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The role of the relative voltage and phase for frequency coupling in a dual-frequency capacitively coupled plasma

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Frequency coupling in multifrequency discharges is a complex nonlinear interaction of the different frequency components. An alpha-mode low pressure rf capacitively coupled plasma operated simultaneously with two frequencies is investigated and the coupling of the two frequencies is observed to greatly influence the excitation and ionization within the discharge. Through this, plasma production and sustainment are dictated by the corresponding electron dynamics and can be manipulated through the dual-frequency sheath. These mechanisms are influenced by the relative voltage and also the relative phase of the two frequencies. © 2008 American Institute of Physics. [DOI: 10.1063/1.2972117]

Discharges operated with more than one frequency are being targeted for improved control over the plasma and its parameters. However, in these discharges, complex coupling of the frequencies is an important matter and needs addressing. Here we investigate a capacitively coupled plasma (CCP) operated with two rfs simultaneously. Dual-frequency CCPs are used to achieve separate control of plasma density and ion impact energy onto the substrate.^{1–14} The plasma density is expected to be controlled by the higher frequency, while the ion energy is mainly determined by the lower frequency. This functional separation has been observed in certain parameter ranges of an asymmetric CCP with two frequencies applied to different electrodes.^{1,12} The electron dynamics in a sheath governed by both frequencies simultaneously is significantly more complex and not yet fully understood. There is, in particular, considerable lack of experimental investigations. Frequency coupling and nonlinear interactions of the frequency components are important aspects requiring attention. Here we investigate the influence of the external discharge parameters, such as voltage ratio and relative phase, with a view for manipulation of the electron dynamics and thus plasma production.

Phase resolved optical emission spectroscopy (PROES) provides access to the high energy electron dynamics in the discharge and thus insight into the ionization and sustainment mechanisms.^{15–20} In this article we use PROES to investigate a discharge specifically designed for investigations of the electron dynamics and sustainment mechanisms in a dual-frequency sheath.

An experimental setup is developed for controlled fundamental investigations. It is an asymmetric capacitively coupled rf driven discharge. The discharge system consists of one flat powered electrode with no counterelectrode. Two excitation frequencies, 2 and 14 MHz, are applied to the powered electrode. The chamber wall is grounded. A large sheath potential forms at the powered electrode and a very small potential forms at the grounded chamber wall. This design ensures minimal interaction of the two plasma boundary sheaths in the system. In this work the plasma is operated in neon, a noble gas with simpler plasma chemistry com-

pared to technologically employed molecular gases. The discharge is operated at relatively low powers and pressures below 10 Pa to avoid secondary electrons.¹⁹ Thus, the discharge operates in pure alpha mode where secondary electrons emitted from the electrode surface do not significantly contribute to excitation or ionization. An alpha-mode discharge is preferred since secondary electrons can be very dominant and make it difficult to decipher the sheath dynamics within the discharge.

PROES provides noninvasive access to the dynamics of high energy electrons with excellent spatial and temporal resolution on a nanosecond time scale. The optical emission from rf discharges exhibits temporal variation within the rf cycle. A detailed description of a typical PROES setup can be found in Ref. 19. In the following a brief description is given. The PROES setup consists of a fast, gatable, and intensified charge coupled device (ICCD) camera (PicoStar, LaVision) with an electronically tunable filter (VariSpec CRI) for spectral discrimination of the Ne 2p₁ emission line. The ICCD camera is synchronized with a signal from the rf generators powering the discharge. The absolute phase is calibrated using a voltage sensor installed at the powered electrode. Measurements with a defined gate width can be performed. In this work we use a gate width of 1 ns for investigations within both the 14 and 2 MHz cycles (rf period of 500 ns). A variable delay between the camera gate and the rf voltage allows for phase resolved measurements within both the low and high frequency rf cycles. The intensities measured in the camera gate time are integrated over many rf cycles for a certain phase setting to obtain good signal to noise ratio. The camera offers very high repetition rates, allowing the collection of photons originating from a particular rf phase in every low frequency rf cycle. From the measured phase resolved emission, the excitation $E_i(t)$ for the excited state i is determined by deconvoluting with the effective lifetime ($\tau_{\text{eff}}=1/A_{ik}$) of the excited state using the following:²⁰

$$E_i(t) = \frac{1}{n_0 A_{ik}} \left(\frac{dn_{\text{ph},i}(t)}{dt} + A_{ik} n_{\text{ph},i}(t) \right), \quad (1)$$

where n_0 is the ground state density, A_{ik} is the transition probability of the observed emission, and $n_{\text{ph},i}(t)$ is the num-

