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Diagnostics and two-dimensional simulation of low-frequency inductively coupled plasmas with neutral gas heating and electron heat fluxes


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Diagnostics and two-dimensional simulation of low-frequency inductively coupled plasmas with neutral gas heating and electron heat fluxes

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This article presents the results on the diagnostics and numerical modeling of low-frequency (~460 kHz) inductively coupled plasmas generated in a cylindrical metal chamber by an external flat spiral coil. Experimental data on the electron number densities and temperatures, electron energy distribution functions, and optical emission intensities of the abundant plasma species in low/intermediate pressure argon discharges are included. The spatial profiles of the plasma density, electron temperature, and excited argon species are computed, for different rf powers and working gas pressures, using the two-dimensional fluid approach. The model allows one to achieve a reasonable agreement between the computed and experimental data. The effect of the neutral gas temperature on the plasma parameters is also investigated. It is shown that neutral gas heating (at rf powers ≥ 0.55 kW) is one of the key factors that control the electron number density and temperature. The dependence of the average rf power loss, per electron–ion pair created, on the working gas pressure shows that the electron heat flux to the walls appears to be a critical factor in the total power loss in the discharge. © 2002 American Institute of Physics.

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I. INTRODUCTION

In recent years, there has been a steadily growing interest in the “fine engineering” of low-temperature plasmas for numerous high-tech applications in low-temperature plasma-enhanced synthesis of nanocomposite and biocompatible materials, nanoscale assemblies, ultra-fine etching, patterning and microstructuring of semiconductor wafers in ultralarge scale integration large-scale manufacturing, micromachining, development of active elements in photonic devices, and many others.1 The fine engineering concept involves the development of sophisticated means of generating the plasma to comply with the specific requirements, and controlling the plasma species composition, number densities and fluxes towards surfaces being processed.2,3 In particular, the common requirements for the suitability of low-temperature plasmas for most high-tech applications are high uniformity of ions and active neutral species over the discharge cross section and volume, high product yield with low damage, process selectivity, and reproducibility.4,5 Sources of inductively coupled plasmas (ICPs) with external flat spiral coils6 do meet the above requirements and as such are widely adopted by the semiconductor industry as reference plasma reactors for microelectronic circuitry fabrication.

In applications, the operating conditions of ICPs can vary over broad ranges, e.g., the plasma needs to remain fairly stable when the gas feedstock pressure varies from a few mTorr to a few atmospheres. However, low-pressure operating regimes still remain favorable for most of the existing applications of ICPs. It is remarkable that the rf power transfer efficiency in low-pressure ICP devices is usually very high thus making it possible to generate dense (n_i~8...
Most of the existing modeling and experimental efforts are focused on 13.56 MHz inductively coupled plasmas, widely used nowadays by the semiconductor and other industries. However, sources of lower frequency rf plasmas are gaining rapidly increasing attention due to a number of indisputable advantages. In particular, operating ICP devices with frequencies in the range 0.46–0.5 MHz has proved unexpectedly efficient in producing highly uniform, high-density plasmas in large volumes.9–16 Moreover, low-frequency rf plasma devices have great potential for upscaling9,10 and for controlling the electron energy distribution functions (EEDFs),16 rates of the major gas-phase reactions,17 optical emission intensity, and mode transition thresholds,18 as well as various thin film deposition13 and nanoassembly18 processes. This certainly makes the sources of low frequency ICPs (LF ICPs) attractive as prototypes of large-volume commercial plasma reactors.

To date, most modeling studies of the electromagnetic effects and particle/power balance in ICPs have been made using either the fluid model19–22 or the particle-in-cell effects and particle/power balance in ICPs have been made large-volume commercial plasma reactors.4,6–8

Most recently, it has been recognized that heating of the neutral gas in low-temperature ICP sources during plasma processing can result in notable distortions of the uniformity of the neutral number densities across the chamber leading to variations of the plasma parameters, composition, temperatures, and number densities of the reactive species, as well as chemical reaction rates.20 Similarly, gas heating strongly affects the discharge electrodynamics and particle kinetics in microwave surface wave sustained plasmas.30,31

Here, we aim at incorporating the neutral gas heating effects into the two-dimensional (2D) fluid simulation of low-frequency ICPs and verify the model experimentally using Langmuir probe and optical emission spectroscopy (OES) techniques. Thus, the key internal discharge parameters, such as the plasma density, electron temperature, and EEDF are examined by Langmuir probe diagnostics. The high-resolution OES technique is used to study the optical emission intensity (OEI) of the abundant plasma species in different ranges of the working gas pressure. The modeling work is based on an improved 2D fluid model for an argon plasma, accounting for the heating of the neutral gas and the electron heat flux to the walls. The computed electron/ion number densities, electron temperatures, and optical emission intensities of the excited argon species appear to be consistent with the experimentally measured values. The dependance of the average power loss per an electron–ion pair created on the working gas pressure is studied as well.

