On low pressure electronegative capacitive discharges

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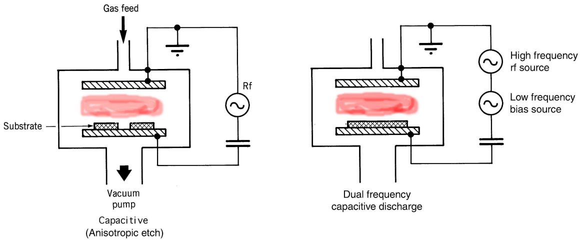
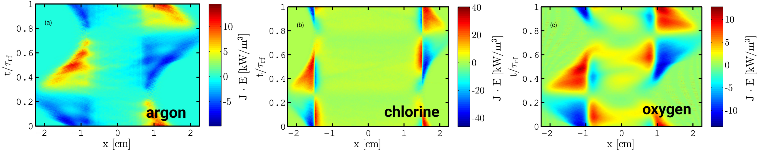
Introduction



Lieberman and Lichtenberg (2025), Principles of Plasma Discharges and Materials Processing, 3rd edition, Wiley

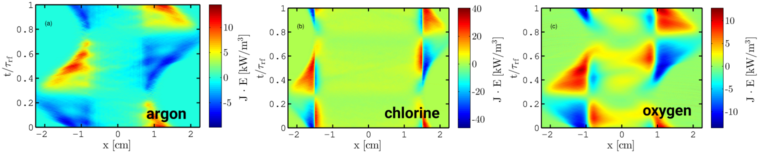
- The capacitive discharge is sustained by applying radio-frequency (rf) voltage or current between electrodes when a neutral gas at low pressure is injected
- The discharge is sustained by radio-frequency (rf) currents and voltages, introduced through capacitive sheaths

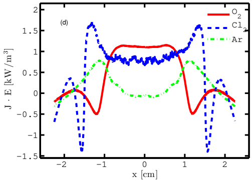
Introduction



- The two main electron power absorption mechanisms in these discharges are
    - momentum transfer due to the moving sheaths which leads to stochastic (pressure or collisionless) heating
    - currents in the main body of the plasma discharge lead to Ohmic (or collisional) heating in the bulk and sheath regions
- We explore this using 1D particle-in-cell/Monte Carlo collision simulation for three different chemistries
    - weakly electronegative oxygen discharge
    - highly electronegative chlorine discharge
    - electropositive argon discharge

Introduction





- The electron power absorption
  - $p = 1.33$ Pa
  - $f = 13.56$ MHz
  - $V_0 = 222$ V
  - gap = 45 mm
  - $\gamma_{\rm see} = 0.0$
- For argon the discharge is in α-mode
- Chlorine and oxygen discharges operate in hybrid α- and drift ambipolar (DA)-mode

Outline

- The 1D particle-in-cell/Monte Carlo collision simulation
- The low pressure oxygen discharge
- The low pressure chlorine discharge
- The argon discharge
- Argon discharge diluted by chlorine
- Summary

The 1D particle-in-cell/Monte Carlo collision simulation

The oopd1 1d-3v PIC/MCC code

- We use the `oopd1` (objective oriented plasma device for one dimension) code to simulate the discharges
- The `oopd1` code was originally developed by the Plasma Theory and Simulation Group at UC Berkeley later Michigan State University
- It has 1 dimension in space and 3 velocity components for particles (1d-3v)
- The `oopd1` code is supposed to replace the widely used `xpdx1` series (`xpdp1`, `xpdc1` and `xpds1`)
- It is developed to simulate various types of plasmas, including processing discharges, accelerators and beams
  - Modular structure
  - Includes relativistic kinematics
  - Particles can have different weights

Gudmundsson et al. (2013) *Plasma Sources Sci. Technol.*, **22**(3) 035011

Wen et al. (2021) *Plasma Sources Sci. Technol.*, **30**(10) 105009

The low pressure oxygen discharge

Introduction

- Oxygen forms a weakly electronegative discharge
- The oxygen discharge is of vital importance in various materials processing applications such as
  - ashing of photoresist
  - etching of polymer films
  - oxidation and deposition of thin film oxides
- The presence of negative ions has a strong influence on the kinetics and dynamics of the oxygen discharge
- The oxygen chemistry is rather involved, in particular due to the presence of **metastable molecular and atomic** oxygen and their role in dissociative attachment and detachment processes

