Analysis of electron density in oxygen rf plasma by 160 GHz microwave interferometry: instabilities and laser photodetachment of negative ions

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2 Experimental setup
3 Electron densities
4 Instabilities
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1. **Motivation**

2. Experimental setup

3. Electron densities

4. Instabilities

5. O⁻ densities

6. Summary
Motivation

- negative ions
  - providing information about elementary processes
  - strong influence on reaction kinetics


- standard technique for negative ion diagnostics
  - combination of laser photodetachment and probe diagnostics

- disadvantages of probes
  - strong invasive
  - need various model assumption
  - interaction laser pulse with probe material
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- goal of project B5
  - investigation of oxygen rf plasmas
  - measurement of O\textsuperscript{-} density
  - combination of laser photodetachment and microwave interferometry

- advantages microwave interferometry
  ⊕ minimal-invasive
  ⊕ applicable in reactive as well as electronegative plasmas
  ⊕ no model assumption
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- current status
  ✓ establishment of 160GHz microwave interferometry
  ✓ measurement of electron density
  ✓ measurement of instabilities
  ✓ measurement O\(^{-}\) density
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Instabilities

O\textsuperscript{-} densities

Summary
Microwave interferometry

$$\Delta \Phi = \frac{\pi}{n_c \lambda_{MWI}} \int_{z_1}^{z_2} n_e(z) \, dz$$

- $$\omega_{MWI} \gg \omega_{Pe}$$
- quasi optical description
- optimization between aperture and spatial resolution

- $$f_{MWI} = 160.28 \text{ GHz}$$
- $$\lambda_{MWI} = 1.87 \text{ mm}$$
- $$n_c \approx 3.2 \cdot 10^{22} \text{ m}^{-3}$$
Microwave interferometry

\[ \Delta \Phi = \frac{\pi}{n_c \lambda_{MWI}} \int_{z_1}^{z_2} n^e(z) \, dz \]

- \( \omega_{MWI} \gg \omega_{Pe} \)
- quasi optical description
- optimization between aperture and spatial resolution

- \( n^e \sim 1 \cdot 10^{16} \text{ m}^{-3} \)
- \( n^o \sim 0.5 \ldots 0.7 \cdot n^e \)

\( f_{MWI} = 160.28 \text{ GHz} \)
\( \lambda_{MWI} = 1.87 \text{ mm} \)
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\( \Delta \Phi = 0.03^\circ \)
Microwave interferometry

**Motivation**

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- Quasi optical description
- Optimization between aperture and spatial resolution

**Experimental setup**

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**Electron densities**

- \( n^e \sim 1 \times 10^{16} \text{ m}^{-3} \)
- \( n^e \sim 0.5 \ldots 0.7 \cdot n^e \)

**Instabilities**

- \( \Delta \Phi \approx 0.03^\circ \)

**O^− densities**

- \( n^e \sim 0.5 \ldots 0.7 \cdot n^e \)
Gaussian beam propagation theory

- Beam radius $w$
  
  $$w = w_0 \cdot \sqrt{1 + \left( \frac{z \lambda_{MWI}}{\pi w_0^2} \right)^2}$$

- $1/e$ decay of the electric field

- Radius of curvature $R$
  
  $$R = z + \frac{1}{z} \left( \frac{\pi w_0^2}{\lambda_{MWI}} \right)^2$$
Optical components

- mirrors
- horn antenna

- $340 \times 240 \times 70 \text{mm}^3$
- self-developed calculation and manufacturing

- numerical calculation and manufacturing by IfP University of Stuttgart
Gaussian beam propagation measurement

- measured microwave distribution

\[
\text{fit} = \frac{a}{\sigma_{\text{Gauss}} \cdot (2\cdot\pi)^{1/2}} \cdot \exp \left( -\frac{(x-x_0)^2}{2\cdot\sigma_{\text{Gauss}}^2} \right)
\]

\[
a = 7.62 \text{ V/mm}
\]

\[
x_0 = -7.17 \times 10^{-5} \text{ mm}
\]

\[
\sigma_{\text{Gauss}} = 11.95 \text{ mm}
\]

\[
\sigma_{\text{rmse}} = 0.03
\]

- evaluated distributions for several positions

\[
\hat{w} = w \sqrt{2} \quad (\text{theo.})
\]

\[
\tilde{w} \quad (\text{exp.})
\]
Experimental setup

- asymmetric capacitively coupled rf discharge (cc-rf)
  - gases: Ar, O\textsubscript{2} @ 5 sccm
  - pressure: 10...100 Pa
  - frequency: 13.56 MHz
  - electrode: 10 cm in diameter
  - power: 10...100 W
  - self-bias: −80...−600 V

- microwave interferometer (PLL, heterodyne)
  - \( f_{\text{MWI}} \) = 160.28 GHz
  - \( \lambda_{\text{MWI}} \) = 1.87 mm
  - \( \Delta t \) = 0.2 \( \mu \text{s} \)
  - \( \Delta y \) = 10 mm
  - \( \Delta \Phi \) = 0.016°
  - \( \Delta n_{L}^e \) = 5.3 \( \times 10^{13} \) m\textsuperscript{-2}
Microwave propagation through the device