The paper is organized as follows. In Sec. II, we describe the experimental setup, ancillary equipment, and diagnostic tools. The basic assumptions, equations and boundary conditions of the 2D model are presented in Sec. III. In Sec. IV, the profiles of the plasma parameters are computed for different rf powers, working gas pressures and temperatures and compared with the results of the Langmuir probe measurements. A satisfactory agreement between the results of optical emission spectroscopy and the computed OEI profiles is also reported. The results obtained and pathways for further improvement of the model and experimental techniques are discussed in Sec. V. A brief summary of this work and the outlook for future research are given in the Conclusion, Sec. VI.

II. EXPERIMENTAL DETAILS

Experimental measurements have been carried out in the low-frequency (~0.46 MHz) inductively coupled plasma source described in detail elsewhere.12,15 The plasma is generated in a cylindrical, stainless steel vacuum chamber with inner diameter 2R = 32 cm and length L = 20 cm (Fig. 1). The chamber is cooled by a continuous water flow in between the inner and outer walls of the chamber. The top plate of the chamber is a fused silica disk, 35 cm in diameter and 1.2 cm thick. A 450 l/s turbomolecular pump backed by a two-stage rotary pump is used to evacuate the plasma chamber. The inflow rate and pressure of the working gas are regulated by a combination of a gate valve and MKS massflow controllers. The pressure is measured by an MKS Baratron capacitance manometer. The operating pressure of argon gas feedstock is typically maintained in the range $p_0 = 20–200$ mTorr. The global plasma parameters, such as the electron/ion number densities, plasma potential and effective electron temperature have been estimated by means of a time-resolved rf-compensated single Langmuir probe technique. The electron energy distribution function has been calculated using the Dryuvestein routine employing the second derivative of the Langmuir probe current-voltage characteristics.5 A number of holes in four rectangular side.
ports and in the bottom plate of the chamber allow one to insert the Langmuir probe into the plasma and move it in radial and axial directions. The optical emission from the ICP discharge has been collected using a light receiver mounted on different port holes and transmitted via the optical fiber to a SpectroPro-750 spectrometer (Acton Research Corporation) with the resolution of 0.023 nm. The OES of excited neutral and/or ionized argon atoms have been investigated in the wavelength range 350–850 nm. Further details of the Langmuir probe and optical emission intensity measurements can be found elsewhere.12,15 Due to the limited space of the article, the experimental results are presented in the computation sections to enable a direct comparison.

III. MODEL DESCRIPTION

A. Basic assumptions

Theoretically, the plasma chamber is modeled by considering a metal cylinder of the inner radius \( R \) and length \( L \), with a dielectric disk of width \( d \) and permittivity \( \varepsilon_d \) atop. The components of the electromagnetic field are calculated assuming that the chamber is uniformly filled by the plasma with the electron/ion number density equal to the spatially averaged plasma density \( \bar{n} \).

The main results of this article are relevant to the plasma with the plasma density \( n_p \) in the range \( 20–100 \) mTorr, where the contribution of nonlocal averaged plasma density with the electron/ion number density equal to the spatially averaged plasma density \( \bar{n} \).

The averaged plasma density is computed from the particle and power balance equations (see the following subsection) and substituted into the electromagnetic fields Eqs. (1)–(3), which are continuously updated when \( \bar{n} \) varies.

B. Electromagnetic fields

The components of the transverse-electric (TE) electromagnetic field in the chamber fully filled by the uniform plasma with the density \( \bar{n} \) are

\[
B_z^m = A \sum_{n=1}^{\infty} \alpha_{1n} \kappa_n^m \left( \frac{4}{\pi c R^2} \right)^{3/2} \frac{J_0\left(\kappa_n^m R\right)}{\kappa_n^m R},
\]

and

\[
B_r^m = A \sum_{n=1}^{\infty} \alpha_{1n} \kappa_n^m \left( \frac{4}{\pi c R^2} \right)^{3/2} \frac{J_1\left(\kappa_n^m R\right)}{\kappa_n^m R},
\]

and

\[
E_{\phi} = -\frac{\omega}{c} \sum_{n=1}^{\infty} \alpha_{1n} \kappa_n^m \left( \frac{4}{\pi c R^2} \right)^{3/2} \frac{J_1\left(\kappa_n^m R\right)}{\kappa_n^m R},
\]

where

\[
\xi_n^m(z) = \sinh\left(\gamma_n^m(L-z)/\cosh(\Gamma_n^m d)\cosh(\gamma_n^m L)\right),
\]

\[
\eta_n^m(z) = \cosh\left(\gamma_n^m(L-z)/\cosh(\Gamma_n^m d)\cosh(\gamma_n^m L)\right),
\]

\[
D_\phi(TE) = \Gamma_n^m \coth(\Gamma_n^m d) + \gamma_n^m \coth(\gamma_n^m L),
\]

