.e. $j_c(t) = -j_c(t + T_{\text{RF}}/2)$ holds, where $T_{\text{RF}}$ is the RF period. In the dual-frequency case this is no longer true (with $T_{\text{RF}} = 1/f_{\text{LF}}$), as there is a low-frequency component superimposed on the high-frequency waveform. In the total electron conduction current density both the high frequency (HF) and LF contributions need to be taken into account. Given that the LF current changes sign only after half of the total RF period, locally, i.e. in the two halves of a given HF period, the currents will not be equal in magnitude. This is shown in figures 4(a) and (b) for the HF cycle marked by the black circle (at $\approx 120 \text{ ns}$). The vertical dashed magenta and green lines indicate the times of maximum HF currents, respectively.

The presence of the LF current, which during the time of this specific HF cycle is negative, will lead to different magnitudes of the currents in the two halves of the HF cycle. Thus, locally, the argument of temporal asymmetry, which was the basis of the explanation for a nonzero ambipolar power absorption in space and time average, cannot be used. However, this argument is still applicable globally, as for each HF cycle during the first half of the total RF cycle, an equivalent...
Figure 4. Spatio-temporal distribution of the electron conduction current density, $j_e$, in units of A m$^{-2}$ for 4 Pa (a) and 30 Pa (b), and spatial distribution of the electron conduction, displacement and ion conduction current density at specific time instances, indicated by the coloured dashed lines in panels (a) and (b) for 4 Pa (c) and 30 Pa (d). The black lines indicate the sheath edges, and the black/white points in panels (a) and (b) indicate the equivalent HF cycles, see text. Discharge conditions: $L = 25$ mm, $(\phi_1, f_1) = (250$ V, $27.12$ MHz), $(\phi_2, f_2) = (500$ V, $1.937$ MHz).

HF cycle can be found in the second half of the RF cycle, where the currents are exactly opposite. The equivalent HF cycle is defined as the cycle shifted in time by half the LF period, and mirrored around the midplane of the discharge, e.g. for $j_e(x, t)$, the equivalent spatio-temporal point is $j_e(L - x, t + T_{RF}/2)$. The equivalent cycle for the one denoted by the black circle is marked with a white circle in figures 4(a) and (b). The vertical dashed red line shows the time instance of the maximum current during the sheath collapse of this local HF cycle near the grounded electrode. After mirroring it in space with respect to the discharge center, this current will be exactly opposite to the current during the local sheath collapse of the HF cycle marked with the black dot, near the powered electrode. Thus, globally, the symmetry of the current density is still present. This can also be seen in figures 4(c) and (d), where the conduction currents at the three specific time instances, indicated by the dashed lines in figures 4(a) and (b), are shown (with times $t_1, t_2, t_3$, see figures 4(a) and (b), together with the displacement and ion conduction current densities at the time instance indicated by the magenta line (i.e. at time $t_1$). The magnitudes of the conduction currents corresponding to $t_2$ and $t_3$, i.e. at times of maximum current during the respective sheath collapse of the mirror cycles, are equal, whereas the conduction currents at $t_1$ are higher due to the presence of the LF current, which at this time has the same sign as the HF current. We can also see in figures 4(c) and (d) that neither the displacement nor the ion conduction current are important in the ambipolar regions.

To understand the reasons behind the differences in the discharge dynamics at low and high pressure, figure 5 shows the spatio-temporal distribution of the electron density, $n_e$, and the ionization source function, $S_{\text{ion}}$, for 4 Pa (panels (a) and (b)) and 30 Pa (panels (c) and (d)), respectively.

