

Surface effects in a capacitive argon discharge at intermediate pressure

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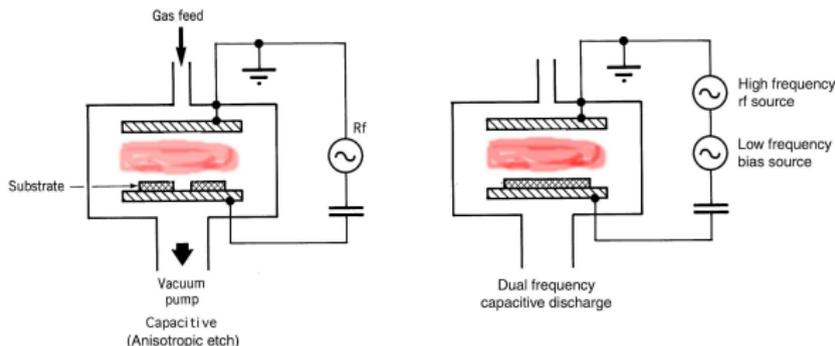
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The XXV Europhysics Conference on the Atomic and
Molecular Physics of Ionized Gases,
Paris, France, July 22., 2022



Introduction



Lieberman and Lichtenberg (2005), Principles of Plasma Discharges and Materials Processing, Wiley

- One of the most widely used types of low-pressure discharges is sustained by radio-frequency (rf) currents and voltages, introduced through capacitive sheaths
- At low pressure nonlocal effects are important and the electron-neutral ionization/excitation frequency is typically fairly uniform across the discharge gap

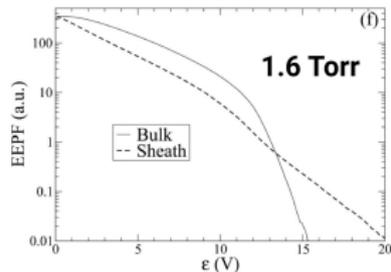
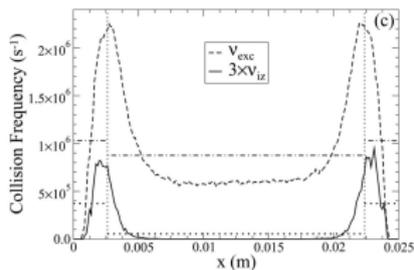


Introduction

- Radio frequency (rf) capacitively coupled plasma (CCP) discharges operated in the intermediate pressure regime (0.2 – 6.0 Torr) are of increasing importance
- In this pressure regime, the mean free path for both ions and electrons is comparable to or smaller than the electrode spacing
- Hence the plasma characteristics are significantly different from that in a low pressure capacitive discharge – the electron-neutral ionization and excitation are localized at the sheath edges
- One-dimensional particle-in-cell/Monte Carlo collisional simulations performed on a capacitive 2.54 cm gap driven by a sinusoidal rf current density amplitude of 50 A/m^2 at 13.56 MHz, with the base case being 1.6 Torr argon discharge



Introduction – argon at 1.6 Torr



Kawamura et al. (2020) JVSTA **38** 023003

- The profile of the ionization and excitation frequencies exhibit a peak near the sheath edges and the ionization is almost nonexistent in the bulk region
- The EEPFs gathered within the central bulk (solid) and sheath (dashed) regions
- The bulk EEPF is Druyvesteyn-like with a strongly depressed tail above the argon excitation energy of 11.55 V – no bulk electrons with high energy
- Excited argon and secondary electrons were neglected

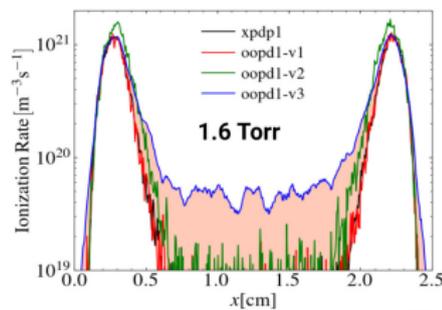
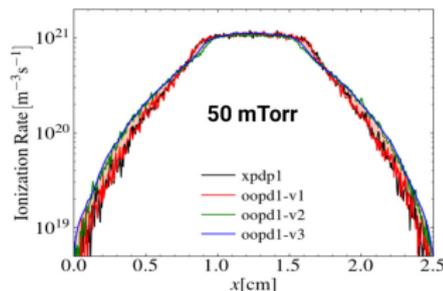