\[
\alpha_{1n} = \left\{ \begin{array}{ll} \frac{8}{\pi} & \text{for } n = 1, \\ \left[\frac{\pi c R^2}{2} \right]^{1/2} & \text{for } n \geq 2 \end{array} \right.
\]

\[ J_0(\kappa_n^m R) \]

where \( \phi \) is the azimuthal angle, \( \Gamma_n^m = \left[(\kappa_n^m)^2 - \omega\varepsilon_d \right]^{1/2} \) and \( \gamma_n^m = \left[(\kappa_n^m)^2 - \omega\varepsilon_\infty \right]^{1/2} \) are the inverse rf field penetration lengths into dielectric and plasma, respectively.12 Furthermore, we have \( J_0(x) \) is a Bessel function of the jth order, \( J_0(\rho_1) = 0 \), \( \kappa_n^m = \rho_1 / R \), \( \varepsilon_d = 1 - \omega_p^2 / [\omega(\omega + i\nu_{ce})] \) is the dielectric constant of the uniform plasma and \( \omega_p = \sqrt{4\pi n_0 e^2/m_e} \) is the electron Langmuir frequency. Here, \( \nu_{ce} \) is the electron–neutral collision frequency for momentum transfer, \( A \) is a constant, and \( e \) and \( m_e \) are electron charge and mass, respectively.

The averaged plasma density is computed from the particle and power balance equations (see the following subsection) and substituted into the electromagnetic fields Eqs. (1)–(3), which are continuously updated when \( \bar{n} \) varies.

C. Particle and power balance

Since the column and inductive coils are fairly uniform in azimuthal direction, the problem is two dimensional and the plasma parameters depend on \( r \) and \( z \) only. It is assumed that the ion temperature \( T_i \) is equal to the temperature of the working gas \( T_g \). The latter was a variable parameter in the computations. The plasma is treated within the ambipolar model that assumes the plasma quasineutrality and the equality of electron and ion fluxes. The effect of negative ions is neglected so that the overall charge neutrality condition can be written as \( n_e = n_i = n \), where \( n_e \) is the electron number density.

Accordingly, the particle balance equation for the electrons or ions is

\[
\frac{\partial n}{\partial t} + \nabla \cdot (n \mathbf{v}) = 0,
\]

where \( \mathbf{v} \) is the electron or ion fluid velocity, and \( \mathbf{v} \) is the ionization rate. We recall that in the ambipolar diffusion-controlled regime

\[
\mathbf{v} \approx -\nabla (nT_e) / nm_e \nu_{in},
\]

where \( \nu_{in} = \sqrt{u_{ei} + u_{ei}/T_e} / \lambda \) is the ion–neutral collision frequency, \( m_i \) is the ion mass, and \( T_i \approx T_e \). Here, \( \lambda = 1/(n_i\sigma_{in}) \) is the ion mean free path, where we have used \( \sigma_{in} = 8 \times 10^{-15} \) cm\(^2\), \( u_{ei} = \sqrt{T_e / m_i} \), the average ion thermal velocity, and \( n_{ne} = q_p / T_e \) is the number density of neutrals, and \( T_e \) is the electron temperature. We note that in computation \( T_e \) was self-consistently derived from the set of hydrodynamic, plasma particle and rf power balance equations. On the other hand, in the experiment, \( T_e \) was measured through the averaged electron energy \( \langle E \rangle \) as \( T_e = 2/3 \langle E \rangle \), as required by the Dryuvesnut’s routine.

The rf power balance in the discharge is described by

\[
\frac{3}{2} n_e \frac{\partial T_e}{\partial t} + \nabla \cdot \mathbf{q}_{ex} = -n_e I_e + S_{ext},
\]

where \( I_e \) is the collision integral for the electrons, and \( \mathbf{q}_{ex} = -(5/2 - g_u) n_e \mathbf{T}_e \mathbf{v}_{ex} / (m_e c^2) \) is the heat flux density with \( g_u = (T_e / v_{ex}) \mathbf{v}_{ex} \mathbf{v}_{ex} / T_e \). The term \( S_{ext} \) denotes the Joule heating of the electrons by the rf field.
\[ S_{\text{ext}} = n v_{\text{e}} m_{\text{e}} U_{\text{osc}}, \]

where \( u_{\text{osc}} \approx \sqrt{\frac{eE_{\phi}^2}{\left(2m_{\text{e}}^2(\omega^2 + \nu_{\text{en}}^2)\right)^{1/2}}} \) is the time-averaged oscillation velocity of the electrons in the rf field. The equilibrium state corresponds to setting \( \delta_1 = 0 \) in Eqs. (4) and (5). Our approach is valid for fixed rf power absorption in the plasma column, with

\[ P_{\text{in}} = \int_0^L \int_0^R S_{\text{ext}} 2\pi r \, dr \, dz, \]

which also yields the constant \( A \) in Eqs. (1)–(3). The rate of the electron–neutral collisions \( \nu_{\text{en}} \) that depends on \( T_{\text{e}} \) has been determined using the elastic scattering rate coefficients.\(^{20}\)