- gaussian beam propagation
- optical axis 20 mm above electrode
- minimal beam radius 5 mm
- axial spatial resolution of 10 mm

J MEICHSNER et al. Surface and Coating Tech. 98 (1998) 1565-1571
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Literature comparison

**Argon**

- $13 \, \text{Pa}$
  - $n_e \left[ \text{m}^{-3} \right]$ vs. $V_{PP} \left[ \text{V} \right]$
  - Overzet et al., exp.
  - $x \times 10^{16}$

- $33 \, \text{Pa}$
  - $n_e \left[ \text{m}^{-3} \right]$ vs. $V_{PP} \left[ \text{V} \right]$
  - Overzet et al., exp.
  - $x \times 10^{16}$

**Oxygen**

- $13.8 \, \text{Pa}$
  - $n_e \left[ \text{m}^{-3} \right]$ vs. $V_{RF/Ampl} \left[ \text{V} \right]$
  - Katsch et al., exp.
  - $x \times 10^{16}$

- $10 \, \text{W}$
  - $n_e \left[ \text{m}^{-3} \right]$ vs. pressure [Pa]
  - Stoffels (30 sccm), exp. (5 scem)
  - $x \times 10^{15}$

[References]

**Time dependence (10 Hz pulsed)**

- **Argon**
  - Decay time: 300…1000 µs
  - Influence of rf generator
  - Rise time in ms range

- **Oxygen**
  - Decay time: 500…5000 µs
  - Electron peak in the afterglow
    - 10…100 Pa
    - |self-bias| < 250 V

- Detachment by $O (^3P)$, $O_2 (X^3Σ_g^-)$ and $O_2 (a^1Δ_g)$

  \[
  O^- + O_2 (X^3Σ_g) \rightarrow O_2 (X^3Σ_g^-) + O (^3P) + e \\
  O^- + O_2 (a^1Δ_g) \rightarrow \text{products} + e
  \]
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Attachment induced ionization instability

- attachment $\leftrightarrow$ detachment

- properties and parameter
  - start-up time
    $1 \ldots 5 \text{ ms}$
  - frequency
    $100 \text{ Hz} \ldots 3 \text{ kHz}$
  - peak to peak
    $0.2 \ldots 3.5 \cdot 10^{15} \text{ m}^{-2}$


Appearance of instabilities

- Stability map

\[ Q = \frac{\partial}{\partial T_e} \left( k_{a}^{O_2} \right) - \frac{\partial}{\partial T_e} \left( k_{iz}^{O_2} \right) \]

- Mechanism

\[ Q > 1 \iff \text{instability} \]
\[ Q < 1 \iff \text{no instab.} \]

- Investigation by Nighan et al.

- \[ e + O_2 \rightarrow O + O^- \]
- \[ e + O_2 \rightarrow O_2^+ + 2e \]

> 60 Pa dominated by instabilities

< 60 Pa stability islands
Appearance of instabilities

- stability map

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<thead>
<tr>
<th>Pressure [Pa]</th>
<th>Power [W]</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>40</td>
<td>40</td>
</tr>
<tr>
<td>60</td>
<td>60</td>
</tr>
<tr>
<td>80</td>
<td>80</td>
</tr>
<tr>
<td>100</td>
<td>100</td>
</tr>
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> 60 Pa dominated by instabilities
< 60 Pa stability islands

- mechanism

- investigation by Nighan et al.

\[
Q = \frac{\partial}{\partial T_e} \left( k^{O_2}_{a} \right) = \begin{cases} 
> 1 \iff \text{instability} \\
< 1 \iff \text{no instab.}
\end{cases}
\]

\[
e + O_2 \xrightarrow{k^{O_2}_{a}} O + O^- \\
e + O_2 \xrightarrow{k^{O_2}_{iz}} O_2^+ + 2e
\]

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③ Electron densities
④ Instabilities
⑤ O\textsuperscript{-} densities
⑥ Summary
Experimental setup

- Nd:YAG laser
  - wavelength: 512 nm
  - photon energy: 2.3 eV
  - puls energy: 400 mJ
  - repetition rate: 10 Hz
  - pulse width: 7 ns
  - diameter: 13 mm
  - overlap angle: 12.6°
  - overlap length: ~15 cm
  - axial distance: 20 mm

Other components:
- Elliptical mirror
- Horn antenna
- Microwave interferometer
- Phase-shifter
- Oscilloscope
- PC
- BNC
- GIB
Experimental setup

Nd:YAG laser

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Detachment signal

\[ \text{O}^- + h\nu \rightarrow \text{O} + e \]

- measured with microwave interferometer
- **first** time in low pressure oxygen cc-rf plasma
- averaged 5000 times
- electron peak $\triangle$ negative atomic oxygen ion density
Test of saturation

- Detachment ratio - laser energy variation

\[
\frac{\Delta n_e}{n_0^{O^-}} = \left[ 1 - \exp \left( -\frac{\sigma}{h \nu A} \right) \right]
\]

- \( \sigma = 6.4 \cdot 10^{-22} \text{ m}^2 \)
- Measurements performed at 350 mJ