The oxygen discharge

- We consider a discharge that consists of:
  - electrons
  - the ground state oxygen molecule $O_2(X^3\Sigma_g^-)$
  - the metastable oxygen molecule $O_2(a^1\Delta_g)$
  - the metastable oxygen molecule $O_2(b^1\Sigma_g)$
  - the ground state oxygen atom $O(^3P)$
  - the metastable oxygen atom $O(^1D)$
  - the negative oxygen ion O^-
  - the positive oxygen ions O^+ and O_2^+

Gudmundsson and Lieberman (2015) *Plasma Sources Sci. Technol.*, **24** 035016

Hannesdottir and Gudmundsson (2016) *Plasma Sources Sci. Technol.*, **25** 055002

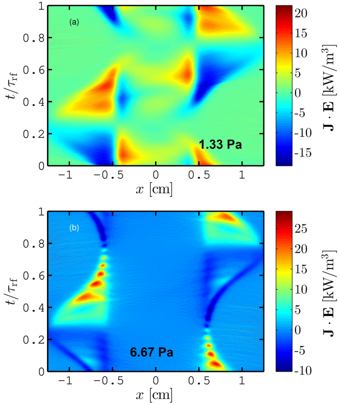
Gudmundsson and Proto (2019) *Plasma Sources Sci. Technol.*, **28** 045012

- We apply a global model beforehand to calculate the partial pressure of the various neutrals

Thorsteinsson and Gudmundsson, *Plasma Sources Sci. Technol.*, **19** 055008 (2010)

Oxygen CCP – pressure dependence

- A parallel plate capacitively coupled oxygen discharge at driving frequency of 13.56 MHz for gap separation of 45 mm
- The spatio-temporal electron heating $\mathbf{J}_{\mathrm{e}} \cdot \mathbf{E}$ at 1.33 Pa (upper graph) and 6.67 Pa (lower graph)
- At 1.33 Pa there is a significant electron power absorption within the electronegative core
- At 6.67 Pa the electron power absorption occurs almost solely in the sheath region

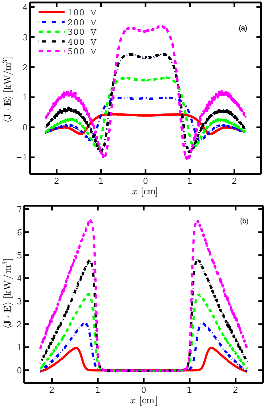


Gudmundsson and Snorrason (2017) JAP **122** 193302

Hannesdottir and Gudmundsson (2016) PSST, **25** 055002

Oxygen CCP – pressure dependence

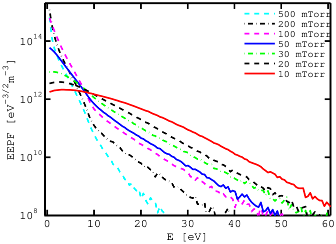
- The time averaged electron power absorption $\langle \mathbf{J}_e \cdot \mathbf{E} \rangle$ at 1.33 Pa (upper graph) and 6.67 Pa (lower graph)
- At 1.33 Pa there is significant electron power absorption within the electronegative core
- At 6.67 Pa, the electron power absorption the electronegative core is roughly zero, and the electron power absorption is almost entirely located in the sheath regions



Gudmundsson and Snorrason (2017) JAP **122** 193302

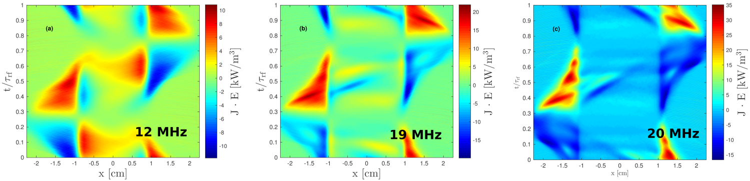
Oxygen CCP – pressure dependence

- At low pressure ($<$ 4 Pa) the EEPF is Druyvesteyn like and becomes bi-Maxwellian as the pressure is increased
- These results contradict what is commonly found for the capacitively coupled argon discharge where the EEPF evolves from being bi-Maxwellian at low pressure to being Druyvesteyn like at high pressure



Gudmundsson and Ventéjou (2015) JAP **118** 153302

Oxygen CCP – frequency dependence



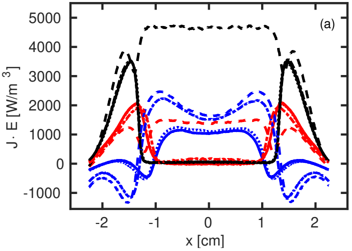
Gudmundsson et al. (2018), PSST **27**(2) 025009

- Oxygen discharge at 1.33 Pa
- At 12 MHz significant power absoption is observed in the plasma bulk but also in the sheath region
- At 19 MHz the heating and cooling in the sheath regions has increased, however there is contribution to the electron power absorption in the bulk region (note the change in scale)
- At 20 MHz there is almost no electron power absorption in the plasma bulk

