On electron power absorption in low pressure electronegative capacitive discharges

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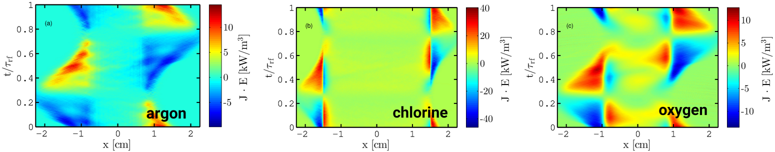
78th Gaseous Electronics Conference
Seoul, Republic of Korea
October 16., 2025

Introduction



p = 1.33 Pa, f = 13.56 MHz, V_0 = 222 V, gap = 45 mm and $\gamma_{\rm see}$ = 0.0

- 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 – detachment
  - highly electronegative chlorine discharge – recombination
  - electropositive argon discharge

Outline

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

The low pressure oxygen discharge

Introduction

- 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
- 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

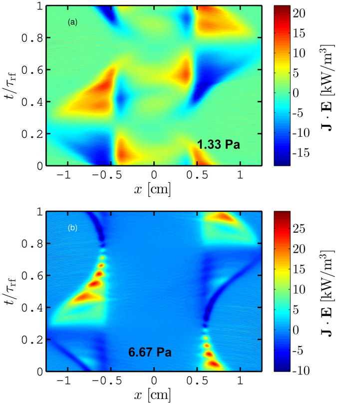
Hannesdottir and Gudmundsson (2016) *Plasma Sources Sci. Technol.*, **25** 055002

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

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 $\mathbf{J}_{\rm e} \cdot \mathbf{E}$ 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

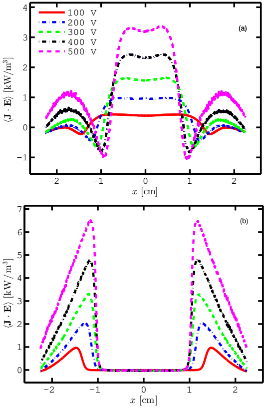
Hannesdottir and Gudmundsson (2016) PSST, **25** 055002



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

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



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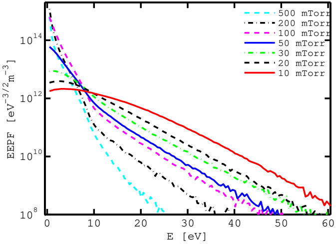
Oxygen CCP – pressure dependence

- 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

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 electron absorbed power can be determined as follows

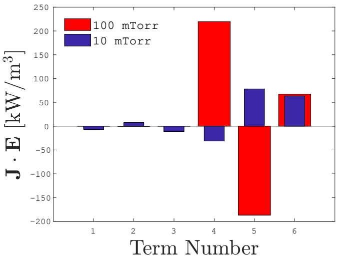
$$J_\mathrm{e} \cdot E = \underbrace{\underbrace{m_\mathrm{e} u_\mathrm{e} n_\mathrm{e} \frac{\partial u_\mathrm{e}}{\partial t}}_{\mathrm{I}} - \underbrace{m_\mathrm{e} u_\mathrm{e}^3 \frac{\partial n_\mathrm{e}}{\partial x}}_{\mathrm{II}} - \underbrace{m_\mathrm{e} u_\mathrm{e}^2 \frac{\partial n_\mathrm{e}}{\partial t}}_{\mathrm{III}}}_{\text{inertia}}$$

$$+ \underbrace{\underbrace{e u_\mathrm{e} T_\mathrm{e} \frac{\partial n_\mathrm{e}}{\partial x}}_{\mathrm{IV}} + \underbrace{e n_\mathrm{e} u_\mathrm{e} \frac{\partial T_\mathrm{e}}{\partial x}}_{\mathrm{V}}}_{\text{pressure}} + \underbrace{\underbrace{m_\mathrm{e} n_\mathrm{e} \nu_\mathrm{c} u_\mathrm{e}^2}_{\mathrm{VI}}}_{\text{Ohmic}}$$

- 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

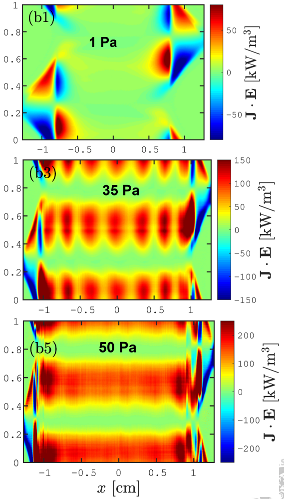
The low pressure chlorine discharge

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
- 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^+

Electron power absorption

- The spatio temporal behavior of the total electron power absorption $\mathbf{J}_{\mathrm{e}} \cdot \mathbf{E}$ 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 DA- and α-mode)
- DA power absorption is expected within the electronegative core



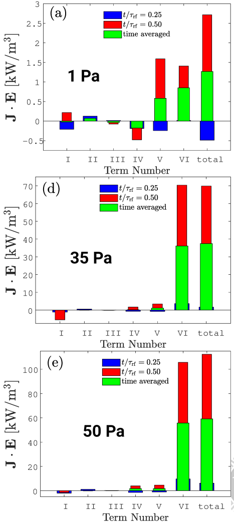
Schulze et al. (2011) PRL **107**(27) 275001