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Pressure dependence of detachment signal @40W

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<th>20 Pa</th>
<th>30 Pa</th>
<th>60 Pa</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n_{L}^O$ [$10^{14}$ m$^{-2}$]</td>
<td>2</td>
<td>5</td>
<td>7</td>
</tr>
<tr>
<td>$\tau$ [\mu s]</td>
<td>75</td>
<td>37</td>
<td>3.2</td>
</tr>
<tr>
<td>$n_{L}^e$ [$10^{14}$ m$^{-2}$]</td>
<td>40</td>
<td>5</td>
<td>2</td>
</tr>
<tr>
<td>$\alpha = \frac{n_{L}^O}{n_{L}^e}$ [1]</td>
<td>0.05</td>
<td>1</td>
<td>3.5</td>
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- Significant change in decay time constant and electronegativity
- Transition between two different plasma modes
- In each mode the decay time constant decreases with rising pressure as expected
Power dependence of detachment signal @30 Pa

- all values show step-like behaviour
- the boundary value is 50 W
- this changes are may be combined with the transition from \( \alpha \)-mode to \( \gamma \)-mode
- \( \gamma \)-mode characterized by lower „electron temperature“ at much higher electron density
(a) excitation due to electron heating during the sheath expansion phase (I)
- $\alpha$-mode discharge

(b) additional excitation pattern (II) due to secondary electrons
- $\gamma$-mode of discharge
0D-attachment-detachment model for low decay time constant

(i) Assumptions

- constant density of other species e.g. $O_2^+$, $O_2 (X^3Σ_g^−)$, $O_2 (a^1Δ_g)$ and $O (3P)$
- $n^e + n^− = n^{+0} \equiv \text{const}$
- neglecting detachment by $O (3P)$

(ii) Considered processes

\[
\begin{align*}
  &e + O_2 (X^3Σ_g^−) \xrightarrow{k_a^{O2}} O^- + O (3P) \quad \text{attachment} \\
  &e + O_2 (a^1Δ_g) \xrightarrow{k_a^{Δ}} O^- + O (3P) \\
  &O^- + O_2 (X^3Σ_g^−) \xrightarrow{k_d^{O2}} O_2 (X^3Σ_g^−) + O (3P) + e \quad \text{detachment} \\
  &O^- + O_2 (a^1Δ_g) \xrightarrow{k_d^{Δ}} O_3 (\text{or products}) + e
\end{align*}
\]

(iii) Rate equation

\[
\frac{dn^e}{dt} = -\left( k_a^{O2} (T_e) n^{O2} + k_a^{Δ} (T_e) n^{Δ}\right)n^e + \left( k_d^{O2} n^{O2} + k_d^{Δ} n^{Δ}\right)n^- = -\tilde{K}_a n^e + \tilde{K}_d n^-
\]

0D-attachment-detachment model for low decay time constant

(iv) Solution

\[
\frac{\tilde{K}_a}{\tilde{K}_d} = \frac{n^{-0}}{n^{e0}} = \alpha \\
\Delta n^e(t) = n^{-0} \cdot \exp \left[ -\left( \tilde{K}_a + \tilde{K}_d \right) \cdot \frac{t}{1/\tau} \right]
\]

\[
\tilde{K}_a = \frac{1}{\tau} \cdot \frac{1}{1 + \frac{1}{\alpha}} \\
\tilde{K}_d = \frac{1}{\tau} \cdot \frac{1}{1 + \alpha}
\]

(v) Estimated densities

- neglecting detachment with \( \text{O}_2 \left( X^3\Sigma_g^- \right) \)
  reveals density of \( \text{O}_2 \left( a^1\Delta_g \right) \) (\( \hat{n}_A \)) and \( \text{O}_2 \left( X^3\Sigma_g^- \right) \) (\( \hat{n}_O^2 \))

\[ \hat{n}_A = \frac{\tilde{K}_d}{k_d} \]

\( \hat{n}_O^2 \) remaining part of the density

---

0D-attachment-detachment model for low decay time constant

(vi) Estimated attachment rate coefficient $k_a$

- Literature attachment rate coefficient
  \[ k_{a, O_2} = 8.8 \times 10^{-17} \exp \left( \frac{-4.4}{T_e} \right) \]
  
- $k_{a, A} = 2.28 \times 10^{-16} \exp \left( \frac{-2.29}{T_e} \right)$

\[ \tilde{K}_a = k_{a, O_2}^O (T_e) n^{O_2} + k_{a, A}^A (T_e) n^{A} \]

- Density taken from (v)
- Decreasing „electron temperature“ from 6…2 eV
- High „electron temperature“ hint to $\alpha$-mode
- Important to consider $O_2 (a^1 \Delta_g)$
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Summary

- applied microwave interferometry successfully
  - resolution 10 cm, 0.2 µs,
    \[ \Delta \Phi = 0.016^{\circ}, \Delta n_e^L = 5.3 \cdot 10^{13} \text{ m}^{-2} \]
  - systematic analysis of electron densities \( n_e^L \sim \cdot 10^{15} \ldots 5 \cdot 10^{16} \text{ m}^{-2} \)
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