Theoretical and experimental study of ion flux formation in an asymmetric radio-frequency capacitive discharge

I V Schweigert\textsuperscript{1}, D A Ariskin\textsuperscript{1}, T V Chernoiziumskaya\textsuperscript{2} and A S Smirnov\textsuperscript{2}

\textsuperscript{1} Khristianovich Institute of Theoretical and Applied Mechanics, Siberian Branch, Russian Academy of Sciences, Novosibirsk 630090, Russia
\textsuperscript{2} St.-Petersburg State Polytechnical University, St. Petersburg 195251, Russia

E-mail: ischweig@itam.nsc.ru

Received 12 June 2010, in final form 6 November 2010
Published 7 January 2011
Online at stacks.iop.org/PSST/20/015011

Abstract

Ion flux formation in a capacitively coupled radio-frequency discharge in argon in axially symmetric chambers of different geometries is studied in experiments and by means of two-dimensional kinetic modeling by the particle-in-cell Monte-Carlo collisions method. A scaling exponent that relates the ratio of voltage drops within sheaths and the ratio of areas of driven and grounded electrodes $\delta S = A_{rf}/A_0$ is calculated for several types of geometrical discharge asymmetry. It is found that the scaling exponent has a maximum at $\delta S = 2.4$. The dc self-bias voltage rises linearly at $\delta S < 2.4$ and then becomes saturated. It is demonstrated that a change in $\delta S$ can substantially increase the ion energy on the electrode practically without disturbing the plasma parameters. The results of self-consistent calculations are in good agreement with experimental data.

1. Introduction

In various plasma technologies, the ion flux generated by a radio-frequency discharge strongly affects plasma–surface interaction. Ion bombardment results in anisotropy of etching processes, and affects the growth rate and the structure of films grown in the discharge. The applied voltage drop in the sheaths is substantial, and a considerable part of the energy imparted to the discharge can be spent on accelerating ions.

Processes affecting sheath formation and ion acceleration have been studied in detail by many researchers (see, e.g., [1]). The processes of ion treatment of the substrate surface are critically dependent on the energy and flux of the impinging ions. There are various alternative concepts to control these parameters independently. For example, recently a novel approach for achieving separate control of ion flux and energy in capacitively coupled radio-frequency (ccrf) discharges based on the electrical asymmetry effect was proposed by Heil \textit{et al} [2]. The electrical asymmetry effect is provided by the relative phase between two harmonic rf frequencies. In this case, in geometrically symmetric dual-frequency ccrf discharges, the generation of a variable dc self-bias was observed. In [3], this electrical asymmetry effect was verified using a particle-in-cell Monte-Carlo collision (PIC-MCC) simulation of a geometrically symmetric dual-frequency ccrf discharge operated at 13.56 and 27.12 MHz.

Another means of dc self-bias generation is the geometric asymmetry of discharge. In 1970, the significance of the relative size of the target area and the non-target area in contact with an rf sputtering discharge was discussed by Koenig and Maissel [4]. Coburn and Kay [5] measured the plasma potential for several values of a parameter $\delta S = A_{rf}/A_0$, where $A_{rf}$ is the target area and $A_0$ is the total grounded area in contact with the glow discharge. In these experiments, the target was kept fixed and the discharge was confined by a series of short Pyrex cylinders. The range of $\delta S$ covered by the cylinders varied from 0.29 to 0.09.

In plasma technologies, the discharge is practically always geometrically asymmetric because one of the electrodes is connected to the grounded walls of the discharge chamber, and some portion of the discharge current is spent there. In this case, the current density and, correspondingly, the drop in voltage near one electrode are greater than the corresponding values in the vicinity of the opposite electrode. Thus, there appears a dc self-bias voltage, which appreciably increases the...
The detailed measurements and analysis of the ion energy distributions at the grounded electrode of an asymmetrical ccrf discharge in argon were presented in [6] for a strongly asymmetrical case with $\delta S = 13$. Considering the mechanisms described in the theoretical model developed by Wild and Koidl [7], the double peaked ion energy distributions were explained. It was shown that the combination of the rf modulation of the sheath potential and charge exchange determines the structures of the ion energy distribution.

