Electron heating in electronegative capacitively coupled discharge of complex chemistry

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Introduction

- Oxygen forms a weakly electronegative 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
- We use the `oopd1` (objective oriented plasma device for one dimension) **particle-in-cell Monte Carlo collision** code to simulate the discharge
- It has 1 dimension in space and 3 velocity components for particles (1d-3v)
- 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., *Plasma Sources Sci. Technol.*, **22** 035011 (2013)

Outline

- The oxygen discharge
- Capacitively Coupled Oxygen Discharge at 13.56 MHz
  - including both $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma_g)$
  - including secondary electron emission
- Pressure dependence
- Frequency dependence
- Dependence on surface quenching of $O_2(a^1\Delta_g)$
- The effect of $\gamma_{\rm see}(E)$
- Summary

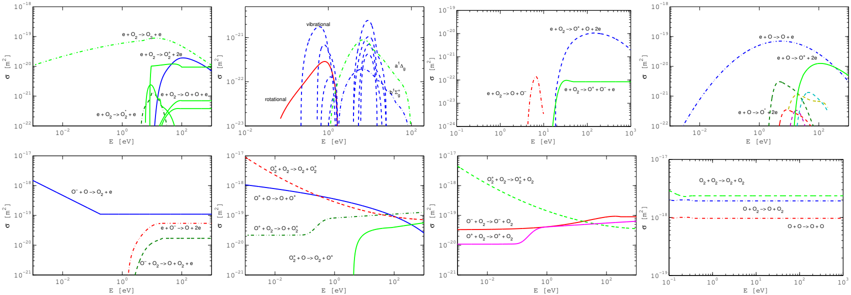
The oxygen discharge

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^+
- The discharge model includes energy dependent secondary electron emission yield
- We assume a parallel plate capacitively coupled oxygen discharge at with electrode separation of 4.5 cm
- We apply a global model[1] beforehand to calculate the partial pressure of the various neutrals

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

The oxygen discharge



- The reaction set for the oxygen is comprehensive and for this study includes up to 67 reactions

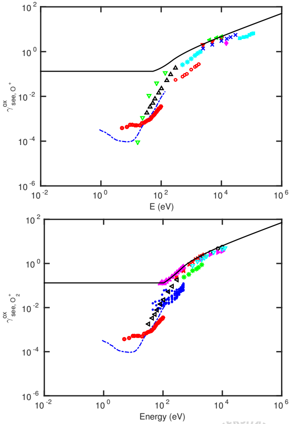
Gudmundsson et al., *Plasma Sources Sci. Technol.*, **22** 035011 (2013)

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

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

The oxygen discharge

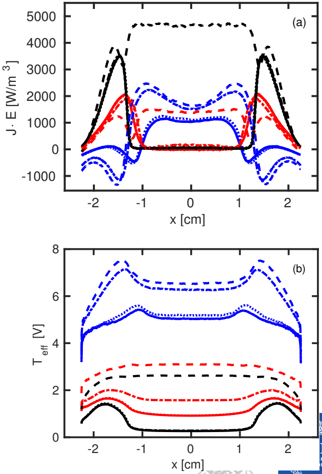
- The discharge model also includes energy dependent secondary electron emission yield
- We have compiled experimental data from the literature on secondary electron emission yields for the species O_2^+, O^+, O_2 and O bombarding various metals and substances
- A fit was made through the available experimental data



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

Oxygen CCP – pressure dependence

- The number of cold electrons increases and negative ion density decreases as the metastables $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma_g)$ are added to the discharge model
- The electron heating in the bulk drops to zero at the higher pressures
- The effective electron temperature profile changes significantly when detachment by singlet metastables is added to the reaction set
- **10 mTorr**, **50 mTorr** and **200 mTorr**



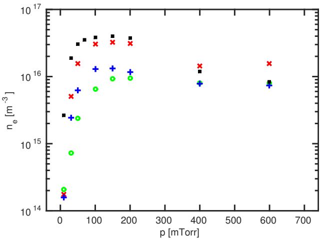
Gudmundsson and Hannesdottir, AIP Conf. Proc. **1811** 120001 (2017)