Pressure dependence – no surface effects



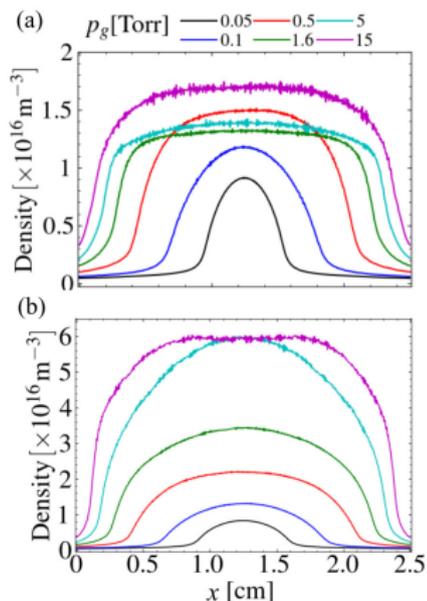
Pressure dependence

- The ionization rate profiles at
 - 50 mTorr (upper)
 - 1.6 Torr (lower)
 - rf current source at 50 A/m² and 13.56 MHz
- The results show varying completeness of the discharge model
- The blue line indicates simulations where the metastable Ar^m, the radiative Ar^r, and the Ar(4p) manifold are included and modeled as time- and space-evolving fluid species
- Without excited species there is no ionization in the bulk



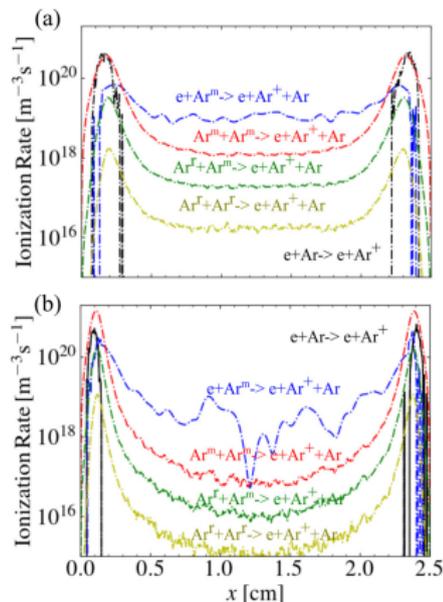
Pressure dependence

- The time averaged ion density profile for various pressures calculated
 - without excited state atoms (upper)
 - including excited state atoms treated as a fluid (lower)
 - rf current source at 50 A/m^2 and 13.56 MHz
- It is found that the presence of the excited species influences the density profile and enhances the plasma density by a factor of 3 at 1.6 Torr