The dc self-bias formation in the sheaths in a rf discharge with non-equal electrodes was theoretically studied by Lieberman [8] for concentric spherical electrodes. In this geometry, the current density was uniform over each electrode due to the symmetry. Three electrode sheath models were considered: collisionless ions, collisional with constant mobility ions and a constant-ion cross-section collisional law. Assuming $\phi_p/(\phi_p - U_{bias}) = \delta S^q$, where $\phi_p$ and $\phi_p - U_{bias}$ are the electrode voltage drops, and using the continuity of the rf current flow, the scaling exponent $q = 2.21$ was obtained for typical rf materials processing discharges.

Analyzing the two-dimensional (2D) effects in the breakdown and plasma sustaining in the grounded slot and vessel around the symmetrical ccrf plasma in [9], the authors took $q = 2$. This value was found by Lieberman and Savas [10] for the cylindrical and coaxial geometry. For strong asymmetric discharge geometry, the 1D cylindrical model was used in PIC-MCC simulations in [11].

The goal of this work is a numerical and experimental study of the ion flux formation in a low-pressure ccrf discharge with different types of geometrical asymmetry. As the discharge has a complicated 2D geometry, the distribution of currents should be calculated with 2D numerical simulations. The sheath thickness and the distribution of the charged particle concentrations depend on the discharge operation mode determined by the current density [1, 12].

We perform 2d3V PIC-MCC simulations and vary the geometrical parameter of the chamber $\delta S$ from 2.17 to 3.9. We study the variation of the dc self-bias $U_{bias}$ from gas pressure and applied voltage. Assuming that the dependence of the sheath voltage drops from the electrode areas in the following form, $\phi_p/(\phi_p - U_{bias}) = \delta S^q$ (where $\phi_p$ is the plasma potential), we calculate the scaling exponent $q$.

This paper is arranged as follows. The experimental setup is described in section 2. The kinetic model for the 2D description of the ccrf discharge is given in section 3. The plasma parameters in chambers with different geometries are compared in section 4. The ion flux onto the electrode is analyzed in section 5. The calculated results for the self-bias voltage, plasma potential, and scaling exponent for different gas pressures and applied voltages are discussed in section 6. The conclusions are drawn in section 7.

2. Experimental setup

The electric parameters of a 13.56 MHz capacitive discharge in argon were measured in the experiments performed in two chambers with different configurations of the driven electrode. The gas-discharge chambers used are schematically shown in figure 1. A voltage is applied to the top electrode with an area $A_{top}$, while the bottom electrode with an area $A_b$ is grounded. In chamber A (see figure 1(a)), the effective area of the driven electrode is substantially smaller than the grounded electrode area ($\delta S = A_{top}/A_b < 1$), because the side surface of the discharge chamber is also subjected to a zero potential. With this ratio of the electrode areas $\delta S$, the discharge is visibly asymmetric at low gas pressures, and a potential drop in the sheath adjacent to the driven electrode prevails. The potential drop at the grounded electrode sheath is small and depends only weakly on the gas pressure and on the input power. This configuration does not allow the flux of high-energy ions from the discharge plasma to be studied, because the ion energy analyzer is usually mounted in the high-vacuum chamber behind the grounded electrode. Such a configuration of the driven electrode, however, allows measurements of the radial distribution of the plasma concentration and also the electron temperature by a moving Langmuir probe.

In chamber B, the driven electrode is supplemented with a cylindrical side part, which is shielded to prevent the breakdown on the side walls of the chamber (see figure 1(b)). In this configuration, it is only the bottom electrode surface that is grounded, and the ratio of the areas of the driven and grounded electrodes is $\delta S > 1$. In chamber B, in the asymmetric ccrf discharge with a prevailing potential drop in the grounded electrode, a sheath is formed, and it is possible to study the high-energy ion flux with the use of an energy analyzer.

The gas pressure in the experiments is varied from 6 to 70 mTorr. The radius of chamber A is $R = 15$ cm, and its
height is 13 cm. The discharge glows between the electrodes with a radius of 11 cm; the distance between the electrodes is \( d = 3.8 \) cm. The radius of chamber B \( R = 11 \) cm is equal to the electrode radius.