Oxygen CCP – pressure dependence

- At **1.33 Pa** excluding the metastable states in the simulation has very small influence on the heating mechanism
- At **6.67 Pa** the metastable states have a significant influence on the heating mechanism and even more significant at **26.67 Pa**
- This is due to detachment by the metastable $O_2(a^1\Delta_g)$ molecule

$$O^- + O_2(a^1\Delta_g) \rightarrow \text{products}$$



Gudmundsson and Ventéjou (2015) JAP **118** 153302

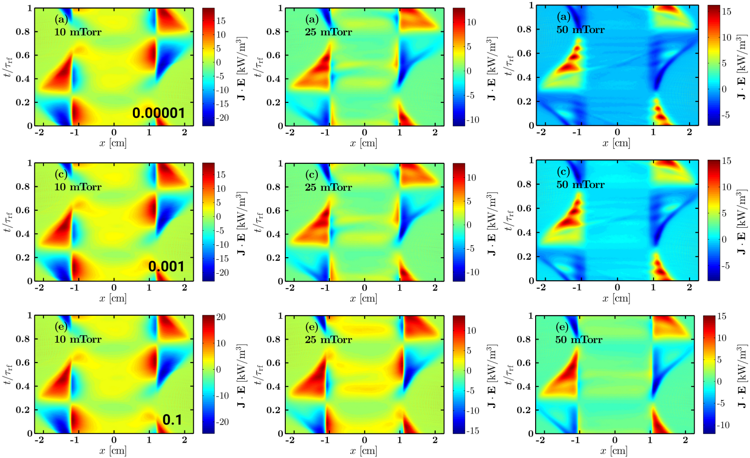
Gudmundsson and Lieberman (2015) PSST **24** 035016

Gudmundsson and Hannesdottir, AIP

Conf. Proc. (2017) **1811** 120001

dashed line – no detachment
solid line – detachment by $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma_g^+)$

Oxygen CCP – surface quenching of $O_2(a^1\Delta_g)$



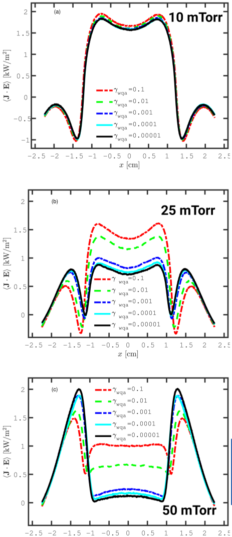
Proto and Gudmundsson (2018) PSST **27** 074002 (2018)

Oxygen CCP – surface quenching of $O_2(a^1\Delta_g)$

- At 1.33 Pa (10 mTorr) almost all the electron heating occurs in the plasma bulk (the electronegative core) and the electron heating profile is independent of the surface quenching coefficient
- At 6.67 Pa (50 mTorr) only for the highest surface quenching coefficients 0.1 and 0.01 there is some electron heating observed in the bulk region
- Typical value is 0.007 for iron

Sharpless and Slanger (1989) *Journal of Chemical Physics* **91** 7947

Proto and Gudmundsson (2018) PSST **27** 074002 (2018)



Electron power absorption – Bolzmann term analysis

- To determine the electron power absorption mechanisms we apply Boltzman term analysis

Surendra and Dalvie (1993) PRE **48**(5) 3914 and Schulze et al. (2018) PSST **27**(5) 055010

- The electric field within the electronegative core constitutes contributions through various different phenomena
- The electric field is composed of six terms

$$E = \underbrace{-\underbrace{\frac{m_e}{e}\frac{\partial u_e}{\partial t}}_{\mathrm{I}} + \underbrace{\frac{m_e}{e}\frac{u_e^2}{n_e}\frac{\partial n_e}{\partial x}}_{\mathrm{II}} + \underbrace{\frac{m_e}{e}\frac{u_e}{n_e}\frac{\partial n_e}{\partial t}}_{\mathrm{III}}}_{\text{inertia}} \underbrace{- \underbrace{\frac{T_e}{n_e}\frac{\partial n_e}{\partial x}}_{\mathrm{IV}} - \underbrace{\frac{\partial T_e}{\partial x}}_{\mathrm{V}}}_{\text{pressure}} \underbrace{- \underbrace{\frac{m_e u_e \nu_c}{e}}_{\mathrm{VI}}}_{\text{Ohmic}}$$