The spatio-temporal electron density profile for 4 Pa (figure 5(a)) shows a relatively narrow bulk region, in accordance with figure 3(a). The motion of the sheath, indicated by the white lines (calculated according to Brinkmann’s criterion [64]) is characteristic of dual-frequency discharges [37]: the local high-frequency sheath width is large whenever the low-frequency sheath is small, and decreases when the LF sheath is fully expanded. This ultimately leads to the phenomenon of frequency coupling, where the mean ion energy and flux cannot be independently controlled in a dual-frequency discharge, as the flux, determined by the HF-sheath expansion, depends on the LF-sheath width. The traditional explanation for this phenomenon is largely based on an assumption of a hard wall-like sheath: as the LF sheath expands, it reaches a region of high ion density, which the local HF sheath cannot penetrate as much as during a small LF sheath where the ion density is low. Because, in this model, the ionization rate depends on how fast the HF sheath can locally expand, which is proportional to $d/dt$, as this quantity is smaller when the LF sheath is fully expanded, the ionization caused by the local HF sheath near this electrode decreases. This is visible in figure 5(b), which shows the ionization source function, $S_{\text{ion}}$ for 4 Pa. The ionization rate is calculated in the simulation according to its definition, i.e. $S_{\text{ion}} = \langle \sigma_{\text{ion}}(v)n ne \rangle = \int d^3v \sigma_{\text{ion}}(v)n ne$, with $n_e = \int d^3v ne$. The distribution function is sampled along the trajectories of each charged particle traced, i.e. in each
Figure 5. Spatio-temporal distribution of the electron density $n_e$ (a) and (c), and the ionization source function $S_{\text{ion}}$ (b) and (d), for 4 Pa (upper row) and 30 Pa (lower row). The white lines indicate the sheath edges. Discharge conditions: $L = 25 \text{ mm}$, $(\phi_1, f_1) = (250 \text{ V}, 27.12 \text{ MHz})$, $(\phi_2, f_2) = (500 \text{ V}, 1.937 \text{ MHz})$.

timestep $\sigma_{\text{ion}} v n_e$ is calculated for each electron, and is added to the corresponding spatio-temporal bin of $S_{\text{ion}}$, weighted by the electron weight. The $S_{\text{ion}}$ is then properly normalized at the end of the simulation. The ionization maxima are observed at the time of the expanding HF sheaths, i.e. these are $\alpha$-peaks, whose maximum value decreases as the LF sheath expands. In this low-pressure case, due to the long mean free path, energetic electrons can reach the opposite sheath and ionize along their trajectory. This is the reason why there is significant ionization in the bulk region as well: energetic electrons accelerated during the local HF-sheath expansion near the grounded electrode can reach the local sheath edge near the powered electrode, and similarly at the other electrode. We note that the ionization profile has a somewhat more complex structure, especially during the time of maximum LF-sheath expansion at the powered electrode, due to the nonlocal character of the transport. There are altogether three distinct features: (i) ionization from energetic electrons accelerated by the opposing sheath, (ii) ionization due to electrons accelerated by the local sheath expansion (whose contribution is rather small) and (iii) ionization from reflected beam electrons (whose trajectories are also visible in figure 5(a)).

Increasing the pressure changes this situation: in figure 5(c), the electron density in the case of 30 Pa appears to increase significantly, decreasing the maximum of the LF-sheath width and leading to a wide bulk region. Similarly to the low-pressure case, whenever the LF sheath is close to its full expansion, the local HF sheath shrinks. Due to the high pressure and the wide bulk region, the effect of the frequency coupling is more easily seen in the ionization source function in figure 5(d). Whenever the local HF-sheath width is large, the corresponding ionization caused during its expansion is considerable. Thus, most of the ionization happens during the phase when the LF-sheath width is small, as whenever it is close to its full expansion (between 150 ns and 350 ns near the powered electrode), when the local HF-sheath width is small, the ionization becomes very low.

As shown in, e.g. [42], in low-pressure CCPs ionization is mainly caused by the ambipolar electric field, outside the local sheath edge. This is in contrast to the assumptions of the hard wall model. This means that the mechanism leading to frequency coupling should be reinterpreted within the framework of the Boltzmann term analysis. For this, the spatio-temporal dynamics of the ambipolar electric field have to be understood in detail. Figure 6 shows the potential of the total electric field between the local sheath edges at each electrode. The base point of the potential is taken to be the center of the discharge. The potential inside the sheath regions is not shown. Since the dominant electric field contribution is the ambipolar electric field, we can investigate the structure of $\Phi$ and draw conclusions about the dynamics of the ambipolar electric-field term.