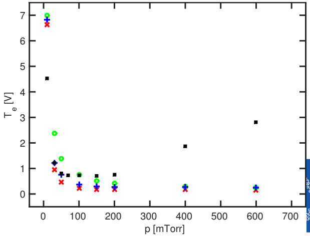
The oxygen discharge

- Comparison to experimental findings:
  - o $\gamma_{\rm see} = 0.0$,
    4.4 % $O_2(a^1\Delta_g)$
  - + $\gamma_{\rm see} = 0.0$,
    4.4 % $O_2(a^1\Delta_g)$ and 4.4 % $O_2(b^1\Sigma_g)$
  - x $\gamma_{\rm see} = \gamma_{\rm see}(E)$,
    4.4 % $O_2(a^1\Delta_g)$ and 4.4 % $O_2(b^1\Sigma_g)$
  - ▪ Experimental findings by Kechkar

    (S. Kechkar, Ph.D. Thesis, Dublin City University, January 2015)

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





Capacitively Coupled Oxygen Discharge single frequency at 13.56 MHz

– pressure dependence –

including $O_2(a^1\Delta_g)$, $O_2(b^1\Sigma_g)$ and $\gamma_{\rm see}(E)$

Oxygen CCP – pressure dependence

- We apply a voltage source with a single frequency

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

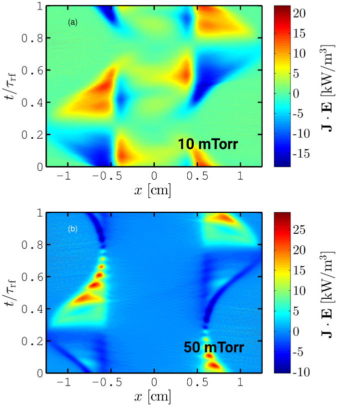
- The electrodes are circular with a diameter of 14.36 cm
- The gap between the electrodes is 4.5 cm
- We set $V_{\rm rf} = 222$ V and $f = 13.56$ MHz
- The neutrals (O_2 and O and the metastables) are treated as background gas at $T_{\rm g} = 300$ K with a Maxwellian distribution
- If the kinetic energy of the neutrals reaches a certain threshold they are tracked
- The dissociation fraction and the metastable fraction is found using a global model

Oxygen CCP – pressure dependence

- The spatio-temporal electron heating $\mathbf{J}_e \cdot \mathbf{E}$ at 10 and 50 mTorr
- At 10 mTorr there is a significant electron heating within the electronegative core
- At 50 mTorr the electron heating occurs almost solely in the sheath region

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

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

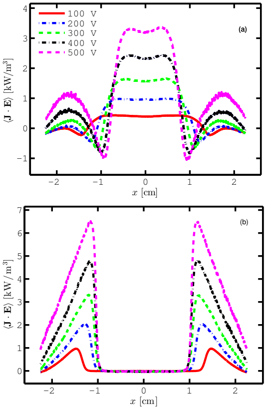


Gudmundsson and Snorrason (2017) JAP **122** 193302

Oxygen CCP – pressure dependence

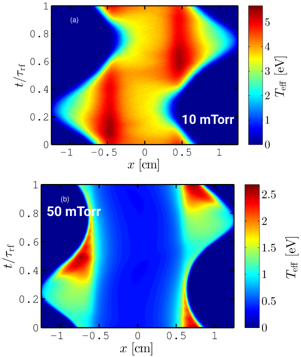
- The time averaged electron heating $\langle \mathbf{J}_\mathrm{e} \cdot \mathbf{E} \rangle$ at 10 and 50 mTorr
- At 10 mTorr there is significant electron heating within the electronegative core
- At 50 mTorr, the heating rate in the electronegative core is roughly zero, and electron heating is almost entirely located in the sheath regions

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



Oxygen CCP – pressure dependence

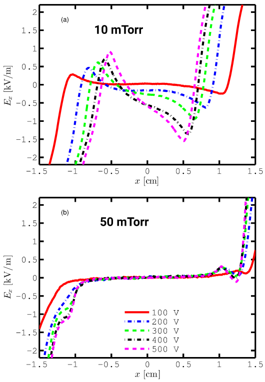
- At 10 mTorr the effective electron temperature is high within the plasma bulk (the electronegative core) throughout the rf period and peaks within the plasma bulk during the sheath collapse phase
- At 50 mTorr a peak in the effective electron temperature within the plasma bulk in the sheath expansion phase and is low within the plasma bulk throughout the rf period



