Mode operation of inductively-coupled argon plasmas studied by optical emission spectroscopy

S Iordanova and I Koleva
Faculty of Physics, Sofia University, BG-1164 Sofia, Bulgaria
E-mail: koleva@phys.uni-sofia.bg, snejana@phys.uni-sofia.bg

Abstract. Mode operation of inductively driven plasmas in argon gas at low pressures is studied by optical emission spectroscopy. The plasma source is a tandem type source with a driver and an expansion plasma region. The driver region of the discharge is in the classical form of a cylindrically shaped inductive discharge, with a coil positioned over a gas discharge tube. The inductively coupled plasma discharge is maintained at high frequency \( f = 27 \text{ MHz} \), applied power varied in the limits \( P = (20 – 700) \text{ W} \) and gas pressure in the range \( p = (8 – 260) \text{ mTorr} \). The Ar line intensity dependencies on the applied power injected into the discharge is analysed. With the power increase a mode transition of the discharge regime is not observed. The investigations on the discharge mode operation are supported by theoretical calculations based on kinetic modelling of main processes contributing to the line intensity behaviour.

1. Introduction
Inductively coupled plasmas (ICPs) or inductive discharges at low gas pressure exhibit two modes of operation: week capacitive discharges known as an E-mode and true inductive discharges known as a H-mode. These regimes have been observed and reported in the literature [1–6]. At low input rf powers the discharge operation is associated with an E-mode characterized by low electron densities and high electron temperatures. At higher input power, the discharge is operating in a H-mode. Operation of the discharge in one of the two regimes is also possible. The transition between these two modes, reminiscent of hysteresis, is marked at a certain critical value of the input current or power and it is connected with a large increase in the electron density and the optical emission. Experimentally the transition occurs at a single well defined coil current function of various discharge operational and geometrical parameters. The E-to-H transition appears at larger coil current than the H-to-E transition. In particular, the H-mode is stable compared to the E-mode even when both are allowed at a given coil current. A full quantitative understanding of the phenomena is still far from complete, and investigations on the matter are of general interest for the operation of ICPs discharges at low pressures.

The theoretical results, reported in the literature [4, 5, 7, 8], show that the hysteresis can be qualitatively understood in terms of electron power balance assuming that either the power dissipated or the power absorbed by the plasma electrons has nonlinear dependence on the electron density. In noble gases the E-to-H transition may be connected with the dynamics of the electron density and metastable density and competition between direct and stepwise ionization of the atoms by electron impact [7, 9].
Most of knowledge of the E-to-H transition is based on measurements of electrical properties such as coil current [2], power deposition, effective impedance, charge particle density and also on measurements of EEDF [3]. Investigation of the E-to-H mode transition using optical emission spectroscopy (OES) measurements in an argon rf discharge are reported in [1, 2, 6]. Observations on the dynamics of the spectral line intensities through the mode transition in ICPs discharges at low pressure are associated with mechanisms of direct and stepwise excitation. In the E-mode, the intensity of 1s₂ → 2p₁ transition at 750.4 nm is the highest, whereas the 1s₅ → 2p₉ transition at 811.5 nm and the 1s₅ → 2p₆ transition at 763.5 nm are the most intense in the H-mode of the discharge operation.

In this paper, we focus on the use of OES measurements to define the mode operation of investigated inductive discharge. The method is based on studying the line intensity dependency on the applied power. We use also theoretical calculations to infer argon gas discharge mode operation. The calculations, based on collisional-radiative (CR) model, aim to estimate the mechanisms responsible for the population and depopulation of the Ar excited states in the conditions of the applied power increase. The argon spectra, experimentally and theoretically obtained at applied power \( P = 200 \) W and gas pressure \( p = 26 \) mTorr, are in good coincidence.

2. Experimental set-up
The experimental set-up, shown in figure 1, consists of vacuum vessel composing of two parts: gas-discharge dielectric chamber where the plasma is produced (driver region), and a metal chamber providing space for the plasma expansion [10, 11]. The discharge is generated by using 27 MHz rf power supply coupled to the matching device. A nine turns copper coil is positioned tightly over a quartz cylindrical tube with internal and external diameters 4.5 cm and 4.9 cm, respectively, and length of 30 cm. The effect of the electrostatic field is minimized by Faraday shield. The plasma produced in the gas discharge tube expands and fills the metal cylindrical stainless steel chamber with internal and external diameters 22 cm and 23 cm, respectively, and length of 47 cm.

![Figure 1. Schema of the experimental set-up.](image1)

In the experiments, the high-frequency applied power is varied in the limits \( P = (20 – 700) \) W. Argon discharges are sustained at gas pressure in the range \( p = (8 – 260) \) mTorr.

The light coming from the first chamber is collected by an optical waveguide trough a hole in the copper shield, positioned at a distance of 0.5 cm from the end of the quartz tube and 3.5 cm from the end of the coil. The spectrometer system for recording atomic spectra radiation is presented in figure 2. This system, PC2000-UV-VIS-ISA Optic Spectrometer from Ocean Optics Inc based on a crossed Czerny-Turner configuration with no moving parts, grating with 600 groves/mm and a 50 \( \mu \)m entrance.