In figure 6(a), for the case of 4 Pa, one can see that near the powered electrode, $\Phi$ monotonically increases as a function of
position in regions where the local LF-sheath width is small, which means that electrons can gain energy from the ambipolar field, which can lead to ionization. This is in accordance with figure 5(b), as the potential is higher in regions where the ionization is high. When the LF sheath is fully expanded at the powered electrode, the increase in the potential drop is much smaller, since the width of the ambipolar region is reduced. Furthermore, as the position is increased, the potential drop decreases due to the ambipolar electric field at the opposite electrode. This means that the ionization maxima in the bulk region in figure 5(b) are caused by the ambipolar field near the opposite (i.e. grounded) electrode, which can result in energetic electrons which, due to the low pressure, can reach the sheath of the powered electrode with a high energy. The high-pressure case, seen in figure 6(b), shows the consequences of frequency coupling more clearly: as in this case the bulk is wide enough so that electrons accelerated by the opposite sheath cannot reach the electrode with a high energy, i.e. the region of the nonzero ambipolar field at each electrode is spatially well separated, one can see that the potential is much smaller in the region where the LF sheath is fully expanded compared to a local HF-sheath expansion when the LF sheath is small. Based on the spatio-temporal profile of $\Phi$, the reason for frequency coupling lies in the fact that: (i) the ambipolar field, which accelerates electrons that can then ionize, is smaller in the ambipolar region when the LF sheath is close to its full expansion (denoted in figure 1 as region II); and (ii) the region of increasing potential, i.e. the nonzero ambipolar electric field outside the sheath is larger when the LF sheath width is small, thus electrons accelerated in this region can gain more energy (ambipolar regions I and III in figure 1). This means that if one understands the dynamics of the ambipolar electric field, then the mechanism of frequency coupling is understood together with the reason why the spatio-temporal average of the ambipolar power absorption shows nonmonotonic behaviour as a function of pressure (cf figure 2).

As the ambipolar electric field is given by $E_{\text{cv}} = -(T_{e}/en_e) (\partial n_e/\partial x)$, for a thorough understanding of the ambipolar field the parallel electron temperature, $T_{e}$, as well as the normalized electron density gradient, $\nabla n_e/n_e$, need to be investigated. Figure 7 shows the electron temperature for 4 Pa (a) and 30 Pa (c) and the normalized electron density gradient for 4 Pa (b) and 30 Pa (d), respectively.

One can see in figure 7(a) that the parallel electron temperature is high in regions of low electron density, when the local HF sheath is wide. As electrons are accelerated by the ambipolar field during the local HF-sheath expansion, they can effectively increase the electron temperature, increasing the field itself and leading to a self-amplifying mechanism resulting in a high temperature region, as reported in [42]. The temperature decreases in the bulk region, i.e. where the electron density is high. When the LF sheath is close to its full expansion, the sheath edge is in a region of high electron density, and thus the weaker local acceleration of electrons by the ambipolar field cannot increase the temperature effectively, leading to a lower electron temperature. However, there is a temperature increase in this time interval, i.e. between 200 ns and 350 ns in the sheath adjacent to the powered electrode, during the collapsing phases of the local HF sheath. This is caused by energetic electrons accelerated near the opposite electrode and reaching the instantaneous sheath width with a high energy. As the electron density between the electrode and the instantaneous sheath edge is low, the incoming high-energy electrons can increase the temperature, leading to an ‘inverse temporal asymmetry’, where the absolute value of the ambipolar power absorption is higher during sheath collapse than during sheath expansion; this in turn results in an attenuation of the spatio-temporally averaged ambipolar power absorption, as ambipolar cooling happens during sheath collapse. One can also see in figure 7(b), i.e. the normalized electron density gradient, that the temporal dynamics of the temperature profile are responsible for the attenuation of the ambipolar power absorption on time average at low pressure and, ultimately, for the presence of the frequency coupling, as the normalized electron density gradient does not change much as a function of time in the vicinity of the maximum LF-sheath width.

