

The effect of secondary electrons on the separate control of ion energy and flux in dual-frequency capacitively coupled radio frequency discharges

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Dual-frequency capacitive discharges are used to separately control the mean ion energy, $\bar{\epsilon}_{\text{ion}}$, and flux, Γ_{ion} , at the electrodes. We study the effect of secondary electrons on this separate control in argon discharges driven at 2+27 MHz at different pressures using Particle in Cell simulations. For secondary yield $\gamma \approx 0$, Γ_{ion} decreases as a function of the low frequency voltage amplitude due to the frequency coupling, while it increases at high γ due to the effective multiplication of secondary electrons inside the sheaths. Therefore, separate control is strongly limited. $\bar{\epsilon}_{\text{ion}}$ increases with γ , which might allow an *in situ* determination of γ -coefficients. © 2010 American Institute of Physics. [doi:10.1063/1.3481427]

Radio frequency (rf) plasma sources have a wide range of applications in many high-tech areas. Optimization of the interaction of the plasma with the surrounding surfaces has motivated the development of plasma sources, which allow a separate control of the mean ion energy, $\bar{\epsilon}_{\text{ion}}$, and the ion flux, Γ_{ion} , at the electrodes. The most important of these discharges are (i) hybrid (rf-dc,¹ capacitive-helicon,² and capacitive-inductive³) sources and (ii) dual-frequency (df) capacitive rf discharges. The latter class of discharges can be divided into two categories: (i) “classical” df discharges driven at substantially different frequencies^{4–11} and (ii) electrically asymmetric sources driven by a fundamental frequency and its second harmonic with fixed, but adjustable phase shift between the driving frequencies.^{12–15}

Classical df discharges are most frequently used in industry and are typically driven by a voltage waveform, $V(t) = V_{\text{HF}} \cos(2\pi f_{\text{HF}} t) + V_{\text{LF}} \cos(2\pi f_{\text{LF}} t)$ with $V_{\text{LF}} > V_{\text{HF}}$. The idea to obtain separate control of ion properties in these discharges is the functional separation of both frequencies due to the substantial frequency difference ($f_{\text{HF}} \gg f_{\text{LF}}$). The high frequency (HF) voltage amplitude, V_{HF} , is assumed to sustain the plasma and, consequently, to control the charged particle density and the ion flux. The low frequency (LF) voltage amplitude, V_{LF} , is assumed to control the acceleration of the ions determining $\bar{\epsilon}_{\text{ion}}$, without affecting Γ_{ion} .

In previous particle-in-cell (PIC) simulation studies different results with regard to the effect of V_{LF} on Γ_{ion} were found, e.g., in Ar/CF₄/N₂ a complicated behavior of Γ_{ion} as a function of V_{LF} was observed by Georgieva and Bogaerts,¹⁶ depending on the applied frequencies. Neglecting secondary electrons Donkó found the ion flux to decrease as a function of V_{LF} in an argon discharge driven at 1+100 MHz at about 3 Pa,¹⁷ while Boyle *et al.* found it to remain constant at about 6.6 Pa.¹⁸ Booth *et al.* experimentally found Γ_{ion} to increase as a function of V_{LF} in a discharge operated at 2 and 27 MHz in a mixture of argon and oxygen at the same pressure.¹⁹ In the latter work γ is most likely relatively high due to oxi-

dized Si electrodes. In PIC simulations of electronegative CF₄ discharges²⁰ at $\gamma=0$ a similar effect of V_{LF} on the peak ion density in the plasma bulk, n_{peak} , and on Γ_{ion} was found as in argon discharges.¹⁷

Here, our aim is to resolve the mystery of these apparently contradicting results in argon discharges by systematically studying the effect of secondary electrons on the quality of separate control of ion energy and flux in a classical df discharge operated in argon at $f_{\text{HF}}=27.12$ MHz and $f_{\text{LF}}=f_{\text{HF}}/14 \approx 1.937$ MHz. The importance of secondary electrons in these discharges has been pointed out before,^{19,21} but their effect has never been quantified. Using a PIC simulation we investigate the effect of V_{LF} and γ (at constant V_{HF} and different neutral gas pressures) on Γ_{ion} and $\bar{\epsilon}_{\text{ion}}$ at the electrodes.