Assuming that the excitation and ionization of the neutral gas goes mainly via the electron impact processes, one infers that the collision integral \( I_{\text{e}} \) is equal to the average power lost by an electron colliding with a neutral. Accordingly,

\[ I_{\text{e}} = (3m_{\text{e}}/m_{\text{n}}) T_{\text{e}} \nu_{\text{en}} + \sum_j \nu_j E_j + \nu^E, \]

where \( m_{\text{n}} \) is the mass of the neutral, \( \nu_j \) is the excitation rate from the ground state to level \( j \) with a threshold energy \( E_j \), and \( E^E \) is the ionization threshold energy. Stepwise ionization and excitation processes are not accounted in the present simulation.

For argon gas, the rates for ionization and excitation to the states \( 4s \) and \( 4p \) are\(^{34}\)

\[ \nu^I = 2.3n_N \times 10^{-8} T_{\text{e}}^{0.68} \exp(-E_i/T_{\text{e}}) \, \text{s}^{-1}, \]
\[ \nu^E = 5.0n_N \times 10^{-9} T_{\text{e}}^{0.74} \exp(-E_{4s}/T_{\text{e}}) \, \text{s}^{-1}, \]

and

\[ \nu_{4p} = 1.4n_N \times 10^{-8} T_{\text{e}}^{0.71} \exp(-E_{4p}/T_{\text{e}}) \, \text{s}^{-1}, \]

where \( n_N \) is in cm\(^{-3} \), \( T_{\text{e}} \) in eV, \( E_i = 15.76 \) eV, \( E_{4s} = 11.5 \) eV, and \( E_{4p} = 13.2 \) eV. It is notable that the above rate coefficients were calculated by assuming the EEDFs Maxwellian.\(^{34}\)

We now consider the boundary conditions for integrating Eqs. (4)–(5). Because of symmetry, the radial gradients of the electron temperature and density are equal to zero at the chamber axis (\( r = 0 \)). At the column edge (\( r = R \)) the radial component of the fluid velocity satisfies the well-known Bohm sheath criterion \( v_j(R, z) = \sqrt{T_j(R, z)} m_j^{-1/2} \). Similarly, at the side walls \( z = z_s \), where \( z_s = 0 \) or \( L \), we have \( v_j(r, z_s) = \sqrt{T_j(r, z_s)} m_j^{-1/2} \). Likewise, the boundary conditions for heat flow are\(^{13}\)

\[ q_{\text{e}}(R, z) = T_{\text{e}}(R, z) \left(2 + \ln(m_e/m_i)\right) x n(R, z) \sqrt{T_{\text{e}}(R, z)/m_i}, \]

and

\[ q_{\text{e}}(r, z_s) = T_{\text{e}}(r, z_s) \left(2 + \ln(m_e/m_i)\right) x n(r, z_s) \sqrt{T_{\text{e}}(r, z_s)/m_i}, \]

where \( q_{\text{e}} \) and \( q_{\text{e}} \) are the radial and axial components of the heat flux density, respectively.

The set of Eqs. (4)–(5) has been solved numerically. The details of the codes and numerical procedures can be found elsewhere.\(^{35,36}\) The profiles of the electron density, temperature and ion velocity are computed from Eqs. (4)–(5). The resulting electron density distribution is used to compute the spatially averaged plasma density \( \bar{n} \). Thereafter, the latter is substituted into Eq. (3) to obtain the azimuthal electric field \( E_{\phi}(r, z) \). The computation is initialized using profiles of \( n, v \), and \( T_{\text{e}} \) estimated from less accurate analytical or computational results. The computation goes through a number of temporal steps and is terminated when a steady state is reached.

IV. NUMERICAL AND EXPERIMENTAL RESULTS

A. Effect of rf power

We now report on the spatial profiles of the electron number density and temperature in the LF ICP obtained numerically using the 2D fluid model and experimentally using the Langmuir probe technique.

The calculated 2D profiles of the electron number density and temperature at \( p_0 = 28.5 \) mTorr and \( P_{\text{in}} = 612.4 \) W are shown in Fig. 2. One can see that the electron temperature is maximal near chamber top at \( r \approx 8 \) cm. The profile of \( T_{\text{e}} \) is linked to the spatial distribution of \( E_{\phi}(r) \) (3). Since the electron temperature is somehow elevated near the fused
silica window, the maximum of the plasma density is shifted approximately 3 cm upwards (z ≈ 7 cm) from the central cross section of the chamber center (z0 = 10 cm). We note that in the classical case of uniform distribution of T_e over the entire chamber, one would expect a cosine-like solutions for the plasma density, with the maximum at z = z0.5