Electron power absorption – Bolzmann term analysis

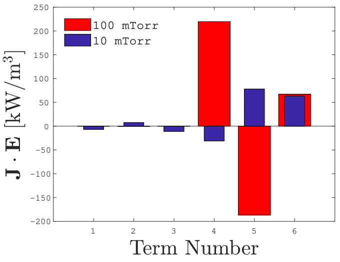
- The electron absorbed power can be determined as follows

$$J_\mathrm{e} \cdot E = \underbrace{m_\mathrm{e} u_\mathrm{e} n_\mathrm{e} \frac{\partial u_\mathrm{e}}{\partial t}}_{\mathrm{I}} - \underbrace{m_\mathrm{e} u_\mathrm{e}^3 \frac{\partial n_\mathrm{e}}{\partial x}}_{\mathrm{II}} - \underbrace{m_\mathrm{e} u_\mathrm{e}^2 \frac{\partial n_\mathrm{e}}{\partial t}}_{\mathrm{III}} + \underbrace{e u_\mathrm{e} T_\mathrm{e} \frac{\partial n_\mathrm{e}}{\partial x}}_{\mathrm{IV}} + \underbrace{e n_\mathrm{e} u_\mathrm{e} \frac{\partial T_\mathrm{e}}{\partial x}}_{\mathrm{V}} + \underbrace{m_\mathrm{e} n_\mathrm{e} \nu_\mathrm{c} u_\mathrm{e}^2}_{\mathrm{VI}} \quad (1)$$

- Terms I and III – power absorption due to electron inertia
- Term II – the electron density gradient
- Term IV – ambipolar field – electron density gradient
- Term V – the electron temperature gradient
  - Terms IV and V are pressure or collisionless heating
- Term VI – electron neutral collisions or Ohmic heating

Oxygen CCP – Bolzmann term analysis

- The pressure terms are important – significant part of the electron power absorption at both 1.33 Pa and 13.3 Pa is due to the pressure terms
- The Terms IV (ambipolar) and V (electron temperature gradient) flip signs and are sharply smaller in the absolute value at 1.33 Pa
- The Ohmic term's magnitude (Term VI) is similar at both pressures
- Note that Ohmic power absorption is important even at low pressure



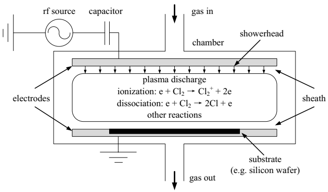
Proto and Gudmundsson (2020) JAP **128** 113302

The low pressure chlorine discharge

The chlorine discharge

- Chlorine is an electronegative diatomic gas that is widely used in plasma etching of both semiconductors and metals, in particular poly-silicon gate and aluminum interconnects
- Chlorine atoms are believed to be the primary reactant in plasma etching
- The chlorine molecule has
  - a low dissociation energy (2.5 eV)
  - a near-zero threshold energy for dissociative attachment
- All electronic excitations of the molecule appear to be dissociative, and no metastable molecular states are of importance

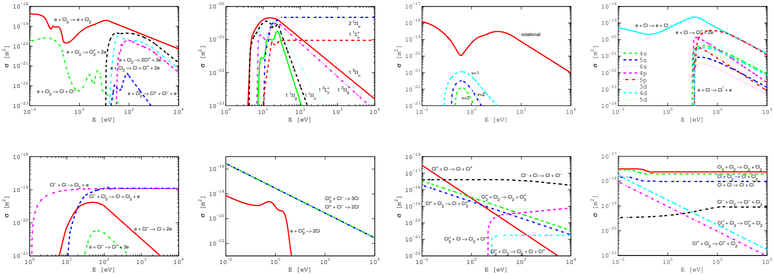
The chlorine discharge



- We consider a discharge that consists of:
  - electrons
  - the ground state chlorine molecule $Cl_2(X\,^1\Sigma_g^+, v = 0)$,
  - the ground state chlorine atom Cl($3p^5\,{}^2P$)
  - the negative chlorine ion Cl^-
  - the positive chlorine ions Cl^+ and Cl_2^+
- We apply a global model beforehand to calculate the fraction of Cl atoms

Thorsteinsson and Gudmundsson (2010) *Plasma Sources Sci. Technol.*, **19**(1) 015001