Our studies are based on an electrostatic PIC simulation complemented with Monte Carlo treatment of collision processes. The code is one-dimensional in space and three-dimensional in velocity space, the electrodes are infinite, plane, and separated by a distance $L=2.5$ cm. The gas temperature is 400 K and electrons are reflected at the electrodes²² with a probability of 20%. As secondary electrons may be accelerated inside the sheaths to a maximum velocity $v_{\text{max}} = \sqrt{2q(V_{\text{HF}} + V_{\text{LF}})/m}$, a very small time step ($\Delta t \leq \Delta x/v_{\text{max}} \sim 1$ ps for the conditions investigated) is required to fulfill the Courant condition. Here Δx is the division of the computational grid, m and q are the electron mass and the elementary charge. Time-synchronized PIC schemes are inefficient at this point, since the number of γ electrons is typically small, compared to the number of “slow” electrons; the tracing of the latter would allow much longer time steps. Therefore, in our code we treat fast and slow electrons as different species and use different time steps to fulfill the Courant condition. Electrons emitted from the electrodes as well as all electrons originating from ionization processes are initially treated as fast electrons and will be transferred to the group of slow electrons, if their energy falls below a threshold (15 eV) in the central region of the discharge.

Figure 1 shows n_{peak} , Γ_{ion} , and $\bar{\epsilon}_{\text{ion}}$ (Ar⁺ ions) as a function of γ and V_{LF} . The three columns of the figure corre-

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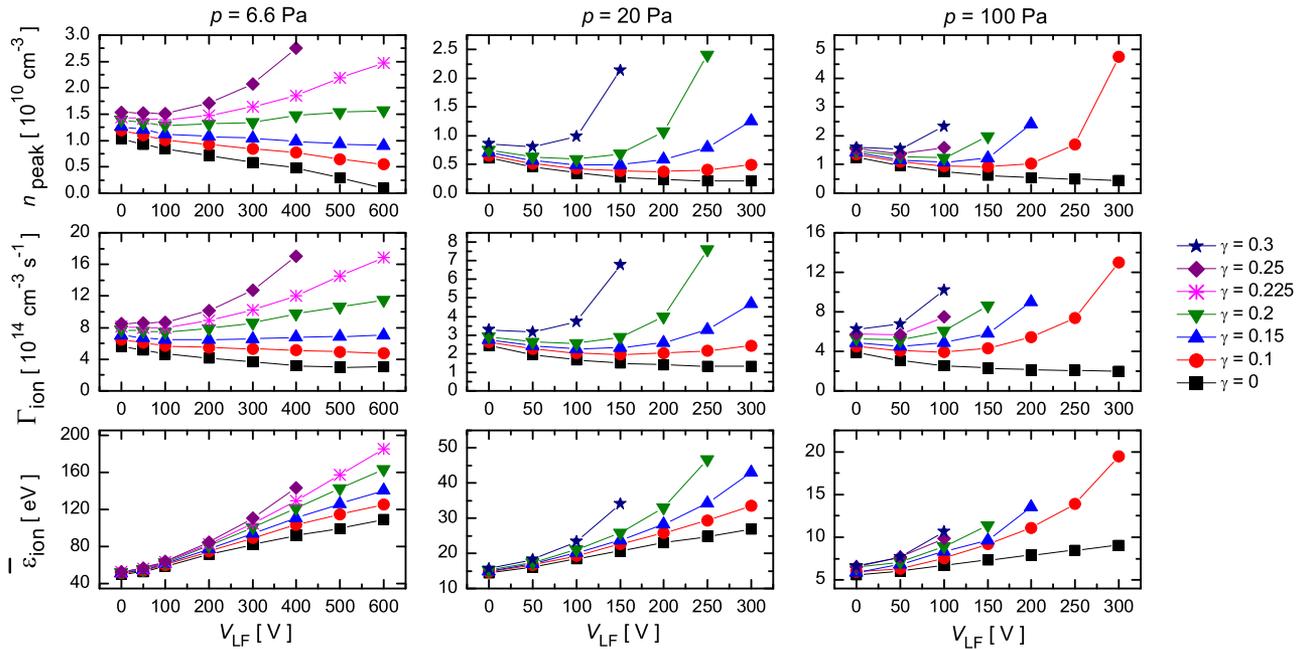