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

Electron power absorption

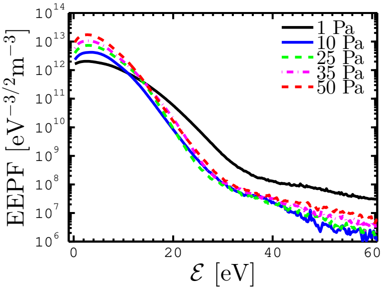
- The space averaged electron power absorption profile terms
  - $t/\tau_{\text{rf}} = 0.25$ blue bar
  - $t/\tau_{\text{rf}} = 0.5$ red bar
  - time averaged green bar
- At 1 Pa the pressure terms and the Ohmic term contribute to the electron power absorption
- At higher pressures Ohmic power absorption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

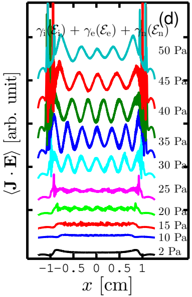


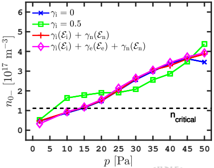
Striations

- 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





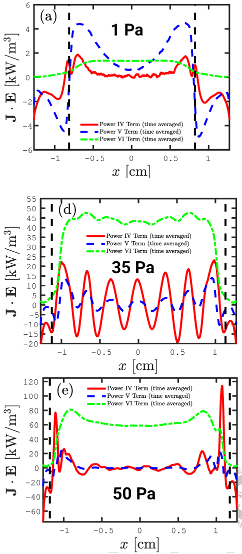
Mahdavipour and Gudmundsson (2025) PSST **34**(4) 045005

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

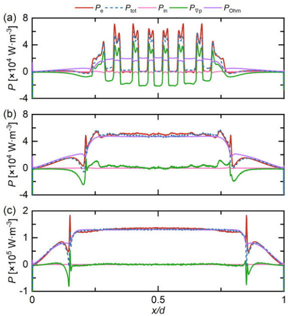


Striations – electron power absorption

- 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

$$P_{\nabla p} = \underbrace{e u_e T_e \frac{\partial n_e}{\partial x}}_{\text{IV}} + \underbrace{e n_e u_e \frac{\partial T_e}{\partial x}}_{\text{V}}$$

  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

Argon discharge diluted by chlorine

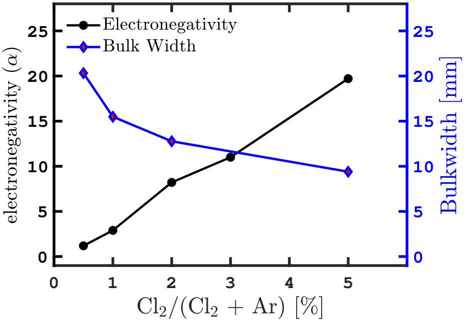
Current source operated at a single frequency

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

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

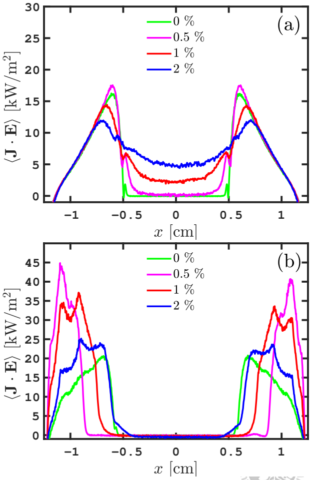
Argon discharge diluted by chlorine

- 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



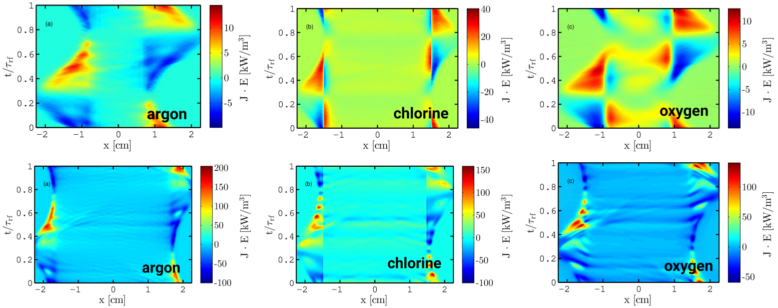
Argon discharge diluted by chlorine

- 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



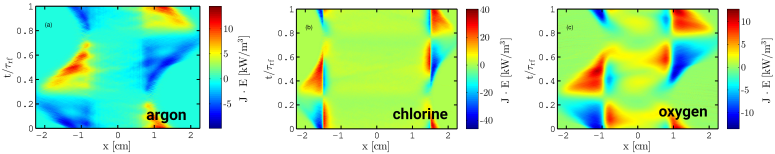
Summary

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

Thank you for your attention

The slides can be downloaded at

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

Various parts of this work were made by a number of students

- Shuo Huang (UM-SJTU, later University of Michigan and now KLA-Tencor)
- Bruno Ventéjou (LPGP, Université Paris-Sud, Orsay, France)
- Hólmfríður Hannedóttir (University of Iceland later Harvard University and now Institute for Advanced Study)
- Davíð I. Snorrason (University of Iceland now Grein Research)
- Andrea Proto (University of Iceland)
- Garðar A. Skarphéðinsson (University of Iceland)
- Janez Krek (Michigan State and now KLA-Tencor)
- Bahram Mahdavipour (University of Iceland)

in collaboration with

- prof. Michael A. Lieberman (UC Berkeley)
- prof. John P. Verboncoeur (Michigan State)
- Dr. Emi Kawamura (UC Berkeley)
- Dr. De-Qi Wen (Michigan State)

The work was funded by Icelandic Research Fund grants nos. 163086 and 217999 the University of Iceland Research Fund.

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