The computed electron number densities and temperatures have been compared with the ones measured by the Langmuir probe, with the tip positioned at z = 5.6 cm and r = 4.0 cm. Figure 3 displays the computed and measured values of n_e and T_e as functions of the input rf power P_in. The computation has been carried out under two different boundary conditions for the electron heat flux:

(i) the electron heat flux towards the boundary is governed by Eqs. (6) and (7); and

(ii) the electron temperature gradient vanishes at the boundary (∇T_e = 0). This boundary condition has been commonly used in simulations alongside with Eqs. (6) and (7).19,33

From Fig. 3(a) one can see that the plasma density obtained using boundary conditions (i) is closer to the experimentally measured one. Indeed, for nonvanishing heat flux boundary conditions (i), in the rf power range of P_in < 800 W the computed plasma density almost fully recovers the experimental data [Fig. 3(a)]. As the power increases (P_in > 800 W), the simulation yields plasma densities somewhat higher than the measured ones. The discrepancy can possibly be attributed to consistent elevation of the electron temperature at higher power levels [solid lines with circles in Fig. 3(b)], which cannot be reflected by the ambipolar-diffusion particle loss model adopted in our simulation [dashed and dotted curves in Fig. 3(b)], where a power-independent value of T_e is normally the case.5 It is thus natural to expect that the particle loss in the discharge in question transit to other, than the ambipolar-diffusion controlled, mode, with higher T_e (and hence, thermal motion power loss), and lower n_e at fixed rf power levels. Further evidence and discussion on the possible particle loss mode conversion follows in Sec. IV B.

We emphasize that the best agreement with the experiment is achieved by carefully accounting for nonvanishing electron heat fluxes, thus making the boundary conditions (i) more appropriate for modeling inductively coupled plasmas at elevated powers. Specifically, application of (ii) instead of (i) would lead to higher values of the electron number density. Physically, the electron heat fluxes are capable of removing a part of the rf power, that could have otherwise been gainfully used for additional electron-ion pairs creation in the plasma bulk.

Variations of rf power apparently affect the distribution of the electromagnetic fields, which is depicted in Fig. 4. In Figs. 4(a) and 4(b) the axial and radial profiles of the computed electric field E^p_φ are shown for different values of the input power. One can see from Fig. 4 that the electric field increases near the chamber top and decreases more rapidly in the z direction when the input power rises. This is consistent with the well-known dependence of the rf field penetration length on the plasma density.32 It is notable that at z > 5 cm the field component E^p_φ is much smaller than at z = 0. We thus infer that at larger distances from the window the electron heat flux is a key factor in controlling the electron temperature.

The normalized axial profiles of the Rf electric field E^p_φ(z)/E^p_φ(z = 0) in the plasma [solid, dashed, and dotted curves in Fig. 4(c)] have also been compared with the evacuated chamber case [dash-dotted curve in Fig. 4(c)]. We note that the skin length λ_s is maximal in the evacuated chamber case and can be estimated from Eq. (5) of Ref. 32. In particular, for low-frequency ICP (wc5e3.83/R), the skin length in the evacuated chamber can be approximated as R/3.83.32 However, for large-area and higher-density discharges satisfying (R5e0.007), λ_s is inversely proportional to n_e [Eq. (7) of Ref. 32], which is consistent with the data in Fig. 4(c). One can also notice that, while the normalized vacuum and plasma fields are quite different, the rf field skin length in the established inductive discharge mode does not change much with power, which is a common feature of ICPs.

B. Effect of gas pressure

We now turn to the study of the effect of the working gas pressure on the plasma parameters and power loss in the
discharge. The radial profiles (at $z=6$ cm) of the electron number density and temperature computed for four different values of $p_0$ are presented in Figs. 5(a) and 5(b), respectively. For the same conditions as in Figs. 5(a) and 5(b) the normalized axial profiles of plasma density and electron temperature at $r=0$ are shown in Figs. 5(c) and 5(d), respectively. The plasma density in Fig. 5(c) is normalized to its value at $z=6$ cm, whereas the electron temperature is normalized to that at $z=0$.

It is clear from Fig. 5 that the working gas pressure strongly affects the plasma density and electron temperature. We note that the Langmuir probe data suggest that within the pressure range of our interest $n_e$ is proportional to the gas pressure for a given electron temperature. Hence, an increase of gas pressure results in elevation of the average plasma density in the plasma column and in diminishing of the electron temperature. It is also seen that the axial uniformity of the electron temperature is better at lower pressures. Furthermore, the nonuniform profiles of $T_e$ affect the electron density distribution in the chamber [Figs. 5(a) and 5(c)]. Indeed, the density peak shifts towards the chamber top as the gas pressure rises. At $p_0=5$ mTorr the peak is close to the discharge center ($z \approx 10$ cm), which is a clear indication of the ambipolar-diffusion controlled regime at low discharge pressures. However, at $p_0=50$ mTorr the maximum of the plasma density is shifted approximately 4 cm towards the chamber top. This can indicate a possible gradual onset of a different particle loss mode, similar to what has been reported in Sec. IV A for the elevated rf powers.