### Figure 6.
Spatio-temporal distribution of the plasma potential, $\Phi$, between the instantaneous sheath edges for 4 Pa (a) and 30 Pa (b). The reference point of the potential is taken to be the center of the discharge. The black lines indicate the sheath edges, the dashed grey lines are the positions corresponding to the maximum sheath widths. Discharge conditions: $L = 25 \text{ mm}$, $(\phi_1, f_1) = (250 \text{ V}, 27.12 \text{ MHz})$, $(\phi_2, f_2) = (500 \text{ V}, 1.937 \text{ MHz})$.
As the gas pressure is increased, the number of energetic electrons that are able to traverse the bulk region diminishes, and the corresponding attenuation effect will be smaller. This is the reason why, in figure 2, $P_{\nabla T}$ increases at low pressure reaching a maximum at $\approx 7$ Pa. Upon increasing the pressure to 30 Pa, the spatio-temporal profile of the parallel electron temperature (figure 7(c)) shows somewhat different behavior compared to the low-pressure case: although the locally increased electron temperature regions during local HF-sheath expansions are still present, the temperature increase during local HF-sheath collapse phases, when the LF sheath is close to its full expansion, is absent, as a result of the increased pressure and the correspondingly increased bulk length and shorter electron mean free path. Furthermore, the attenuation of the acceleration effect of the local HF-sheath expansion in a wide LF sheath is also visible, which is a manifestation of the frequency coupling effect. Due to the increased pressure, in the bulk region, the ohmic electric field is dominant. This is in accordance with figure 3(d), where there is a significant power absorption on the time average within the bulk region originating from ohmic heating. Electrons accelerated by the ambipolar field during a local HF-sheath expansion will gradually lose their energy via collisions, and thus their effect of increasing the local electron temperature diminishes as they fly towards the bulk. This is the reason for the peculiar shape of the electron temperature profile in figure 7(c) in the middle of the discharge: the electron temperature is constant in time and has a value of $\approx 3$ eV, which is a result of energetic electrons coming from either electrode ‘meeting’ in the center of the discharge. This is the reason for the local increase of $P_{\nabla T}$ in figure 3(d): there is a local increase in the electron temperature at the position of the discharge center, for which then $E_{\nabla T} \propto -\nabla T_\parallel < 0$, and thus electrons gain energy in this region. The electron density gradient profile in figure 7(d) looks similar to that at low pressure (figure 7(b)). The parallel electron temperature is lower at higher pressure, one can understand why $\Phi_{\nabla n}$ is smaller in figure 6(b) compared to figure 6(a): due to the higher pressure and the higher electron density, electrons accelerated by the ambipolar field during a local HF-sheath expansion cannot increase the parallel electron temperature effectively (as the higher the electron density is, the less effect a few energetic electrons can have on the electron temperature, which essentially measures the variance of the electrons’ velocity); thus, the aforementioned self-amplifying mechanism gets increasingly less pronounced as the pressure is increased. This will lead to two important consequences: (i) the electron temperature near the full expansion of the LF-sheath width will decrease compared to lower pressures, and thus the magnitude of the ambipolar electric field will also be smaller; and (ii) the spatio-temporal average of $P_{\nabla n}$ will also decrease with increased pressure, as the positive contribution coming from the self-amplifying increase in the electron temperature is attenuated. This is the reason why the ambipolar power absorption decreases monotonically as
the pressure is increased in figure 2. Thus, the basic mechanism of frequency coupling in the Boltzmann term analysis picture is fundamentally related to the spatio-temporal dynamics of the ambipolar electric field, and is thus quite different from the classical explanation.