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

Oxygen CCP – pressure dependence

- The axial electric field at $t/\tau_{\rm rf} = 0.5$ for both 10 and 50 mTorr
- At 10 mTorr there is a significant electric field strength within the electronegative core
- This strong electric field within the plasma bulk (the electronegative core) indicates a drift-ambipolar (DA) heating mode
- This electric field is a combination of a drift field and an ambipolar field
- At 50 mTorr the electric field is zero within the electronegative core



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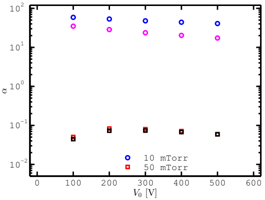
Oxygen CCP – pressure dependence

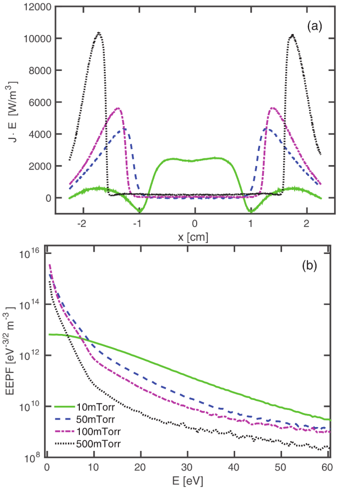
- The electronegativity is significantly higher when operating at 10 mTorr than when operating at 50 mTorr
- At 10 mTorr, the discharge is operated in a combined drift-ambipolar (DA) and α-mode
- At 50 mTorr, the discharge is in a pure α-mode and sheath heating dominates
- The transition from the combined DA-α-mode to the pure α-mode coincides with a significant decrease in the electronegativity



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

Oxygen CCP – pressure dependence

- At low pressure the EEPF is convex, the population of low energy electrons is relatively low
- As the pressure is increased the number of low energy electrons increases and the number of higher energy electrons (> 10 eV) decreases
- Thus the EEPF develops a concave shape or becomes bi-Maxwellian as the pressure is increased



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

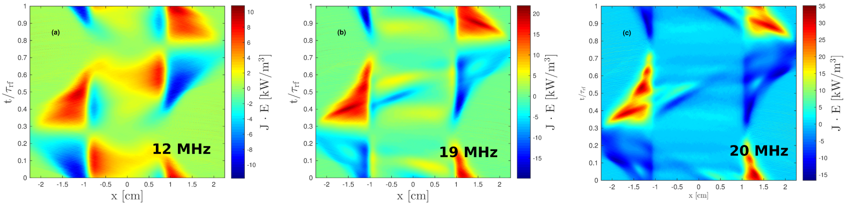
Gudmundsson and Ventéjou (2015) JAP **118** 153302

Capacitively Coupled Oxygen Discharge single frequency at 10 mTorr

– driving frequency dependence –

including $O_2(a^1\Delta_g)$, $O_2(b^1\Sigma_g)$ and $\gamma_{\rm see}(E)$

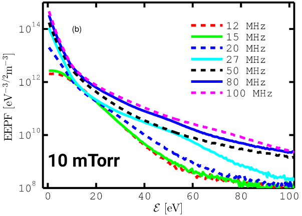
Oxygen CCP – frequency dependence

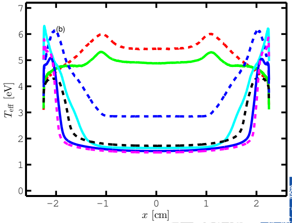


- At 12 MHz significant heating is observed in the plasma bulk but also in the sheath region
- At 19 MHz the heating and cooling in the sheath regions has increased, however there is contribution to the electron heating in the bulk region (note the change in scale)
- At 20 MHz there is almost no electron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