To measure the ion flux distribution (IFD) function over energy, there is an orifice 0.1 cm in diameter at the center of the grounded bottom electrode; this orifice connects the discharge chamber with the diagnostic chamber located below. The diagnostic chamber is evacuated independent of discharge chamber evacuation, and the pressure in the diagnostic chamber is below 1 mTorr. The diagnostic chamber contains a four-grid electrostatic energy analyzer of the confining field. We used this analyzer to register the energy distribution function of the ion flux moving from the discharge plasma toward the grounded electrode.

3. Kinetic model

The system of equations in a 2D model of a ccrf discharge with cylindrical symmetry includes the kinetic equations for electrons and ions (which are 3D in terms of velocity and 2D in space) and Poisson’s equation. The energy distribution function for electrons \( f_e(\vec{r}, \vec{v}) \) and ions \( f_i(\vec{r}, \vec{v}) \) are found from the Boltzmann equations,

\[
\frac{\partial f_e}{\partial t} + \vec{v} \cdot \frac{\partial f_e}{\partial \vec{r}} = -eE \frac{\partial f_e}{m \partial v_r} = J_e, \quad n_e = \int f_e \, d\vec{v}_e, \tag{1}
\]

\[
\frac{\partial f_i}{\partial t} + \vec{v} \cdot \frac{\partial f_i}{\partial \vec{r}} = -eE \frac{\partial f_i}{M \partial v_r} = J_i, \quad n_i = \int f_i \, d\vec{v}_i, \tag{2}
\]

where \( v_r, v_i, n_e, n_i, m \) and \( M \) are the electron and ion velocities, concentrations and masses, respectively; \( J_e \) and \( J_i \) are the collisional integrals for electrons and ions.

Knowing the energy distribution functions for electrons and ions, we can calculate the mean energy of electrons and ions,

\[
e_{e,i}(\vec{r}) = n_{e,i}^{-1} \int m_{e,i} v_{e,i}^2 f_{e,i} \, d^3v_{e,i}. \quad \tag{3}
\]

Poisson’s equation describes the electric potential distribution,

\[
\Delta \phi = 4\pi e \left( n_e - \sum_{i=1}^{N} n_i \right), \quad \vec{E} = -\frac{\partial \phi}{\partial \vec{r}}. \tag{4}
\]

The boundary conditions for Poisson’s equation are the voltage \( U = 0 \) on the grounded electrode and \( U = U_0 \sin(\omega t) + U_{bias} \) on the driven electrode. The self-bias voltage \( U_{bias} \) is calculated from the condition of a zero total current onto the grounded surfaces and surfaces with applied voltage.

System (1)–(4) is solved self-consistently by the particle-in-cell method with sampling of collisions by the Monte-Carlo method (PIC-MCC) [13]. The ccrf discharge operates in argon. The kinetics of electrons includes elastic scattering of electrons on atoms, excitation of metastable states and ionization. The cross sections of electron scattering for argon are taken from [14, 15]. The emission of secondary electrons from the electrodes due to bombardment by ions with the secondary emission coefficient \( \gamma = 0.1 \) is also considered. In our PIC-MCC calculations, we found that the contribution of the secondary electrons in the ionization process is visible. For example, for the case of \( U_0 = 360 \) V and \( P = 70 \) mTorr, the maximum of the electron concentration \( n_e = 1.2 \times 10^{10} \) cm\(^{-3}\) for \( \gamma = 0 \) and \( n_e = 1.41 \times 10^{10} \) cm\(^{-3}\) for \( \gamma = 0.1 \)

4. Comparison of discharge parameters in chambers of different geometries

Let us consider the plasma parameters obtained in the experiment and in the self-consistent numerical solution of system (1)–(4). The discharge operates in chambers of
Figure 4. Distributions of the electrical potential in chambers A (a), (c) and B (b), (d) for $P = 70$ mTorr and $U_0 = 155$ V. The potential profiles in (c), (d) are shown for $r = 0$.

different geometries (see figure 1), with a fixed loaded power of 10 W, $P = 5$, 30 and 70 mTorr, and $\gamma = 0.1$. Chambers A and B have different ratios of the areas of the driven and grounded electrodes $\delta S$, because the voltage is applied only to the top electrode in chamber A ($\delta S < 1$) and to the top electrode and the side walls of the chamber ($\delta S > 1$) in chamber B.