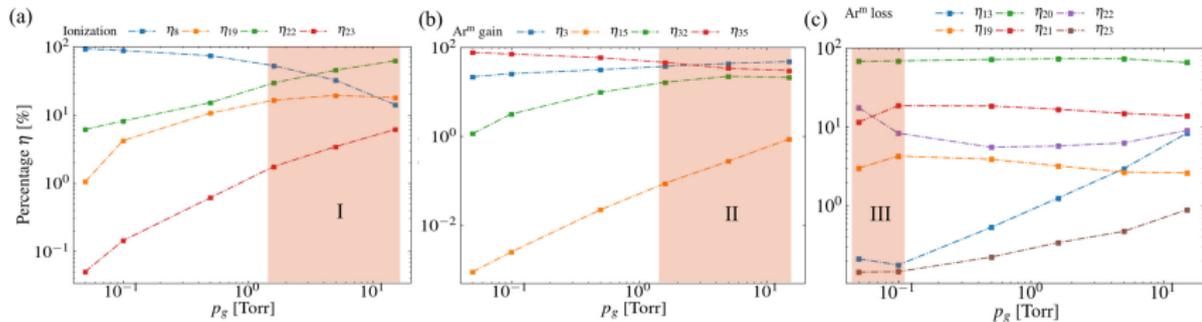


Pressure dependence

- The time-averaged ionization reaction rates for primary electron-neutral impact ionization, metastable pooling, and step-wise ionization at
 - 5 Torr (upper)
 - 15 Torr (lower)
- The main ionization rates are
 - R8: $e + \text{Ar} \rightarrow 2e + \text{Ar}^+$ is the most important ionization source only near the sheath
 - R22: $\text{Ar}^m + \text{Ar}^m \rightarrow e + \text{Ar}^+ + \text{Ar}$ is also important in the sheath region
 - R19: $e + \text{Ar}^m \rightarrow 2e + \text{Ar}^+$ plays a significant role in the bulk



Pressure dependence



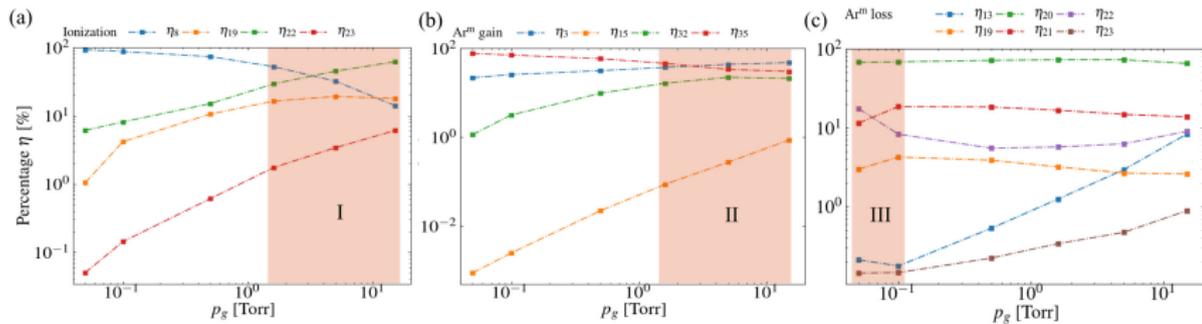
Wen et al. (2021) PSST **30** 105009

- Percentage (η_j) of the total reaction rate of each reaction j versus background pressure p_g

Ionization

- R8: $e^- + \text{Ar} \rightarrow 2e^- + \text{Ar}^+$ dominates at low pressure
- R22: $\text{Ar}^m + \text{Ar}^m \rightarrow e^- + \text{Ar}^+ + \text{Ar}$ – Penning ionization
- R19: $e^- + \text{Ar}^m \rightarrow e^- + \text{Ar}^+ + \text{Ar}$ – step wise ionization take over at higher pressure

Pressure dependence



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- Ar^m gain, $j \in \{3, 15, 32, 35\}$
 - R35: $\text{Ar}^r \rightarrow \text{Ar}^m + h\nu$ dominates at low pressure
 - R3: $e + \text{Ar} \rightarrow \text{Ar}^m + e$ dominates at high pressure
- Ar^m loss, $j \in \{13, 19 - 23\}$
 - R20: $e + \text{Ar}^m \rightarrow e + \text{Ar}^r$ always dominates the loss



Surface effects – secondary electron emission



Surface effects

- One-dimensional particle-in-cell/Monte Carlo collisional simulations were performed on a capacitive 2.54 cm gap, 1.6 Torr argon discharge driven by a sinusoidal rf current density amplitude of 50 A/m² at 13.56 MHz

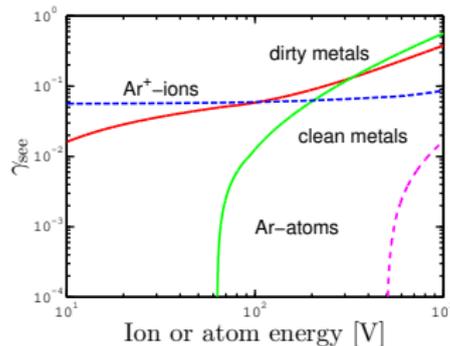
Table 3. An overview of the four cases explored.