The gas pressure also controls the rf power deposition process by affecting the average power loss per electron–ion pair. To elucidate the role of the electron heat flux in the plasma column we have separated the average power loss per electron $\theta_e$ into the collisional $\theta_c$ and heat flux $\theta_f$ components, so that $\theta = \theta_e + \theta_f$. Here

$$\theta_e = \frac{1}{\pi LR^2 n} \int_0^R \int_0^L n(r,z) I_e(r,z) 2\pi r dz$$

and

$$\theta_f = \frac{1}{\pi LR^2 n} \int_0^R \int_0^L \mathbf{v} \cdot \mathbf{q}_e 2\pi r dz$$

$$= \frac{1}{\pi LR^2 n} \int_S q_{eos} dS,$$

where $S$ is the total surface area of the rf discharge, and $q_{eos}$ is the component of electron flux density perpendicular to the surface.

The dependence of $\theta_e$ and $\theta_f$ on the working gas pressure are displayed in Fig. 6(a). The relative contribution of the heat transport component to the total power loss per electron $\theta_f/\theta \times 100\%$ is shown in Fig. 6(b). One can see that the heat flux contribution to the total power loss varies from 40% at low pressures ($p_0 \approx 10$ mTorr) to 17% at 100 mTorr. It should be noted that both $\theta_e$ and $\theta_f$ decline with pressure, as does the contribution of the heat flux to the total power loss per electron. The results of Fig. 6 clearly confirm the importance of the power loss through the heat flux to the chamber walls, which also strongly affects the value of the plasma density (Fig. 3).

C. Effect of the neutral gas temperature

We remark that the discharge in question is powered with relatively high rf powers. Therefore, in the plasma regions remote from the water-chilled chamber walls, areas of elevated neutral gas temperature may appear. To study the effect of the neutral gas temperature on the plasma properties, $T_g$ was varied in computations. The radial electron density and temperature profiles calculated at $z=6$ cm, $p_0$...
In the plasma parameters as a decrease of the working gas density. This can best be understood by noting the apparent link $p_0 = n_N T_g$, which means that the fixed pressure conditions require that the neutral gas density, which enters the expressions for the most of the reaction rate coefficients, has to diminish when $T_g$ rises.

D. Distribution of excited species

We now examine the spatial distribution of the excited argon atoms in the $3p^55p$ configuration. The profile of the excited atom density $n^*(r,z)$ can be calculated from

$$n^*(r,z) = n(r,z) \nu^*(r,z),$$

(8)

where $\nu^*(r,z) = \nu_0^*(r,z) \exp(-U^* / T_e)$ is the excitation rate with $U^* \approx 14.5$ eV being the threshold energy for the excited level. Generally, $\nu_0^*$ is a slowly varying function of $T_e$. Thus, evaluating $n^*(r,z)$, one can assume that $\nu_0^*$ does not depend on $r$ and $z$.

The normalized profiles of the excited atoms for the two different sets of our experimental conditions are shown in Figs. 8(a) and 8(b). According to Fig. 8, the resulting spatial distributions of the excited argon species are governed by the electron density and temperature profiles. One can observe that the radial profiles of the excited atoms are hollow near the chamber top. Meanwhile, the maximum of the OEI shifts towards the chamber axis as the axial position $z$ increases.
Likewise, when the gas pressure grows, the ratio of \( n^*(r) / n^*_{\text{max}} \) decreases, where \( n^*_{\text{max}} \) is the maximal density of the excited species along the radius.

We note that the radial OEI profiles have been consistently related to the integral (along the chamber axis) of the optical emission collected by the optical probe via the collimator positioned in eight portholes in the chamber bottom plate. For this purpose, the resulting local density of the excited species Eq. (8) has been integrated along \( z \) direction. Further details of the OES setup and collection of the emission can be found elsewhere.\(^{15}\) The emission of the 420.07 nm argon line is due to the electron transitions from 5\( p \) onto 4\( s \) levels.\(^{39,40}\) The radial distribution of emission intensity is shown in Fig. 9(a), which reveals a good consistency of the computation and experimental results. Analogously, integrating \( n^*(r,z) \) in radial direction, the axial distribution of the intensity has been computed. Experimentally, the optical probe in this case was placed in seven available portholes in the side observation port of the chamber.

Figure 9(b) presents a comparison of the axial OEI profiles obtained experimentally and numerically. One can notice a remarkable agreement of the calculated emission intensities with the experimental data. However, a minor discrepancy can be seen in the radial profile in the vicinity of the discharge center. We should also note that the experiment reveals that the OEI dips (\( \approx 20\% \) less than the maximal value) near the chamber axis. The computation results, correctly following the trend, suggest less remarkable diminishing of the OEI near the chamber axis. The deviation of the computed OEIs from the experimental data is certainly within the accuracy of the model that assumes independence of \( n_0^* \) on \( r \) and \( z \).