The calculated distributions of the electron concentration $n_e$ in the chambers are shown in figure 2 for $P = 70$ mTorr and $U_0 = 158$ V. The electron concentration at the center of the discharge gap is $4.9 \times 10^9$ cm$^{-3}$ in chamber A and $4.5 \times 10^9$ cm$^{-3}$ in chamber B. In chamber A, the concentration $n_e$ decreases with distance from the axis of symmetry toward the side wall. In chamber B, there is a second peak of the plasma concentration near the electrode edge owing to enhanced ionization.

Figure 3 shows the distribution of the electron energy $\epsilon_e$. The energy of the electrons changes from 2.9 to 3.5 eV over the volume of chambers A and B. Note that the maximum energy of the electrons in chamber B is observed between the grounded bottom electrode and the side walls, which is under a voltage. The ionization rate here, however, is not very high, because the gap length is smaller than the characteristic length of ionization by electrons.

The distribution of the electric potential $\phi$ in figure 4 demonstrates the appearance of the self-bias voltage induced by the difference in the areas of the driven and grounded electrodes. Figures 4(a) and (c) show the distribution of $\phi$ for chamber A, where the voltage is applied to the top electrode with a smaller area; in this case, the self-bias voltage is $-100$ V. Figures 4(b) and (d) refer to chamber B where the grounded bottom electrode has a smaller area; the potential drop here is 110 V.

Let us compare the measured and calculated radial distributions of the electron concentration and energy at the discharge gap center. Figure 5(a) shows the measured and calculated concentrations of electrons in chamber A for different gas pressures. The calculated profiles of $n_e$ in chambers A and B are plotted in figure 5(b). In chamber A, which has a greater radius, the plasma concentration monotonically decreases toward the side wall. An increase in the electron concentration near the edge of the bottom electrode is observed in chamber B. As a whole, the change in the chamber geometry has a weak effect on the plasma concentration for gas pressures ranging from 15 to 70 mTorr. With increasing pressure, the plasma concentration increases in both chambers from $1.8 \times 10^9$ to $4.5 \times 10^9$ cm$^{-3}$.

Figure 6 shows the radial profiles of the electron temperature $T_e = 2/3\epsilon_e$ at the center of the discharge gap for different gas pressures. The mean electron temperature at $P = 30$ mTorr, which is measured near the electrode edge, agrees well with the numerical data.

The numerical and experimental data in figures 5 and 6 are in good agreement and show that the discharge mode and its parameters remain almost the same in both chambers. Because of the discharge asymmetry, however, the potential drop in chamber B is substantially greater near the grounded electrode, which makes it possible to form a high-energy ion flux to the electrode and to control experimentally the energy distribution function of the ions.
5. Ion flux to the electrode

In the experiment, the IFD over the ion energy was measured in chamber B on the small grounded electrode. As the value of the applied voltage was not measured in the experiment, in the simulation we varied the applied voltage to obtain an IFD close to the experimental curve. The best fitting of the experimental and calculated IFDs was found at the voltage $U_0 = 285$ V for 15 mTorr and at $U_0 = 260$ V for 30 mTorr. Figure 7 shows the measured and calculated IFDs at the center of the bottom electrode for $P = 15$ and 30 mTorr in chamber B. The parameters of the discharge plasma for these IFDs are summarized in table 1. The input power in the experiment was $14$ W for $P = 15$ mTorr and $19.2$ W for $P = 30$ mTorr.

At gas pressures of 15 and 30 mTorr, the ions experience several charge exchange collisions as they cross the sheath; therefore, the IFDs in figure 7 have a series of peaks. The ion maximum energy of the ions is $E_{\text{max}} = 189$ eV (experiment) and 190 eV (simulation) at $P = 15$ mTorr. For a higher gas pressure of $P = 30$ mTorr, $E_{\text{max}} = 167$ eV (experiment) and 175 eV (simulation). The inset in figure 7 shows the calculated distribution of the potential $\phi$ at $r = 0$. The self-bias voltage is $U_{\text{bias}} = 108$ V for both values of the gas pressure. The maximum value of the plasma potential is $\phi_p = 191$ V for $P = 15$ mTorr and 175 V for $P = 30$ mTorr. The maximum ion energy $E_{\text{max}}$ is approximately equal to the plasma potential with respect to the grounded electrode, because the characteristic time of the ion crossing the sheath is much larger than the discharge period.