Case	Ar ^m , Ar ^r , Ar(4p)	Secondary electrons			
		Ion induced	Ground state neutrals	Excited species	Reflected
A	Neglected	Neglected	Neglected	Neglected	Neglected
B	Neglected	0.15	Neglected	Neglected	Neglected
C	Neglected	$\gamma_{\text{sec}}(\mathcal{E}_i)$	Neglected	Neglected	Neglected
D	Included	$\gamma_{\text{sec}}(\mathcal{E}_i)$	$\gamma_{\text{sec}}(\mathcal{E}_n)$	Included	0.2

Surface effects

Table 2. The parameters of the simulation, the energy threshold above which the PIC dynamics of the neutral particles are followed, the wall quenching and reflection coefficients, and secondary electron emission yield upon particle impact.

Species	\mathcal{E}_{thr} (mV)	γ_{q}	γ_{r}	γ_{sec}
Ar	1000	—	1.0	$f(\mathcal{E}_n)$ [46, 47]
Ar ^m	50	0.5	0.5 [45]	0.21 [48]
Ar ^f	50	0.5	0.5 [45]	0.21 [48]
Ar(4p)	50	0.5	0.5 [45]	0.27 [48]
Ar ⁺	—	—	—	$f(\mathcal{E}_i)$ [46, 47]
e	—	—	0.2 [49]	—



Gudmundsson et al. (2021) PSST **30** 125011

based on Phelps and Petrović (1999) PSST **8** R21

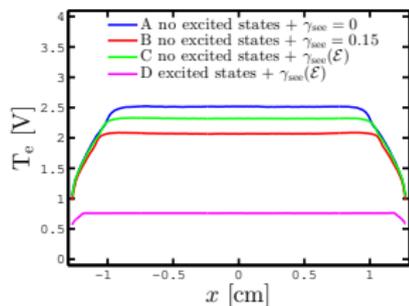
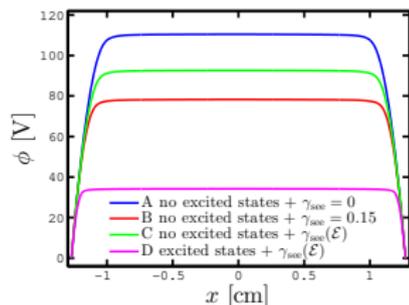
■ Secondary electron emission

- Ion induced, energy dependent
- Due to bombardment of neutrals in the ground state
- Due to bombardment of excited neutrals



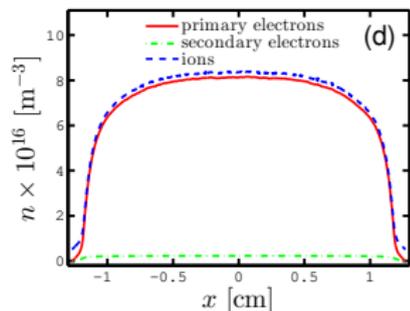
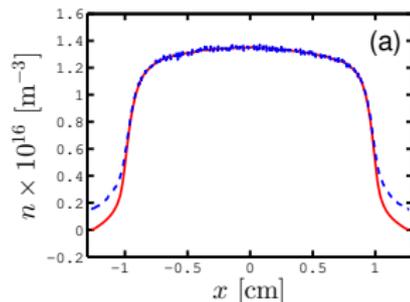
Surface effects

- Adding secondary electron emission from the electrodes to the discharge model decreases the time averaged potential, and adding excited states to the discharge, decreases the potential much further
- The electron temperature profile when the excited state kinetics, energy dependent secondary electron emission (both ion and atom induced) and electron reflection are included has a significantly lower value, about 0.76 V, indicating γ -mode operation



Surface effects

- The time averaged charged particle densities
 - neglecting excited states and secondary electron emission
 - including excited state kinetics and energy dependent secondary electron emission due to ions and atoms bombarding the electrodes, as well as electron reflection
- For a parallel plate capacitive argon discharge at 1.6 Torr with a gap separation of 2.54 cm driven by a 50 A m⁻² sinusoidal current source at 13.56 MHz