![Figure 2. Experimental arrangement for the optical emission spectroscopy measurements.](image2)
slit, provides detection of the argon emission spectra in the wavelength range 200 – 1000 nm. The detector is Sony ILX511, 2048 elements linear CCD-array silicon.

3. Experimental results

3.1. Spectra observation

Argon emission spectra are recorded over wavelength range (200 – 1000) nm from the first chamber as a function of applied power at \( p = 8, 26 \) and 260 mTorr. Line-of-sight integrated optical emission from the plasma is collected. The spectra are clearly dominated by atomic argon lines in the red region (690 – 900) nm, due to \( 1s - 2p \) transitions. The transitions and wavelengths of the intensive lines in the spectra are given in table 1. From 400 to 690 nm pronounced spectral lines are not detected.

According to the observations, the contribution in the light emission of highly excited states from \( 3p^56s, 3p^25p \) and \( 3p^4d \) configurations and also of ions is negligible. Spectra emitted from the driver region of the source in the spectral range (690 – 870) are presented in figures 3(a) and 3(b).

Table 1. Spectroscopic data for Ar transitions.

| Transition | Wavelength (nm) | Transition | Wavelength (nm) |
|------------|----------------|------------|----------------|
| 1 \( 1s_5 - 2p_9 \) | 811.5 | 10 \( 1s_2 - 2p_4 \) | 852.1 |
| 2 \( 1s_5 - 2p_8 \) | 801.5 | 11 \( 1s_5 - 2p_3 \) | 706.7 |
| 3 \( 1s_4 - 2p_8 \) | 842.5 | 12 \( 1s_4 - 2p_3 \) | 738.4 |
| 4 \( 1s_3 - 2p_7 \) | 772.4 | 13 \( 1s_2 - 2p_3 \) | 840.8 |
| 5 \( 1s_4 - 2p_7 \) | 810.4 | 14 \( 1s_5 - 2p_2 \) | 696.5 |
| 6 \( 1s_5 - 2p_6 \) | 763.5 | 15 \( 1s_5 - 2p_2 \) | 772.4 |
| 7 \( 1s_4 - 2p_6 \) | 800.6 | 16 \( 1s_2 - 2p_2 \) | 826.5 |
| 8 \( 1s_4 - 2p_5 \) | 751.5 | 17 \( 1s_2 - 2p_1 \) | 750.4 |
| 9 \( 1s_3 - 2p_4 \) | 794.8 |

The spectra observations reveal that at the whole ranges of the applied power and gas pressure the most intense spectral lines, with wavelengths 811.5 nm and 763.5 nm, are the same as those observed in [1] at the H-mode of the discharge operation. The spectral line intensity at 750.4 nm and 751.5 nm is smaller.

Figure 3. Emission spectra of inductive discharges in argon gas at \( P = 500 \) W and \( p = 8, 26 \) and 260 mTorr in (a) and at \( p = 26 \) mTorr and \( P = 90, 200 \) and 700 W in (b).

Some intense argon lines, with low oscillator strength, are represented as a function of the injected power at gas pressures \( p = 8 \) mTorr and \( p = 260 \) mTorr in figures 4(a) and 4(b), respectively. The
arrows show the behaviour of line intensities with the power increase and decrease. The line intensities change monotonically with the power and mode (E-to-H and H-to-E) transition in the discharge regime operation is not observed.

![Graphs showing line intensities with power for two different gas pressures](image)

**Figure 4.** Evolution of argon lines with low oscillator strength as a function of the applied power at $p = 8$ mTorr in (a) and at $p = 260$ mTorr in (b).

### 3.2. Theoretical calculations

The theoretical calculations, made in this study, aim to predict the line intensities behaviour observed by OES measurements. The calculations, based on the CR model [12], enable to estimate the relative contribution of the mechanisms responsible for the population and depopulation of the first four $1s_{5/2}$, (two metastable ($1s_2$, $1s_3$) and two resonant ($1s_4$, $1s_5$) states) and next ten $2p_{10,1}$ excited states with the power increase. The input parameters in the modelling are the gas pressure $p$, the plasma column radius $R$, the gas temperature $T_g$, the electron temperature $T_e$ and the electron density $n_e$. The plasma parameters $T_g$, $T_e$ and $n_e$, experimentally obtained in [12] at applied power $P = (90 - 160)$ W ($T_g$ obtained from Doppler broadening measurements of the spectral line profile and $T_e$, $n_e$ determined through the cross points method of line-intensity ratios), are shown in figures 5 and 6. The calculations are made with an assumption of a Maxwellian EEDF according to the measurements in [10]. Extrapolation of the plasma parameters at $P = 200$ W is carried out due to the smooth dependence of

![Graphs showing gas temperature and electron density](image)

**Figure 5.** Gas temperature dependence on the applied power at $p = 8$, 26 and 260 mTorr of the first chamber.

**Figure 6.** Electron density dependence on the applied power at $p = 8$, 26 and 260 mTorr of the first chamber.
the line intensities on the applied power. The calculations of the process contributions in the applied power range \( P = (90 – 200) \) W show that the metastable pooling and destruction occur mainly through radiative decay from the 2\( p \) excited states and electron impact excitation of metastable states to these levels, respectively. These processes have the main contribution over the whole pressure range, compared to electron impact processes within the block of 1\( s \) levels, electron impact ground state excitation to 1\( s \), diffusion to the walls and ionization from the metastables.