FIG. 1. (Color online) Peak ion density (n_{peak} , top row), ion flux (Γ_{ion} , middle row) and mean energy ($\bar{\epsilon}_{\text{ion}}$, bottom row) of Ar^+ ions reaching the electrodes as a function of the low-frequency voltage, at different pressures (6.6, 20, and 100 Pa) and secondary yields γ . $L=2.5$ cm, $V_{\text{HF}}=200$ V at 6.6 Pa, 100 V at 20 Pa, and 100 V at 100 Pa.

spond to different pressures, covering the range $6.6 \text{ Pa} \leq p \leq 100 \text{ Pa}$. The HF voltage amplitude at the different pressures is kept constant: $V_{\text{HF}}=200$ V at 6.6 Pa, 100 V at 20 Pa, and 100 V at 100 Pa.

Generally, the dependence of n_{peak} and Γ_{ion} on V_{LF} is similar.¹⁹ For a given LF voltage both quantities increase as a function of γ due to additional ionization caused by the secondary electrons.

For $\gamma=0$ we find the peak ion density and flux to decrease as a function of V_{LF} similar to previous results of Donkó.¹⁷ Under these conditions the coupling of both frequencies dominates the ionization.^{11,16,23–26} Figures 2(a) and 2(b) illustrate the effect of the frequency coupling on the spatiotemporal ionization rate, $S(x,t)$, within one period T of the fundamental frequency for $\gamma=0$ at 6.6 Pa. The white lines indicate the movement of the sheath edge adjacent to both electrodes. For $V_{\text{LF}}=0$ the discharge is operated as a single HF discharge in α -mode,²⁷ i.e., the ionization is dominated by highly energetic electron beams generated by the HF sheath expansion.^{25,26,28} By switching on the LF voltage the sheath edge is pushed away from the electrode into a region of higher ion density during a substantial fraction of T . During this time the sheath expands more slowly and, thus, the ionization due to electron beams generated by the expanding sheath is reduced. Finally, this causes the ion flux and peak ion density to decrease as a function of V_{LF} .

For high γ coefficients, n_{peak} and Γ_{ion} increase as a function of V_{LF} similar to previous results of Booth *et al.* for Ar/O_2 discharges.¹⁹ This is caused by the more effective multiplication of secondary electrons inside the sheaths at high V_{LF} . Increasing the LF voltage causes the sheaths adjacent to each electrode to become bigger for a given ion density profile. Thus, secondary electrons are multiplied more often inside the sheaths and stronger avalanches of γ -electrons are generated. This enhances the ionization and causes n_{peak} and Γ_{ion} to increase as a function of V_{LF} . The

choice of γ determines the number of secondary electrons generated at the electrode surface. Thus, a higher γ causes stronger avalanches and a stronger increase of n_{peak} and Γ_{ion} as a function of V_{LF} . As the mean free path for the secondary electrons decreases with increasing pressure, the generation of secondary electron avalanches is more effective at higher pressures. Thus, the increase of n_{peak} and Γ_{ion} is amplified at higher pressures. The effect of γ electrons on $S(x,t)$ at 6.6 Pa is shown in Figs. 2(c) and 2(d) for $V_{\text{LF}}=0$ V and $V_{\text{LF}}=500$ V, respectively. For $V_{\text{LF}}=0$ V a high γ causes a weak constant background ionization in the plasma bulk. For $V_{\text{LF}}=500$ V, however, it causes a strong background ionization in the bulk, which is modulated by twice the LF. This background ionization is caused by γ -electron avalanches, generated at each electrode, when the respective sheath is big. During these times secondary electrons are multiplied inside the sheath efficiently and strongly increase the total ionization in the discharge. The discharge is operated in a hybrid α - γ -mode.²⁷ For higher V_{LF} and/or higher γ the discharge will jump into a pure γ -mode, where secondary electrons dominate the ionization.²⁷ It is noted that with increasing γ values, above a certain LF voltage the number of superparticles diverges in the simulation (see Fig. 1) – physically this corresponds to a continuously growing charged particle density, which in an experiment would lead to arc formation.