Surface effects

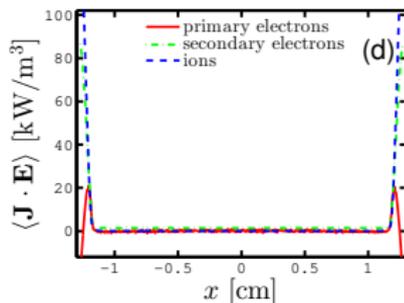
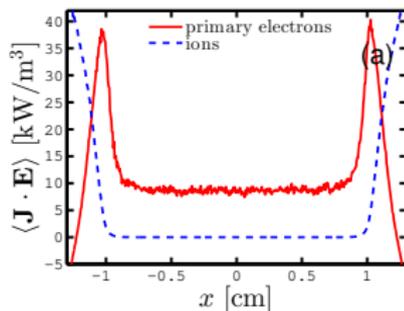
- The time averaged particle power absorption profiles

Upper graph neglecting excited states and secondary electron emission

- The power absorption by the primary electrons within the plasma bulk is roughly 8.7 kW/m^3

Lower graph including excited state kinetics and secondary electron emission

- The primary electrons in the bulk region absorbs almost no power $\sim 0 \text{ W/m}^3$, while the secondary electrons absorb 1.4 kW/m^3



Surface effects

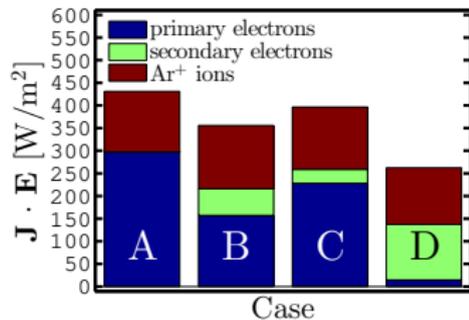
- The time averaged power absorption by the various species

Case A: no excited state kinetics nor secondary electron emission

Case B: no excited state kinetics but constant secondary electron emission

Case C: no excited state kinetics and energy dependent secondary electron emission

Case D: including excited state kinetics, energy dependent secondary electron emission due to ions and neutrals, and electron reflection

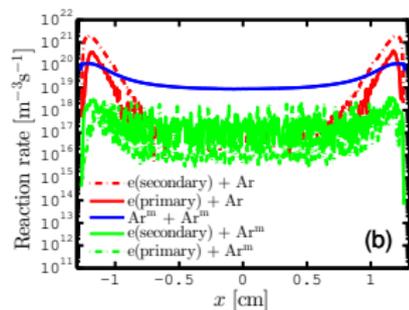
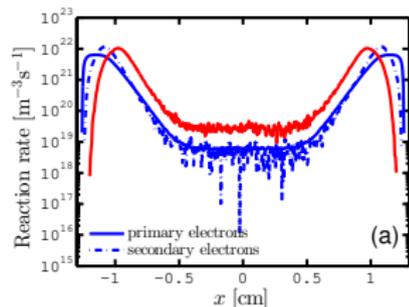


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Surface effects

- The reaction rates for ionization processes neglecting excited states and secondary electrons (red) and including constant secondary electron emission (blue)
- The reaction rates for ionization processes including excited state kinetics, energy dependent secondary electron emission due to ion and atom bombardment of the electrodes, and electron reflection



Surface effects

- Electron impact ionization of ground state argon atoms by secondary electrons dominates (75.7 % contribution) and by primary electrons (10.9 % contribution)
- The third most important process is Penning ionization (metastable pooling) $\text{Ar}^m + \text{Ar}^m \rightarrow e + \text{Ar} + \text{Ar}^+$, which has about a 12.7 % contribution
- Electron impact ionization of the metastable argon atom (multi-step ionization) is small, contributing roughly 0.3 % to the total ionization.