4. Conclusions

The electron power absorption dynamics in a low-pressure capacitively coupled RF argon discharge excited by a dual-frequency waveform has been investigated using 1d3v PIC/MCC simulations in the pressure range between 4 Pa and 70 Pa based on the Boltzmann term analysis: a self-consistent, spatio-temporally resolved method based on the momentum balance equation of electrons.

Based on the spatio-temporal dynamics of the ambipolar power absorption, one can provide a novel interpretation of the mechanism of frequency coupling, which is not based on the hard wall model. The reason for the presence of the frequency coupling within the framework of the Boltzmann term analysis is twofold: (i) the ambipolar region, i.e. the spatio-temporal region where the ambipolar electric field has a significant magnitude, shrinks when the low-frequency sheath is close to its full expansion; thus electrons accelerated in this region will have a smaller energy than electrons at times when the low-frequency sheath is small, and thus these electrons will be less likely to ionize, which leads to the decrease in the ionization in this region. (ii) Because, at times of wide low-frequency sheath, the instantaneous sheath edge is in a region of high electron density, electrons accelerated in this smaller ambipolar region will be less likely to effectively increase the parallel electron temperature. This leads to a further decrease in the spatio-temporally averaged ambipolar electron power absorption in this region, as the self-amplifying mechanism where accelerated electrons can increase the parallel electron temperature, which then increases the ambipolar electric field itself, is less pronounced. This has important consequences on the overall power absorption dynamics.

Throughout the whole pressure range, ohmic heating was found to be the dominant power absorption mechanism on space and time average. The ambipolar power absorption monotonically increased at low pressure to reach a local maximum, and then monotonically decreased as the pressure was increased. The nonmonotonic behaviour of the spatio-temporally averaged ambipolar power absorption as a function of pressure can be understood based on the interpretation of the frequency coupling: at lower pressure (<7 Pa) the ambipolar power absorption showed a decreasing trend, as there can be energetic electrons created at the opposite electrode that can increase the temperature during a local high-frequency sheath collapse, thereby increasing the negative contribution of ambipolar power absorption during one high-frequency cycle. As the pressure is increased and the bulk widens (and the mean free path, λ, decreases), this mechanism gets increasingly less important, as initially, energetic electrons can no longer reach the opposite sheath with a high energy. This is the reason for the local maximum observed in the spatio-temporally averaged ambipolar power absorption at ≈7 Pa in the present case. As the pressure is further increased, the electron temperature decreases because, due to the higher electron density, the ambipolar electric field can no longer increase the parallel electron temperature effectively, which attenuates the self-amplifying mechanism that leads to regions of high electron temperatures. As the positive contribution to ambipolar power absorption decreases, so does the spatio-temporally averaged ambipolar power absorption as well, as the pressure is increased.

Acknowledgments

This work was funded by the German Research Foundation within the framework of the projects, ‘Electron heating in capacitive RF plasmas based on moments of the Boltzmann equation: from fundamental understanding to knowledge based process control’ (No. 428942393) and ‘Controlling the electron dynamics in radio-frequency driven micro plasma jets for efficient CO2 conversion’ (No. 445072286). Support by the DFG via SFB TR 87, Project C1, SFB 1316, Project A4 and by the Hungarian Office for Research, Development, and Innovation (NKFIH 134462) is gratefully acknowledged.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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References

[1] Lieberman M A and Lichtenberg A J 2005 Principles of Plasma Discharges and Materials Processing 2nd edn (New YorkWiley)
[2] Makabe T and Petrović Z L 2006 Plasma Electronics: Applications in Microelectronic Device Fabrication (Boca Raton, FLCRC Press)
[3] Fridman A A and Friedman G G 2008 Physics of Radio-Frequency Plasmas (CambridgeCambridge University Press)
[4] Chabert P and Braithwaite N 2011 Plasma and Chemistry (CambridgeCambridge University Press)
[5] Donnelly V M and Kornblit A 2013 J. Vac. Sci. Technol. A 31 050825
[6] Bi Z h, Liu Y x, Jiang W, Xu X and Wang Y n 2011 Curr. Appl. Phys. 11 S2–8