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FIG. 1. (Color online) Excitation (normalized) in front of the powered electrode in a dual-frequency CCP at 1 Pa for three different voltage conditions: (a) $V(14 \text{ MHz})=25 \text{ V}_{\text{rms}}$, $V(2 \text{ MHz})=125 \text{ V}_{\text{rms}}$, (b) $V(14 \text{ MHz})=125 \text{ V}_{\text{rms}}$, $V(2 \text{ MHz})=125 \text{ V}_{\text{rms}}$, and (c) $V(14 \text{ MHz})=125 \text{ V}_{\text{rms}}$, $V(2 \text{ MHz})=25 \text{ V}_{\text{rms}}$.

ber of photons per unit volume and time. Under the investigated low pressure conditions ($\leq 10 \text{ Pa}$), collisional de-excitation is negligible in comparison to the relatively high optical transition rate of the Ne $2p_1$ state.

Figure 1 shows the excitation measured from the phase resolved emission in front of the powered electrode at 1 Pa for three different voltage conditions: (a) with the 2 MHz voltage five times higher than the 14 MHz voltage, (b) with equal voltages, and (c) with the high frequency voltage dominating. Measurements at 10 Pa were also performed, and the results are similar to those at 1 Pa presented here. The phase axis shows one 2 MHz rf cycle. There are two noticeable differences in the figures. First, as more 14 MHz voltage is added, the sheath width decreases; this is due to the sheath width decreasing with increasing plasma density. Secondly, the high frequency excitation structures become more dominant as the high frequency component increases.

In Fig. 1(a) at a phase of about 70 ns, the sheath can be observed to expand from the electrode into the plasma bulk and along with it a strong excitation structure, emerging from the acceleration of electrons in the expanding sheath electric field. This excitation structure is followed by two pronounced but smaller excitation structures at a distance of around 100 mm from the electrode surface. They also propagate in the direction toward the bulk plasma. As the 14 MHz voltage component is increased, these minor excitation structures become more pronounced and the number observed increases. When the 14 MHz voltage component dominates the 2 MHz voltage, seven of these excitation structures can be identified. These correspond to the seven high frequency cycles that are within one low frequency 2 MHz cycle. The excitations can be explained through the acceleration of electrons during the phases when the sheath momentarily expands due to the high frequency during the low frequency cycle.

In Fig. 1(a), the low frequency dominates the sheath dynamics and electrons are accelerated into the plasma bulk during the phase of sheath expansion. As in traditional single-frequency CCPs, operated in alpha mode, electrons gain energy as the sheath expands into the plasma.¹⁵ Comparing Fig. 1(a) to a pure single-frequency 2 MHz plasma, the phase of the main excitation is similar. The main influence of the additional 14 MHz component is an enhancement in the excitation due to the simultaneous increase in both frequency components as opposed to the 2 MHz only. The

acceleration of electrons and energy gain is governed by the sheath velocity. The velocities of the sheath edge in Fig. 1, in a phase of 70 ns, can be estimated from the slope of the excitation trajectory as 5.3×10^6 , 4×10^6 , and $3.5 \times 10^6 \text{ ms}^{-1}$ for conditions (a), (b), and (c), respectively. This illustrates that the velocity of the sheath edge is governed by the sheath thickness—the larger the sheath, the faster its velocity in order to cover the greater distance in the same time.