The chlorine discharge



- The reaction set for the chlorine is comprehensive and includes 44 reactions

Huang and Gudmundsson (2013) *Plasma Sources Sci. Technol.*, **22**(5) 055020

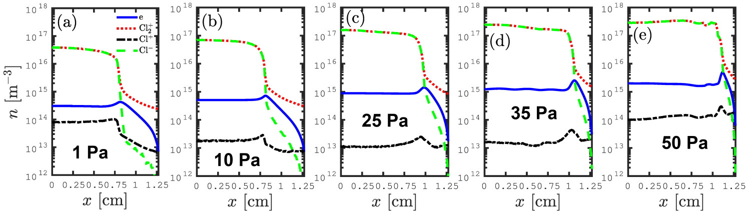
Electron power absorption

Voltage source operated at a single frequency

$$V(t) = V_{\mathrm{rf}} \sin(2\pi f t)$$

gap = 2.54 cm, V_{rf} = 222 V and f = 13.56 MHz

Electron power absorption



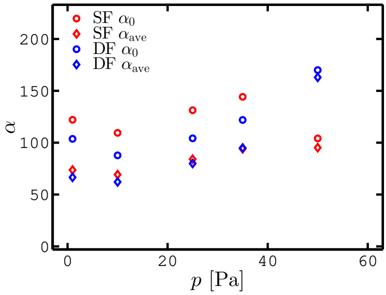
Proto and Gudmundsson (2021) PSST **30**(6) 065009

- The time averaged charged particle density profiles of a parallel plate capacitively coupled chlorine discharge
- At low pressures, the profile for Cl_2^+ ions is cosine-like or parabolic since Cl_2^+ ions are lost mainly due to diffusion to the walls
- As the pressure increases, the recombination between Cl_2^+ and Cl^- ions becomes the major loss mechanism for Cl_2^+ ions and the density profile for Cl_2^+ and Cl^--ions becomes flat in the bulk region

Electron power absorption

- The electronegativity in the discharge center is very high and increases with increased operating pressure
- Negative ion creation proceeds via electron impact dissociative attachment whose cross-sections are high
- Drift-ambipolar (DA) power absorption is expected within the electronegative core

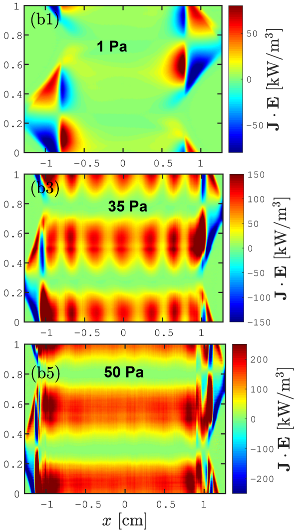
Schulze et al. (2011) PRL **107**(27) 275001



Skarphedinsson and Gudmundsson (2020) PSST **29**(8) 084004

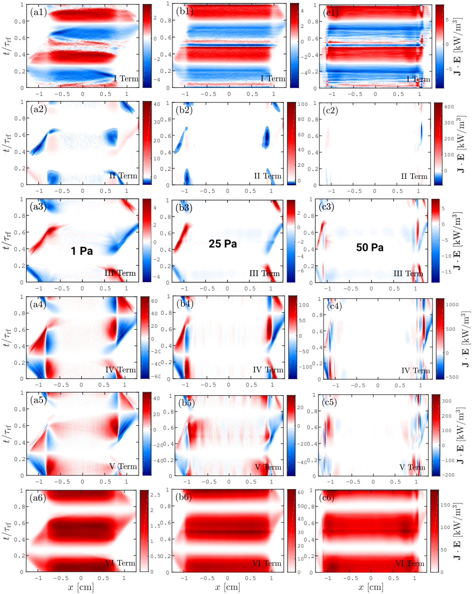
Electron power absorption

- The spatio temporal behavior of the total electron power absorption $\mathbf{J}_e \cdot \mathbf{E}$ over the full gap length for a capacitively coupled chlorine discharge
- At 1 Pa there is clear sign of drift ambipolar heating (DA-mode) and stochastic heating (α-mode)
- At 35 and 50 Pa there are indications of striations in addition to drift ambipolar heating (DA-mode) and stochastic heating (α-mode)



Proto and Gudmundsson (2021) PSST **30**(6) 065009

Electron power absorption



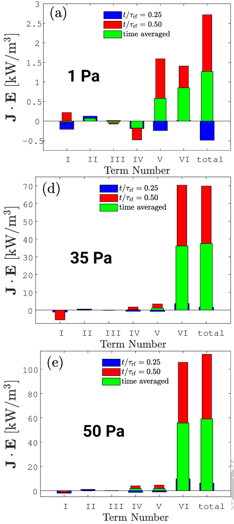
- The spatio temporal behavior of the various terms that constitute the electron power absorption
    - Terms I and III – electron inertia
    - Term I – electron density gradient
    - Terms IV and V – pressure (collisionless) heating
    - Term VI – Ohmic heating