The calculated distribution of the charge characterizing the sheath width near the electrodes and chamber walls for $U_0 = 260$ V and $P = 30$ mTorr is shown in figure 8. In the case considered, the sheath width is 1 cm for the grounded electrode and approximately 0.5 cm for the surface with an applied voltage. At a pressure of 30 mTorr, the mean free path of ions with respect to the charge exchange collisions with neutral atoms is approximately 2 mm; therefore, the ions participate in several collisions while crossing the electrode sheath.

Let us consider the IFD along the electrode surface. Figure 9 shows the distribution of the ion flux density along the surface of the bottom electrode and between the electrodes at $r = 0$. The ion flux density along the bottom electrode remains almost unchanged at $0 < r < 8$ cm and then decreases as the electrode edge is approached (see figure 9(a)). It is of interest to note that the ion flux density onto the top electrode is little different from the ion flux onto the bottom electrode, though the ion energy on the top electrode is considerably lower.

6. Self-bias voltage for various discharge geometries

Let us consider the self-bias voltage in B-like configuration chambers of different geometries. Figure 10 shows the calculated plasma potential relative to the grounded electrode $\phi_p$ and the self-bias voltage $U_{\text{bias}}$ as a function of the ratio of the areas of the driven and grounded electrodes $\delta S$. The ccrf discharge was calculated for chamber B with different...
Figure 7. IFD functions for 15 mTorr, $U_0 = 286$ V (a) and 30 mTorr, $U_0 = 260$ V (b) in chamber B (the experimental and numerical data are shown by crosses and circles, respectively). The inset shows the potential distribution at $r = 0$ for $P = 15$ mTorr (solid curve) and $P = 30$ mTorr (dotted curve).

Table 1. Self-bias voltage $U_{\text{bias}}$, plasma potential $\phi_p$, amplitude of applied voltage $U_0$, concentration of electrons at the center of the discharge gap $n_e$, and ion flux onto the bottom electrode $j_i$ for different gas pressures.

| $P$ (mTorr) | $U_{\text{bias}}$ (V) | $\phi_p$ (V) | $U_0$ (V) | $n_e$ (cm$^{-3}$) | $j_i$ (s$^{-1}$ cm$^{-2}$) |
|-------------|----------------|--------------|----------|------------------|------------------|
| 15          | 109           | 189          | 285      | $2.4 \times 10^9$ | $2.3 \times 10^{14}$ |
| 30          | 108           | 191          | 260      | $3.4 \times 10^9$ | $3.0 \times 10^{14}$ |

parameters: (1) $R = 11$ cm, $d = 3.8$ cm and $\delta S = 1.72$; (2) $R = 7$ cm, $d = 3.8$ cm and $\delta S = 2.17$; (3) $R = 11$ cm, $d = 7$ cm and $\delta S = 2.33$; (4) $R = 6$ cm, $d = 8$ cm and $\delta S = 3.9$. The bottom electrode and the chamber wall are separated by a 0.5 cm gap. It is seen in figure 10 that the plasma potential increases linearly with increasing relative area of the driven electrode for $\delta S < 2.3$. In turn, $U_{\text{bias}}$ increases faster with $\delta S$ and approaches the plasma potential value at $\delta S > 4.6$. A decrease in the gas pressure leads to an insignificant increase in the plasma potential relative to the grounded electrode and the self-bias voltage.

In our calculations, we studied the dependence of the voltage drop in the electrode sheaths on the electrode area ratio. For a symmetric discharge, the ratio of the areas of the driven and grounded electrodes is $\delta S = 1$, and the ratio $\phi_p / (\phi_p - U_{\text{bias}})$ is also equal to unity. Using the results in figure 10, we obtain a scaling exponent $q$ for the expression relating the voltage drop on the electrode sheaths to the area of the electrodes in an asymmetric ccrf discharge $\phi_p / (\phi_p - U_{\text{bias}}) = \delta S^q$. Figure 11 shows the behavior of the exponent $q$ for different pressures and voltages as a function of the geometric parameter $\delta S$. Note that the curve for $q$ obtained by 2D kinetic calculations is a non-monotonic function, which has a maximum at $\delta S = 2.4$: $q_{\text{max}} = 1.7$ for $P = 15$ mTorr, $q_{\text{max}} = 1.7$ for $P = 30$ mTorr, and $q_{\text{max}}$ varies from 1.8 to 2.1 for $P = 70$ mTorr. Note that in an asymmetric 2 MHz capacitive discharge, the value of $q$ obtained in PIC-MCC simulations in [16] also varies from 1.6 to 2.1.