Our results (Fig. 1) also show, that the choice of γ affects $\bar{\epsilon}_{\text{ion}}$: the mean ion energy increases as a function of γ . This is caused by a decrease of the sheath width as a function of γ due to the increasing ion density. Thus, at a given pressure and mean free path the ions undergo fewer collisions inside the sheaths and arrive at the electrodes at higher energies. This effect might be the basis of an *in situ* technique to determine γ -coefficients from the comparison of $\bar{\epsilon}_{\text{ion}}$ obtained from experiment and simulation.

In conclusion, there is only a small process window of particular discharge conditions in classical df discharges,

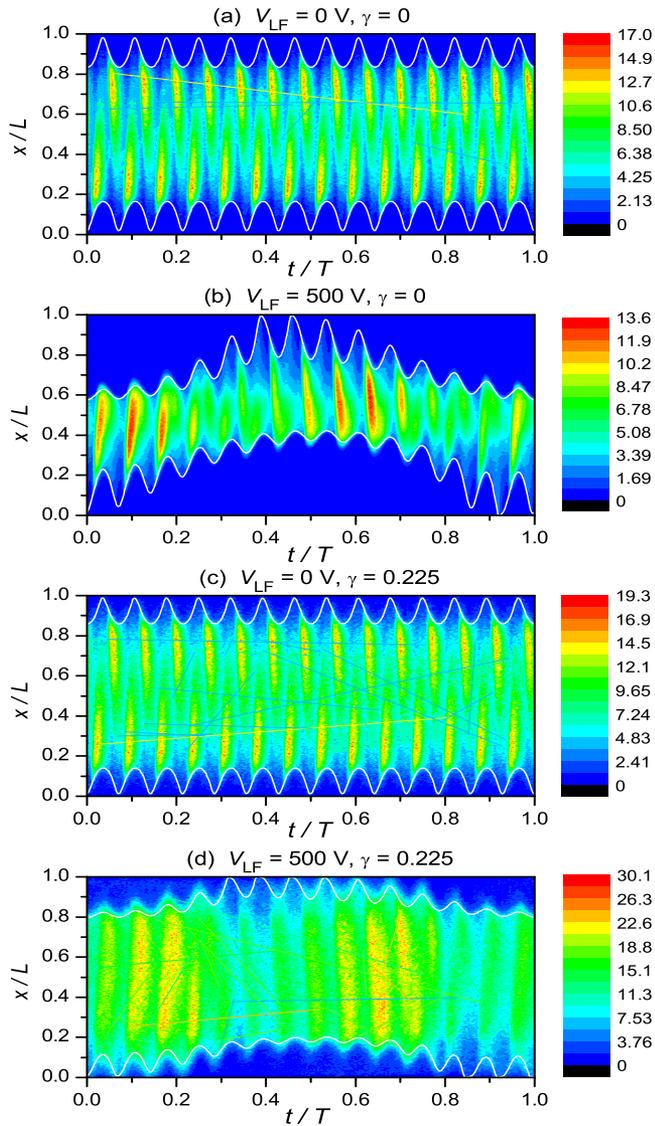


FIG. 2. (Color online) Spatiotemporal ionization rate within one period of the fundamental frequency for different combinations of V_{LF} and γ at 6.6 Pa (units are $10^{14} \text{ cm}^{-3} \text{ s}^{-1}$). The powered electrode is situated at $x=0$. $L=2.5 \text{ cm}$, $V_{HF}=200 \text{ V}$.

where a nearly constant ion flux independently of V_{LF} is ensured, e.g., for $p=6.6 \text{ Pa}$ γ should be within the range $0.1 \leq \gamma \leq 0.15$ (see Fig. 1). Within this process window the effect of secondary electrons on the ionization compensates the effect of the frequency coupling. Generally, however, the quality of the separate control is strongly limited and an alternative approach is required. Our results show that previous, apparently contradicting, results can be explained by the different discharge conditions in the respective works, particularly by the different γ -coefficients.

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