Proto and Gudmundsson (2021) PSST **30**(6) 065009

Electron power absorption

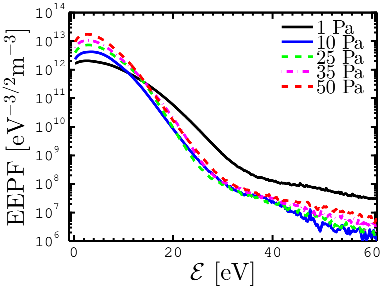
- The space averaged electron power absorption profile terms
  - $t/\tau_{\rm rf} = 0.25$ blue bar
  - $t/\tau_{\rm rf} = 0.5$ red bar
  - time averaged green bar
- At 1 Pa the pressure terms and the Ohmic term contribute to the electron power absorption
- At higher pressures Ohmic power absorption dominates

Proto and Gudmundsson (2021) PSST **30**(6) 065009



Electron power absorption

- The electron energy probability function (EEPF) in the discharge center is Druyvesteyn like at all pressures
- This is expected when there is significant Ohmic heating in the plasma bulk



Striations

Voltage source operated at a single frequency

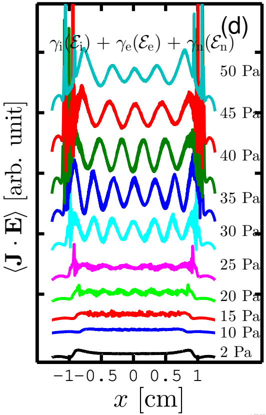
$$V(t) = V_{\mathrm{rf}} \sin(2\pi f t)$$

gap = 2.54 cm, V_{rf} = 222 V and f = 13.56 MHz

$$\gamma_{\mathrm{i}}(\mathcal{E}_{\mathrm{i}}) + \gamma_{\mathrm{n}}(\mathcal{E}_{\mathrm{n}}) + \gamma_{\mathrm{e}}(\mathcal{E}_{\mathrm{e}}, \theta)$$

Striations

- Primary electron power absorption as the pressure is varied
- We observe striation pattern at 15 Pa
- Above 15 Pa the striation amplitude increases with increased pressure up to 35 Pa, and then decreases
- The number of striations decreases with increased pressure



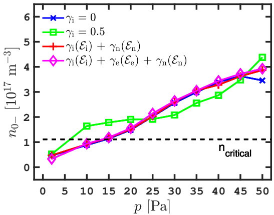
Mahdavipour and Gudmundsson (2025) PSST **34**(4) 045005

Striations

- Striations are known to appear in electronegative discharges when two conditions are simultaneously fulfilled:
  - high enough electronegativity
  - driving frequency that is comparable to the ion plasma frequency
- Based on the ion-ion plasma model striations may appear when $n_{\rm ion} > n_{\rm critical}$ where the critical density is defined as

$$n_{\rm critical} = \frac{\omega_{\rm rf}^2 \epsilon_0 \mu}{e^2}$$

where $\mu = m_+ m_- / (m_+ + m_-)$ is the reduced mass of the positive and negative ions



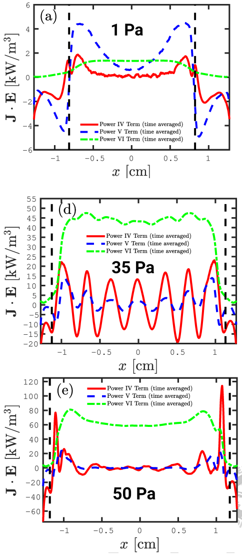
Mahdavipour and Gudmundsson (2025)

PSST **34**(4) 045005

Liu et al. (2017) PSST **26**(5) 055024

Striations - electron power absorption

- The time averaged electron power absorption profile of
  - term IV (red line)
  - term V (blue dashed line)
  - term VI (green dot dashed line)
- At 1 Pa the pressure terms and the Ohmic term contribute to the electron power absorption
- At higher pressures Ohmic power absorption dominates
- At 35 Pa striations are observed – the pressure terms are apparent



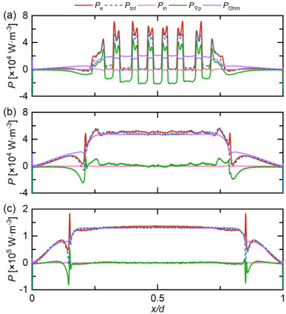
Proto and Gudmundsson (2021) PSST **30**(6) 065009

Striations – electron power absorption

- Similar findings have been reported more recently for a capacitive CF_4 discharge operated at 60 Pa and 6.78 MHz with 2 cm electrode gap (top graph)
- In this case the pressure terms

$$P_{\nabla p} = \underbrace{e u_{\rm e} T_{\rm e} \frac{\partial n_{\rm e}}{\partial x}}_{\rm IV} + \underbrace{e n_{\rm e} u_{\rm e} \frac{\partial T_{\rm e}}{\partial x}}_{\rm V}$$