The relative contributions of the electron impact ground state excitation to the 2\( p \) excited states and of the excitation from the metastables are presented in figure 7.

Figure 7. Relative contribution of direct, at \( P = 160 \) W and \( p = (8 – 260) \) mTorr, (in (a)) and step, at \( p = 26 \) mTorr and \( P = (90 – 200) \) W, (in (b)) excitations to the 2\( p \) levels.

Figure 8. Spectrum simulated through the CR model at plasma parameters, \( p = 26 \) mTorr, \( T_g = 590 \) K, \( n_e = 3 \times 10^{11} \) cm\(^{-3}\) and \( T_e = 3.4 \) eV, obtained through extrapolation of plasma parameter dependencies for \( P = 200 \) W, and experimentally registered spectrum at \( P = 200 \) W.
At gas pressure $p = 8$ mTorr, the direct excitation is comparable with the step excitation. At higher pressures the step excitation dominates. The radiative decay of the $2p$ levels is the most efficient depopulation process, compared to ionization by electron collisions from the $2p$ excited states and superelastic electron collisions to the ground and metastable states.

The simulation of argon spectrum at $p = 26$ mTorr and at $P = 200$ W with plasma parameters $T_g = 590$ K, $n_e = 3 \times 10^{11} \text{ cm}^{-3}$ and $T_e = 3.4$ eV, is in good agreement with the experimentally obtained at the same applied power. The spectra are presented in figure 8. The coincidence of the two spectra shows that the applied CR model well describes the population and depopulation mechanisms of excited states at low gas pressure and from low to high applied power.

The spectra observations also show that the most intense spectral lines – with wavelengths 811.5 nm and 763.5 nm – are emitted from levels populated mainly due to stepwise excitation whereas the emission – with wavelengths 750.4 nm and 751.5 nm – from levels populated through direct excitation is quite lower. Dominant contribution of the step processes is a feature of inductive mode operation [1] of the discharge.

4. Conclusions
Optical emission spectroscopy method is applied to study mode operation regime of cylindrical rf inductively-driven discharge in an argon gas at low gas pressures. The emission spectra and the evolution of the spectral line intensities with the applied power increase are analyzed. Mode (E-to-H and H-to-E) transition in the discharge regime operation is not observed in the line intensity behaviour. Calculations based on the kinetic modelling of the main processes contributing to the line intensity behaviour reveal that the density of the metastable atoms control the density population of the ten $2p_{10,1}$ excited states by electronic quenching. The agreement between the experimental line intensity and those predicted by the collisional radiative model at $p = 26$ mTorr indicates that under the conditions of our experiments the discharge regime operation is maintained primary by the induced electrical field. The plasma parameters referred to the coincidence between the spectra experimentally and theoretically obtained are in compliance with the main feature of the H-mode operation of the inductive discharges at low pressure.

5. Acknowledgements
The authors acknowledge Prof. Dr. A. Shivarova for the useful discussions and stimulation in the preparation of the paper. This work is within the EURATOM project FU06-CT-2003-00139 and project no 1315 of the National Science Fund in Bulgaria.

References
[1] Czerwiec T and Graves D B 2004 J. Phys. D: Appl. Phys. 37 2827
[2] Kortshagen U, Gibson N D and Lawler J E 1996 J. Phys. D: Appl. Phys. 29 1224
[3] Cunge G, Crowley B, Vender D and Turner M M 1999 Plasma Sources Sci. Technol. 8 576
[4] El-Fayoumi I M, Jones I R and Turner M M 1998 J. Phys. D: Appl. Phys. 31 3082
[5] Suzuki K, Nakamura K, Ohkubo H and Sugai H 1998 Plasma Sources Sci. Technol. 7 13
[6] Miyoshi Y, Petrović Z Lj and Makabe T 2002 IEEE Trans. Plasma Science 30 (1) 130
[7] Turner M M and Lieberman M A 1999 Plasma Sources Sci. Technol. 8 313
[8] Yoon N S, Kim B C, Yang J G and Hwang S M 1998 IEEE Trans. Plasma Science 26 190
[9] Demidov V I, DeJoseph Jr C A and Kudryavtsev A A 2004 Plasma Sources Sci. Technol. 13 600
[10] Tsankov T, Kiss’ovski Z, Djermanova N and Kolev S 2006 Plasma Process. Polym. 3 151
[11] Dimitrova M, Djermanova N, Kiss’ovski Z, Kolev S, Shivarova A and Tsankov T 2006 Plasma Process. Polym. 3 156
[12] Iordanova S and Koleva I 2006 Spectrochim. Acta Part B submitted