With dual-frequency excitation electrons gain energy during the phases of increasing voltage, giving rise to multiple excitation maxima in each low frequency cycle. The velocity of these oscillations depends on the motion of the sheath edge. The spatial structure of the sheath is determined by the ion density profile in the sheath. Due to ion flux conservation in the sheath, the ion density drops toward the electrode. Examining the dependence within the low frequency cycle, the local ion density is determined by the phase of the low frequency component. For the same voltage change, the resultant sheath velocity depends on the local ion density at the sheath edge. The lower the ion density, the larger the distance covered by the sheath at the same time, resulting in higher sheath velocities and, hence, increased energy gain for electrons. This is particularly obvious in Fig. 1(c) where the maximum energy gain is actually where the sheath is at a minimum (first half of the low frequency cycle in Fig. 1). This is the origin of the high frequency coupling and nonlinear interaction of the two frequency components in a dual-frequency sheath.

As the high frequency component is increased in relation to the low frequency component, it becomes more important as regards the electron dynamics within the sheath. The main difference occurs at maximum sheath expansion when the relative sheath motion is slow (second half of the low frequency cycle in Fig. 1). During this phase the high frequency component can momentarily increase and briefly expand the sheath toward the plasma bulk. If the high frequency component is high enough, electrons can also be momentarily accelerated and gain enough energy for excitation, as in Fig. 1(c) during the second half of the low frequency cycle. However, the excitation will of course not be as large as during the phase when the sheath is rapidly moving closer to the electrode. The faster the sheath, the larger the excitation maximum. Therefore, in Fig. 1(c) even though there are quite pronounced excitation structures in the second half cycle,

FIG. 2. (Color online) Excitation in front of the powered electrode in a dual-frequency CCP for four different relative phases between the two applied frequency components, (a) 0° , (b) 90° , (c) 180° , and (d) 270° , for the same conditions as in Fig. 1(a).

they are not nearly as large as those during the first half of the low frequency cycle.

The spatial penetration of the energetic electrons can be rather complex. Inelastic collisions cause relaxation of energetic electrons. Depending on discharge conditions the penetration of electrons can also be strongly influenced by the phase of the bulk electric field in relation to the sheath electric field.^{21,22} If the two fields are strongly out of phase with each other, electrons can be retarded earlier in space limiting penetration. On the other hand with the right phase relationship between the fields, electrons can also be efficiently accelerated by both fields that penetrated far into the plasma bulk.

We have observed that under certain conditions a pronounced coupling between the two applied frequencies can occur, limiting the separate control over ion energy and flux. This coupling is particularly pronounced when both frequencies are applied to the same electrode or in electrically symmetric plasmas. It can also be expected that the phase relation between the two frequencies is important. Figure 2 shows a plot of the spatial excitation as a function of time for the same voltage conditions as in Fig. 1(a) where the low frequency voltage component dominates. As in Fig. 1 the excitation structures arise from the acceleration of electrons during the sheath expansion phase into the main plasma bulk region. The main difference in the plots is that the magnitude of the excitation in each maximum varies depending on the relative phase between the two applied frequency components. In Fig. 2(a), where a relative phase of 0° is attributed, there is one dominant excitation maximum followed by a series of smaller maxima. At a phase of 90° two of the maxima are very pronounced, and at a phase of 180° the phase of the most pronounced maximum is shifted in time to approximately 100 ns, compared to approximately 70 ns in Figs. 2(a) and 2(b). By tuning the relative phase of the two frequency components, resonances in the plasma can be obtained. The total sheath velocity determines the excitation structures. The motion of the sheath can be influenced by tuning the increase and decrease in the high frequency component to the acceleration of the low frequency component. For the case considered here the low frequency component dominates. Thus by tuning the relative phase of the high frequency component, the phase and amplitude of high frequency excitation structures during the expansion of the low frequency cycle can be manipulated. This is a very interesting aspect of frequency coupling between the applied frequency components in multifrequency discharge systems. Controlling the relative phase of the frequencies can allow

for manipulating the excitation dynamics and thus ionization and plasma sustainment and production. These mechanisms are governed by the nonlinear interaction of the frequency components and their influence on the sheath dynamics.

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