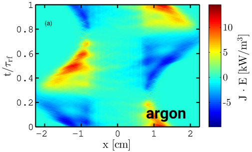
  dominate the power absorption process
- For higher operating frequency the Ohmic term dominates the electron power absorption (lower graphs)

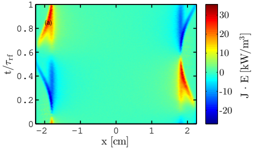


Zhou et al. (2025) PSST **34**(8) 085014

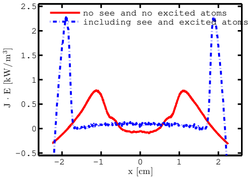
The argon discharge

Introduction



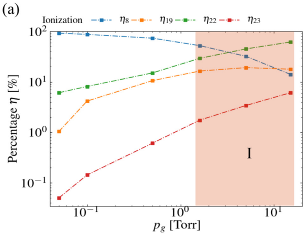


- The electron power absorption
  - $p = 1.33$ Pa
  - $f = 13.56$ MHz
  - $V_0 = 222$ V
  - gap = 45 mm
  - no excited argon states and $\gamma_{\rm see} = 0.0$ (left)
  - with excited argon states and $\gamma_{\rm i}(\mathcal{E}_{\rm i}) + \gamma_{\rm n}(\mathcal{E}_{\rm n}) + \gamma_{\rm e}(\mathcal{E}_{\rm e}, \theta)$ (right)
- For argon the discharge is in α-mode



The argon discharge

- The ionization processes versus pressure neglecting secondary electrons
  - Electron impact ionization
  - Step-wise ionization
  - Metastables pooling ionization
  - Metastables pooling ionization
- The region labeled I ($p_g > 200$ Pa) indicates where the contributions of metastable pooling and step-wise ionization exceed that of electron impact ground state ionization



Wen et al. (2021) *Plasma Sources Sci. Technol.*, **30**(10) 105009

Argon discharge diluted by chlorine

Current source operated at a single frequency

$$J(t) = J_{\mathrm{rf}} \sin(2\pi f t)$$

gap = 2.54 cm, J_{rf} = 50 A/m^2 and f = 13.56 MHz, p = 6.67 Pa

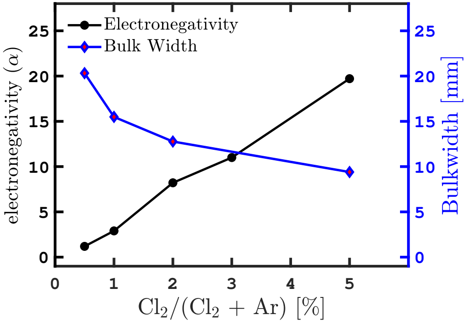
Argon discharge diluted by chlorine

Case	exited species	ions	neutrals	electrons
I	0.0	0.0	0.0	0.0
II	included	0.0	0.0	0.0
III	included	$\gamma_{\mathrm{see,i}}(\mathcal{E}_{\mathrm{i}})$	$\gamma_{\mathrm{see,n}}(\mathcal{E}_{\mathrm{n}})$	$\gamma_{\mathrm{see,e}}(\mathcal{E}_{\mathrm{e}}, \theta)$

- We explored three cases
  - Neglecting excited states of argon and all secondary electron emission
  - Including excited states of argon and neglecting all secondary electron emission
  - Including excited states of argon and all secondary electron emission

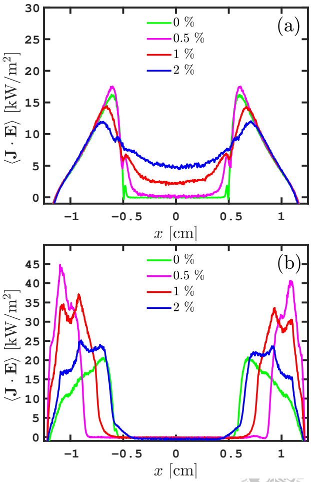
Argon discharge diluted by chlorine

- Ar/Cl_2 mixture at 6.67 Pa
- The electronegativity increases with increased chlorine fraction
- The plasma bulk width decreases with increased chlorine fraction
- Gap width 2.5 cm, driving current density 50 A/m^2 and frequency 13.56 MHz
- The argon model includes excited argon atoms