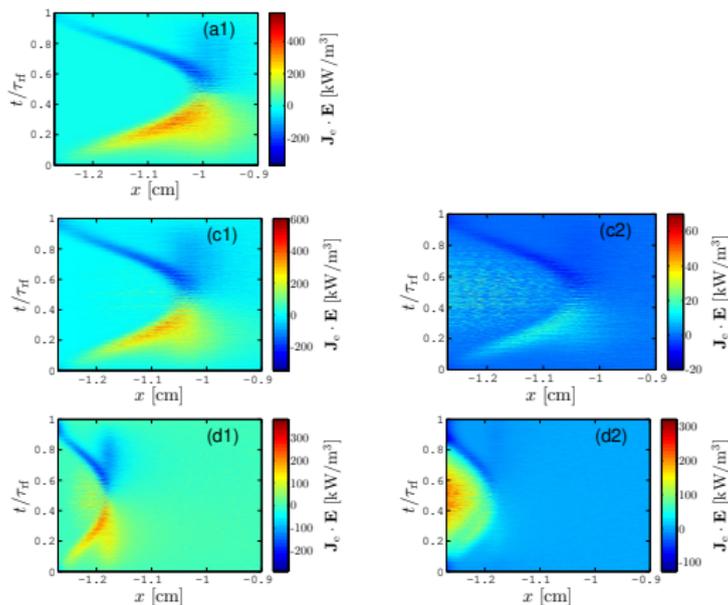
Table 4. The relative contribution of the various ionization processes within the argon discharge for case D.

Reaction	Contribution (%)
$e(\text{primary}) + \text{Ar} \rightarrow \text{Ar}^+ + 2e$	10.9
$e(\text{secondary}) + \text{Ar} \rightarrow \text{Ar}^+ + 2e$	75.7
$e(\text{primary}) + \text{Ar}^m \rightarrow e + \text{Ar}^+$	0.13
$e(\text{secondary}) + \text{Ar}^m \rightarrow e + \text{Ar}^+$	0.18
$\text{Ar}^m + \text{Ar}^m \rightarrow e + \text{Ar} + \text{Ar}^+$	12.7
$\text{Ar}^f + \text{Ar}^m \rightarrow e + \text{Ar} + \text{Ar}^+$	0.39
$\text{Ar}^f + \text{Ar}^f \rightarrow e + \text{Ar} + \text{Ar}^+$	0.0077
$\text{Ar}(4p) + \text{Ar}(4p) \rightarrow e + \text{Ar} + \text{Ar}^+$	1×10^{-8}
$e(\text{primary}) + \text{Ar}(4p) \rightarrow e + \text{Ar}^+$	—
$e(\text{secondary}) + \text{Ar}(4p) \rightarrow e + \text{Ar}^+$	0.0017

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Surface effects



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- The spatio-temporal behavior of the electron power absorption primary electrons (left) secondary electrons (right)

Summary



Surface effects

- When the excited states, and secondary electron emission due to neutral and ion impact on the electrodes are included in the discharge model, the discharge operation transitions from α -mode to γ -mode, in which nearly all the ionization is due to secondary electrons
- Secondary electron production due to the bombardment of excited argon atoms was approximately 14.7 times greater than that due to ion bombardment
- Electron impact of ground state argon atoms by secondary electrons contributes about 76% of the total ionization; primary electrons, about 11%; metastable Penning ionization, about 13%; and multi-step ionization, about 0.3%



Thank you for your attention

The slides can be downloaded at

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

- Gudmundsson, J. T., J. Krek, D.-Q. Wen, E. Kawamura, and M. A. Lieberman (2021). Surface effects in a capacitive argon discharge in the intermediate pressure regime. *Plasma Sources Science and Technology* 30(12), 125011.
- Kawamura, E., M. A. Lieberman, A. J. Lichtenberg, and P. Chabert (2020). Particle-in-cell simulations and passive bulk model of collisional capacitive discharge. *Journal of Vacuum Science and Technology A* 38(2), 023003.
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- Wen, D.-Q., J. Krek, J. T. Gudmundsson, E. Kawamura, M. A. Lieberman, and J. P. Verboncoeur (2021). Benchmarked and upgraded particle-in-cell simulations of capacitive argon discharge at intermediate pressure: The role of metastable atoms. *Plasma Sources Science and Technology* 30(10), 105009.