We compared our PIC-MCC results for the A-type chamber with the experimental data from [5] and found good
agreement. In the A-type chamber with geometric parameter $\delta S = 2.1$, the measured plasma potential is $\phi_p = 37$ V and the voltage drop on the small electrode is $(\phi_p - U_{\text{bias}}) = 147$ V (see figure 4 [5]). Our calculated values of voltage sheath drops are $\phi_p = 38$ V and $(\phi_p - U_{\text{bias}}) = 138$ V for $\delta S = 2.3$ (see figure 4(c)).

Figure 12 shows the calculated IFD over the ion energy on the small electrode for different geometric parameters $\delta S = 1.72, 2.3$ and 3.9. With increasing discharge asymmetry, the plasma potential relative to the smaller electrode increases, which results in a greater maximum energy of the ions.

7. Conclusions

The formation of an ion flux in a low-pressure asymmetric ccrf discharge was studied in experiments and by means of kinetic numerical simulations. 2D simulations were performed by the particle-in-cell method with collision sampling by the Monte-Carlo method. In the experiment, the ion flux was studied by an energy analyzer placed behind the grounded electrode with an orifice in the middle. The ion density and electron temperature...
were measured by a Langmuir probe. Several reactors with different ratios of the areas of the driven and grounded electrodes were considered to study the effect of chamber geometry on the ion flux. The plasma potential relative to the grounded electrode and, therefore, the maximum energy of the ions were demonstrated to increase with increasing area ratio. The measured and calculated parameters of the plasma, such as the electron concentration and temperature, and also the ion energy distribution functions were in good agreement. We found the scaling exponent \( q \) for different degrees of discharge asymmetry, applied voltages and gas pressures. It was shown that the dc self-bias rose practically linearly for \( \delta S < 2.4 \). With a further increase in \( \delta S > 2.4 \), the saturation of \( U_{\text{bias}} \) was observed. Thus the scaling exponent \( q \) attained a maximum value at \( \delta S = 2.4 \).

Acknowledgment

The authors gratefully acknowledge the support of this work by the Russian Foundation for Basic Research (grant No 08 – 02 – 00833a).

References

[1] Lieberman M A and Lichtenberg A J 1994 Principles of Plasma Discharges and Material Processing (New York: Wiley)
[2] Heil B G et al 2008 J. Phys. D: Appl. Phys. 41 165202
[3] Donko Z, et al 2009 J. Phys. D: Appl. Phys. 42 025205
[4] Koenig H R and Maissel L I 1970 IBM J. Res. Dev. 14 168
[5] Coburn J W and Kay E 1972 J. Appl. Phys. 43 4965
[6] Rusu I A, Popa G and Sullivan J L 2002 J. Phys. D: Appl. Phys. 35 2808
[7] Wild C and Koidl P 1991 J. Appl. Phys. 69 2909
[8] Liebermann M A 1989 J. Appl. Phys. 65 4186
[9] Lieberman M A, Lichtenberg A J, Kim S, Gudmundsson J T, Keil D L and Kim J 2006 Plasma Sources Sci. Technol. 15 276
[10] Lieberman M A and Savas S E 1990 J. Vac. Sci. Technol. A 8 1632
[11] Lee J K et al 2005 Plasma Sources Sci. Technol. 14 89
[12] Orlov K E and Smirnov A S 1999 Plasma Sources Sci. Technol. 8 37
[13] Birdsall C K and Langdon A B 1985 Plasma Physics Via Computer Simulation (New York: McGraw-Hill)
[14] Ivanov V V, Popov A M and Rakhimova T V 1995 Plasma Phys. Reports 21 548
[15] Lagushenko R and Maya J 1984 J. Appl. Phys. 59 3293
[16] Schweigert I V 2010 J. Phys. D: Appl. Phys. 43 305204