Figure 10 displays the experimental data on variation of the optical emission spectra with the operating pressure. The experiment has been carried out in the pressure range of 26–80 mTorr (argon gas flow rates were varied from 4 to 84 sccm). The intensity of the selected argon lines is presented in Fig. 11. In the pressure range concerned it is seen that there is a consistent general trend of diminishing of the OEI with \( p_0 \). For computation of the dependence OEI (\( p_0 \)), the 420.07 nm line of neutral argon has been selected. This dependence has been computed for the same axial position (\( z = 9.6 \) cm) as in Fig. 11 and is shown in Fig. 12. In calculations, two different assumptions have been made. The first one assumes that \( n^*(r,z) \sim n_N \) and corresponds to the dotted curve in Fig. 11. The dashed curve is obtained assuming that \( n^*(r,z) \) does not depend on the density of neutrals. In the first case, the agreement between the theory and experiment...
is satisfactory for the gas pressures exceeding 60 mTorr. The latter assumption provides much better consistency of the computational and experimental data within the entire range of operating pressures.

V. DISCUSSION

Here, we discuss the main results obtained and limitations of the modeling and experimental approaches. Generally, the modeling and experimental results appear to be in a good agreement, especially the data for the electron number densities [Fig. 3(a)]. However, in certain cases discrepancies of the order of few tens of percents still remain [e.g., Fig. 3(b) for the electron temperature]. It is thus worthwhile to discuss possible reasons affecting the accuracy of the simulation and experiment.

One of the possible reasons is a higher-than-expected value of the neutral gas temperature in the proximity of the Langmuir probe, causing excessive overheating of the probe tip. In this case, emissive properties of the probe surface should be carefully accounted for. Although we did not systematically measure the neutral gas temperature in this set of experiments, our earlier results give an indication that, in the course of materials synthesis and processing in LF ICPs sustained with 1–2 kW rf powers, the gas temperature in the reactor chamber is maintained in the range of $T_g = 500–700$ K. The above range of the neutral gas temperatures was used in most of the computations in this work. However, we do not exclude a possibility of somehow higher, than 700 K, temperatures in the area of rf power deposition. The probe tip position was, in fact, quite close to the above area, which may have resulted in less accurate values of $T_g$ at the actual measurement position.

To verify the range of the gas temperatures in our experiments, we make a comparison with the available data from the GEC reference cell with $R = 6.5$ cm and $L = 3.1$ cm. At $p_0 = 10$ mTorr, as the total input power raised from 50 to 200 W, the temperature of neutrals increased from 500 to 820 K. At fixed rf input of 200 W, $T_g = 1100$ K was achieved at $p_0 = 30$ mTorr. The estimated rf power densities $W_{rf}$ in Ref. 41 ranged from 0.121 W/cm$^3$ at 50 W to 0.729 W/cm$^3$ at 200 W.
W/cm$^3$ at 300 W. In our experiments with the much larger chamber, variation of the input power from 600 to 1700 W reflected changes in $W_{rf}$ from 0.037 to 0.105 W/cm$^3$. Thus, comparison of the neutral gas temperatures at similar rf power densities (although at quite different experimental conditions including gas pressure) does confirm the reasonability of our results on $T_g$ in the processing chamber.

The other limitation of the modeling effort in this work is the apparent inaccuracy in the excitation/ionization rate coefficients calculated for Maxwellian EEDFs. Indeed, as our experimental measurements of the EEDF suggest (Fig. 13), the latter is not necessary is the case. Figure 13 presents the variation of the EEDF with input power [Fig. 13(a)] and comparison with the corresponding Maxwellian EEDF plotted using the same values of the electron number density and temperature as in the experiment [Fig. 13(b)]. One can see from Fig. 13(a) that the total area under the EEDF increases with rf power, which obviously confirms proportionality of the electron/ion number densities to the power absorbed by the plasma column. However, the power does not noticeably affect the shape of the curve. The EEDF maximum is slowly shifting towards higher values of the electron energy while $P_{in}$ increases. From Fig. 13(b) one can clearly observe that the measured EEDF is somehow different from the corresponding Maxwellian one. Specifically, the peak of the measured EEDF is higher and corresponds to larger values of the electron energy. On the other hand, the Maxwellian EEDF gives a larger number of high-energy electrons, with the energies exceeding 13–14 eV. From Fig. 13 one can notice that the measured EEDF has a shape similar to the Maxwellian shape, with the peak shifted toward higher values of the energy, which is peculiar to Dryvestein-like EEDFs. A possible factor that affects a remarkable deviation of the experimentally measured EEDF from a Maxwellian one is strong electric fields in the rf power deposition area. Also, notable divergence of EEDFs from Maxwellian ones in the skin layer of low frequency ICPs can be attributed to the ponderomotive effect.
It is notable that the electrons with higher energies play an important role in nonelastic electron-impact processes. Thus, the depletion of the electron number density should be accompanied by a decrease of the excitation and ionization rates, which inevitably results in a growth of the electron temperature should the gas pressure remain the same.