Oxygen CCP – frequency dependence

- At low driving frequency the EEPF is convex, the population of low energy electrons is relatively low
- The EEPF remains convex for driving frequency up to 15 MHz and has transitioned to concave or bi-Maxwellian shape at 20 MHz
- Increasing the driving frequency enhances the high energy tail as the number of high energy electrons increases

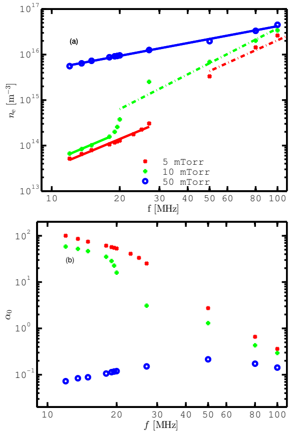




Gudmundsson et al. (2018) PSST **27** 025009

Oxygen CCP – frequency dependence

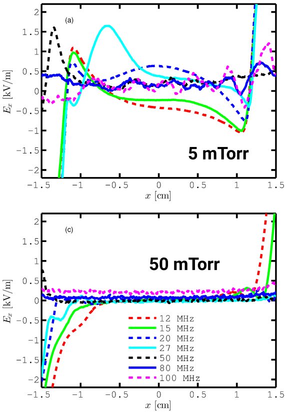
- At 10 mTorr there is a jump in the center electron density between 20 and 27 MHz
- At 10 mTorr $n_{\mathrm{e}} \propto f^{2.11}$ at low frequency, below 18 MHz, and $n_{\mathrm{e}} \propto f^{2.00}$ at higher frequencies, 27.12 MHz and above
- At 50 mTorr $n_{\mathrm{e}} \propto f^{1.16}$ over the entire frequency range explored and no transition is observed
- We see that at 5 and 10 mTorr the electronegativity decreases with increasing driving frequency



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

Oxygen CCP – frequency dependence

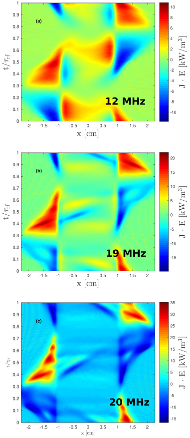
- The electric field profile at $t/\tau_{\mathrm{rf}} = 0.5$ for discharges operated at 5 and 50 mTorr
- We see a significant electric field strength within the electronegative core at low driving frequency and low pressure
- The strong electric field within the plasma bulk (the electronegative core), at low pressure and low driving frequency, indicates a drift-ambipolar (DA) heating mode



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

Oxygen CCP – frequency dependence

- At a low driving frequency and low pressure (5 and 10 mTorr), a combination of stochastic (α-mode) and drift ambipolar (DA) heating in the bulk plasma (the electronegative core) is observed
- The DA-mode dominates the time averaged electron heating
- As the driving frequency is increased, the heating mode transitions into a pure α-mode
- At low pressure (5 and 10 mTorr), this transition coincides with a sharp decrease in electronegativity



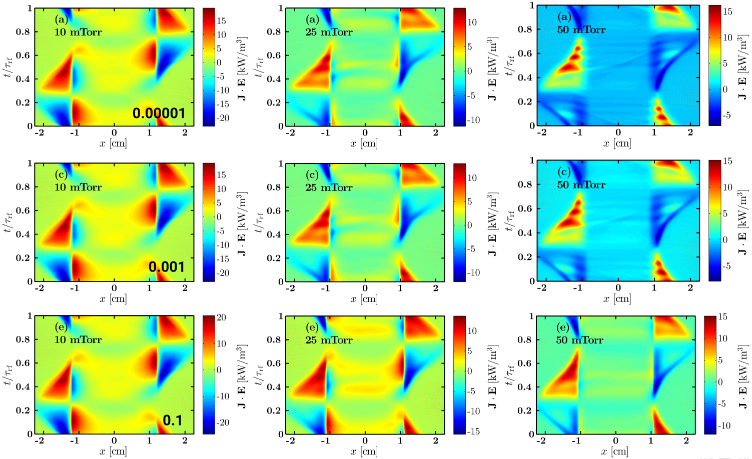
Gudmundsson et al. (2018) PSST **27** 025009