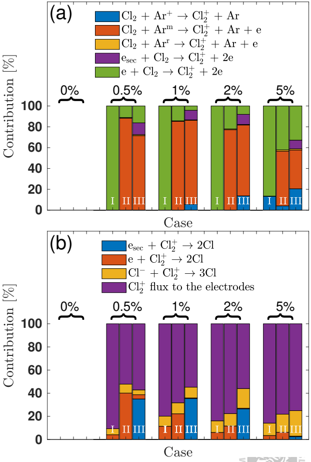
Argon discharge diluted by chlorine

- Ar/Cl_2 mixture at 6.67 Pa
- The time-averaged electron power absorption for varying chlorine dilution
  - (a) neglecting excited argon states
  - (b) including excited argon states



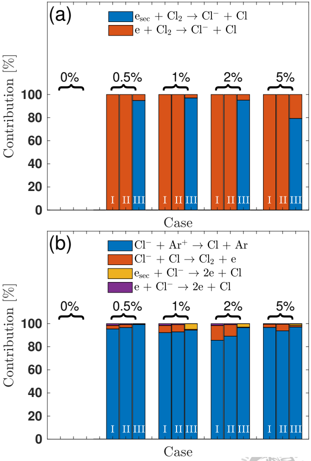
Argon discharge diluted by chlorine

- Ar/Cl_2 mixture at 6.67 Pa
- The creation and loss of the Cl_2^+ ion
- The role of Penning ionization becomes rather significant as excited states of argon are included in the discharge model
- Gap width 2.5 cm, driving current density 50 A/m^2 and frequency 13.56 MHz



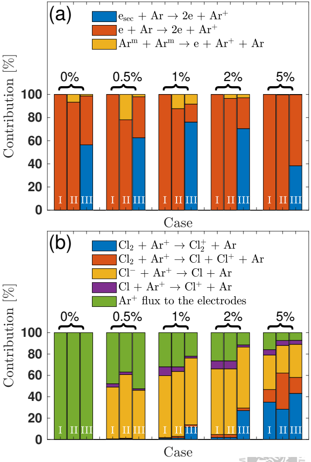
Argon discharge diluted by chlorine

- Ar/Cl_2 mixture at 6.67 Pa
- The creation and loss of the negative Cl^- ion
- The creation of the negative ion Cl^- is almost entirely due to dissociative attachment as for pure chlorine discharge
- The role of secondary electrons becomes significant as they are included in the discharge model
- Ion-ion recombination with Ar^+ is the main loss process



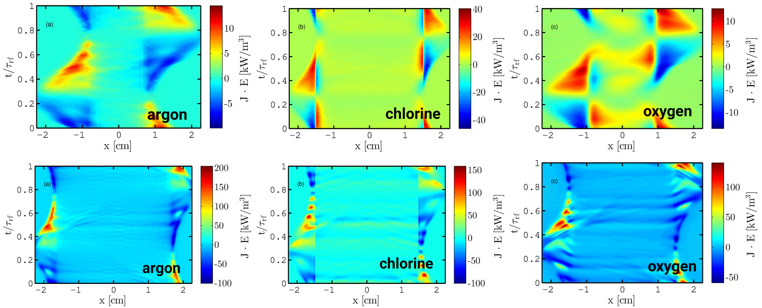
Argon discharge diluted by chlorine

- Ar/Cl_2 mixture at 6.67 Pa
- The creation and loss of the Ar^+ ion
- The contribution of secondary electrons becomes significant as they are included in the discharge model



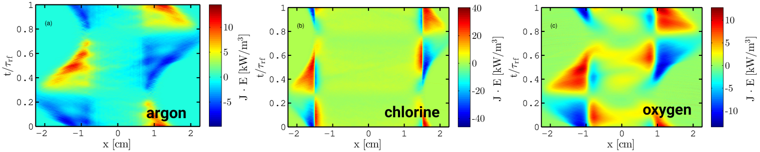
Summary

Summary



- Capcacitively coupled argon, oxygen and chlorine discharges were studied by particle-in-cell/Monte Carlo collision simulations

Summary



- In a chlorine discharge DA electron power absorption dominates and becomes increasingly more Ohmic with increased pressure
- Including the detachment processes by the singlet metastable states has a strong influence on the electronegativity in the oxygen discharge
- A heating mode transition, from hybrid drift-ambipolar (DA) and α-mode to pure α-mode, is observed as the pressure, driving frequency, or electrode gap spacing are increased, or the quenching coefficient of the oxygen metastable $O_2(a^1\Delta_g)$ is varied

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Thank you for your attention

The slides can be downloaded at

`http://langmuir.raunvis.hi.is/~tumi/ranns.html`

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