Another factor affecting relatively higher values of the measured electron temperature is a lower than conventional frequency of the rf generator. At 30 mTorr, the rate of the electron–neutral collisions is much larger than the rf frequency \( \nu_{\text{rf}} \gg \omega \). Hence, the electrons receive the energy from the rf field as if they are in a dc electric field, acquiring somehow higher thermal energy.

Furthermore, the electron–electron collision rate \( \nu_{ee} \) may not necessarily be large enough to “Maxwellize” the EEDF. It can be estimated as \( \nu_{ee} \sim n_e \sigma_{ee} \sqrt{T_e/m_e} \), where \( n_e \) is the electron number density, \( T_e \) is the electron temperature, and \( \sigma_{ee} \sim 2.9 \times 10^{-15} T_e^{-2} \text{ cm}^2 \) is the cross section of electron–electron collisions. For \( n_e \sim 5.0 \times 10^{11} \text{ cm}^{-3} \) and \( T_e \sim 2 \text{ eV} \), \( \nu_{ee} \sim 10^8 \text{ s}^{-1} \), which is the value of the same order of magnitude as the rates of inelastic collisions. 33

Meanwhile, the mean free path of electrons becomes larger at lower pressures. Hence, some part of the electron population receiving the energy from the rf electric field near the chamber top can affect the EEDF non-locally. In this case the solution of a nonuniform Boltzmann equation is required to obtain viable values of the plasma parameters. Finally, the discrepancy between the experiment and computation can certainly be attributed to the experimental uncertainties in the Langmuir probe signal acquisition and measurement techniques.

We emphasize that the best agreement between the computed and measured values of the plasma density [Fig. 3(a)] has been achieved in case of taking into account the heat flux onto the chamber walls (\( \nabla T_e \neq 0 \)). For nonvanishing heat flux boundary conditions, at \( P_{in} \sim 800 \text{ W} \) the numerically obtained value of the plasma density is almost the same as the experimental one. As the power increases (\( P_{in} > 800 \text{ W} \)) the calculated plasma density becomes larger than the one obtained in experiment. The discrepancy can be attributed to the observed increase of \( T_e \) in the subsequent power range [Fig. 3(b)]. Accordingly, the elevated electron temperature forces the plasma density to decrease should \( W_{rf} \) remain the same.

The electron number density calculated in case of the vanishing heat flux onto the chamber walls (\( \nabla T_e = 0 \)) appears to be \( 20\%–30\% \) larger when the one obtained assuming \( \nabla T_e \neq 0 \). It is consistent with the results of Fig. 6 suggesting that under prevailing experimental conditions the electron heat fluxes consume up to \( 20\%–30\% \) of the total power. Thus, not accounting for such a significant power loss channel results in higher, than in the experiment, values of \( n_e \).

The remarkable agreement between the experimental and numerical (dashed) curves in Fig. 12 suggests that when the OEI declines with the gas pressure, it is likely that the excitation rate \( \nu_{*}(r, z) \) indeed does not depend on the density of neutrals. Thus, we can presume that the increase of the working gas pressure results in pronounced elevation of \( T_g \). This tendency is consistent with the available experimental data. 42

Finally, knowledge on the neutral gas temperature and heat fluxes in plasma processing discharges is becoming a matter of outmost importance for a number of applications. In particular, recent results on growth of nanostructured silicon-based films have convincingly demonstrated the neutral gas temperature as a critical factor in management of hydrogenated silicon nanoparticles in \( \gamma \) (powder-generating) regime. 43

**VI. CONCLUSIONS**

Results on the diagnostics and numerical modeling of low-frequency (\( \sim 460 \text{ kHz} \)) inductively coupled plasmas are presented in this article. Comparisons are made between the experimental and numerical data, particularly for the electron density and temperature, which show reasonable agreement. Possible reasons for any discrepancies are discussed. A study of the influence of the gas pressure and temperature on plasma density and electron temperature shows that an increase of working gas temperature has the same effect as a decrease of gas pressure. The electron heat flux to the walls appears to be a factor in the total rf power loss, as is also the neutral gas pressure. Measurements were made of the spatial distribution of the optical emission intensities of the excited argon species in both radial and axial directions. These show good agreement with the calculated emission intensities.

The fairly accurate agreement between the numerical and experimental results confirms the viability of the 2D fluid discharge model in simulating the major parameters of low frequency inductively coupled plasmas. The model can further be improved by involving both local and nonlocal kinetic approaches, stepwise excitation and ionization processes, complex gas chemistries including radicals, molecular complexes, and negative ions, details of the process (e.g., substrate bias, power and mass transport in near-substrate areas), as well as the effects of the near-wall sheath/presheath areas.

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35. V. Lisovsky (private communication).