Capacitively Coupled Oxygen Discharge

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

including $O_2(a^1\Delta_g)$, $O_2(b^1\Sigma_g)$ and $\gamma_{\rm see}(E)$

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

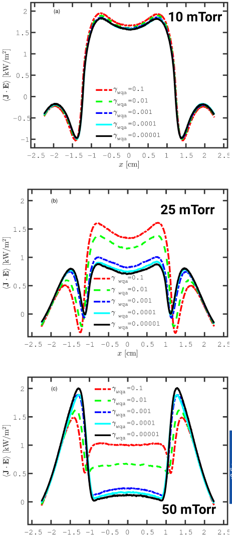


Proto and Gudmundsson (2018) PSST **27** 074002 (2018)

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

- At 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 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)

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



Capacitively Coupled Oxygen Discharge

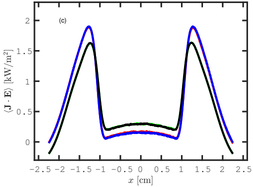
– the effect of $\gamma_{\rm see}(E)$ –

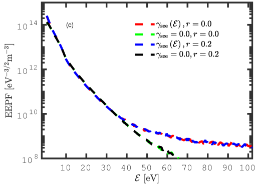
including $O_2(a^1\Delta_g)$, $O_2(b^1\Sigma_g)$ and $\gamma_{\rm see}(E)$

Oxygen CCP – the effect of $\gamma_{\text{see}}(E)$

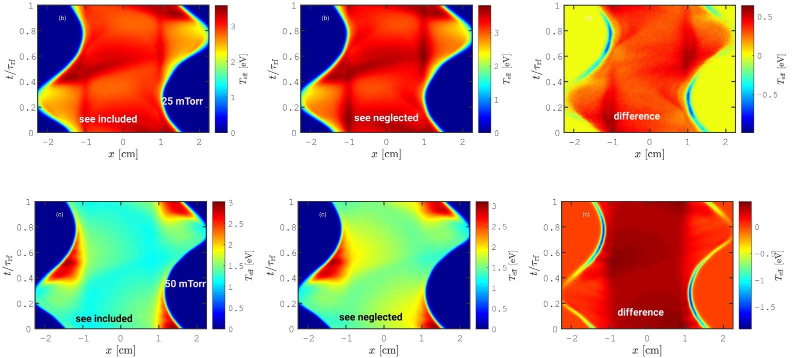
- Adding secondary electron emission yield
  - increases the electron density
  - increases the electron heating rate in the sheath region
  - the sheath region becomes narrower
  - a high energy tail appears in the EEPF

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



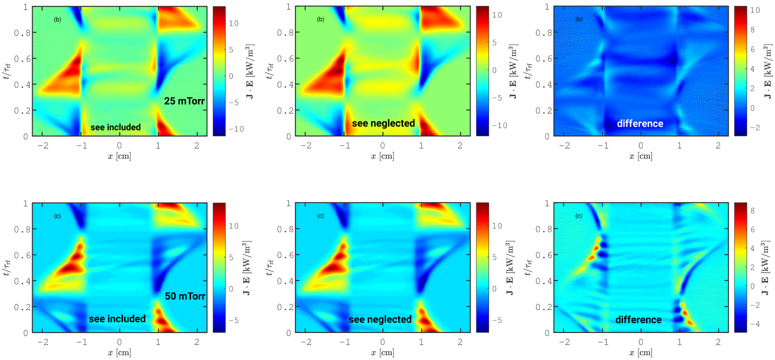


Oxygen CCP – the effect of $\gamma_{\rm see}(E)$



- Including secondary electron emission increases the electron energy

Oxygen CCP – the effect of $\gamma_{\mathrm{see}}(E)$



- Including secondary electron emission decreases the electron power absorption

Summary

Summary

- We demonstrated particle-in-cell/Monte Carlo collision simulation of a capcacitively coupled disharge
- Including the detachment processes by the singlet metastable states has a strong influence on the effective electron temperature and electronegativity in the oxygen discharge
- At low pressure the discharge is operated in a combined drift-ambipolar (DA) and α-mode, and at higher pressure it is operated in the pure α-mode

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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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