63rd Gaseous Electronics Conference & 7th International Conference on Reactive Plasmas

Progress on simulations of multiple-frequency capacitively coupled discharges

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Ruhr-University Bochum, Germany

8 October 2010

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Why multiple-frequency discharges? - Control of ion properties

Inspiration: ICPIG 2005 - M. A. Lieberman: "*Plasma processing in the 21st century*"

Budapest - Belgrade (Z. Petrović) collaboration 2005-2007, Dual-frequency CF_4, CF_4 / Ar discharges

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Budapest - Bochum collaboration, 2007 ➟

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- Electric field reversals in capacitively coupled radio frequency discharges
- Electrical Asymmetry Effect: separate control of ion flux and energy, optimization
- Self-excited nonlinear plasma series resonance oscillations
- Different modes of electron heating
- Charge dynamics in CCRF discharges
- Excitation dynamics in CCRF discharges
- Power dissipation under EAE operation
- The Electrcal Asymmetry Effect in multi-frequency discharges
- Effects of secondary electrons in dual-frequency discharges

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- Charge dynamics in CCRF discharges
- Excitation dynamics in CCRF discharges - **poster by J. Schulze**
- Power dissipation under EAE operation - **poster by E. Schüngel**
- The Electrcal Asymmetry Effect in multi-frequency discharges - **talk by J. Schulze**
- **Effects of secondary electrons in dual-frequency discharges** - **this talk**

The effect of secondary electrons in dual-frequency CCRF discharges

- Many simulation studies assume $\gamma = 0$ (incl. some of ours)
- Dielectric materials often have high secondary emission yield (0.3 .. 0.4) → secondary electrons can have a major influence on the behavior of plasma processing discharges
- At some conditions fast neutrals may as well contribute significantly to secondary emission [e.g. M. M. Turner, High-Frequency Gas Breakdown Workshop, 4 Oct 2010, Paris] (known for DC, too) (here we concentrate on ions only, no fast neutrals, no metastables ...)

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- The effect of secondary electrons on single-frequency discharges ✓
- Ion properties in single frequency discharges ✓
- Ion properties in dual-frequency discharges : success and limitations ✓
- The effect of secondary electrons on the separate control of ion flux and energy
  - in classical dual-frequency discharges
  - in discharges operated under the conditions of the Electrical Asymmetry Effect
- Method: <u>1d3v (standard) PIC/MCC simulation</u> + theoretical analysis + experiments

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INTRODUCTION:

- The effect of secondary electrons on single-frequency discharges ✓
- Ion properties in single frequency discharges ✓
- Ion properties in dual-frequency discharges : success and limitations ✓

RESULTS:

- The effect of secondary electrons on the separate control of ion flux and energy
  - in classical dual-frequency discharges
  - in discharges operated under the conditions of the Electrical Asymmetry Effect

- Method: 1d3v (standard) PIC/MCC simulation + theoretical analysis + experiments

Single-frequency discharges:

alpha / gamma modes,
flux-energy distribution of ions,
ion flux & energy

Alpha / gamma modes in a single-frequency discharge: fluid model

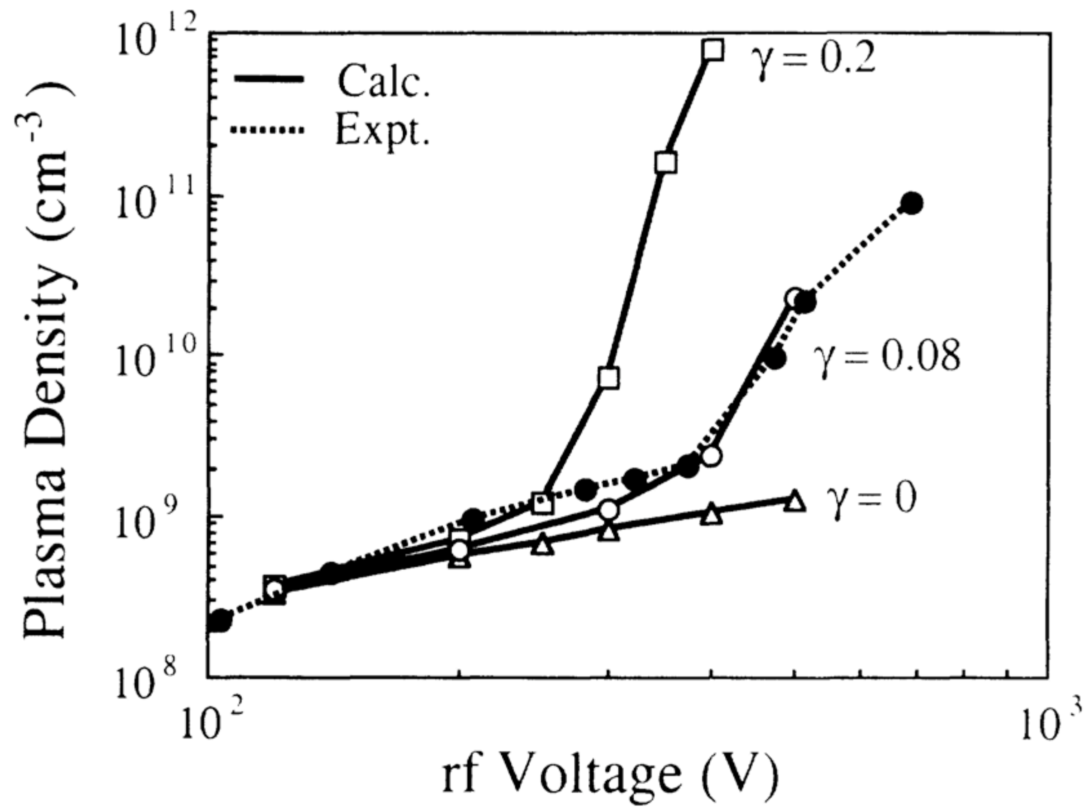
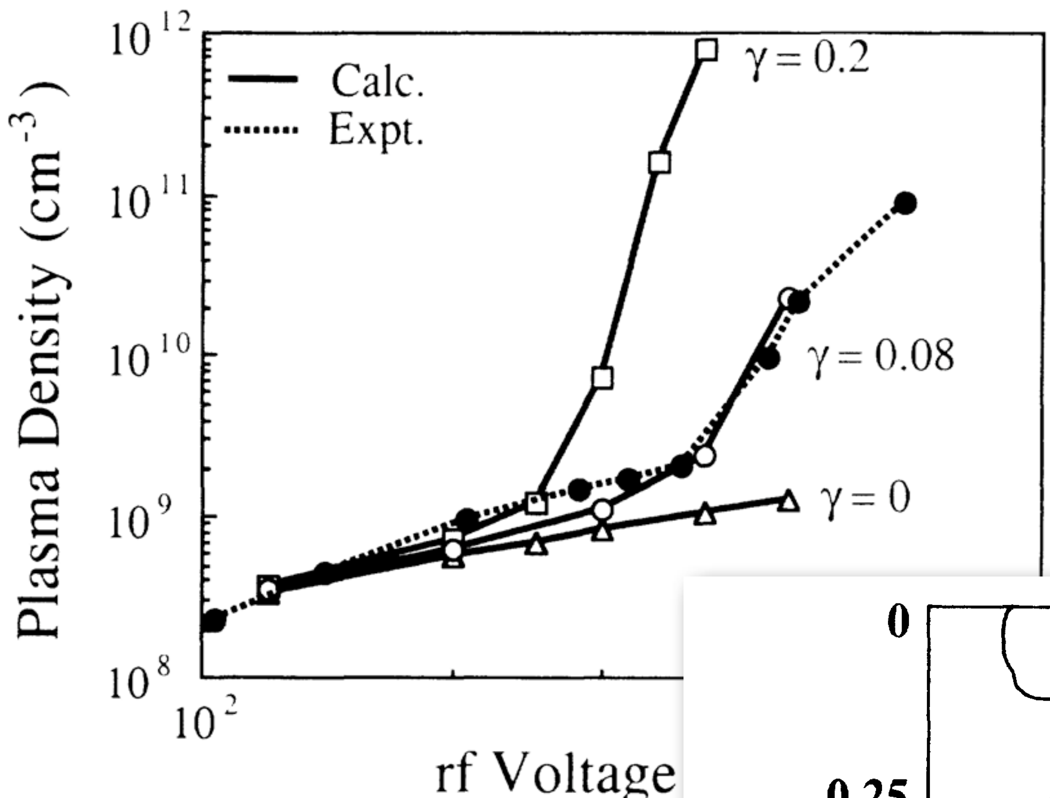


FIG. 2. Variations of the plasma density as a function of the rf voltage amplitude for a discharge in helium ($p = 3$ Torr, $d = 3$ cm, $F = 3.2$ MHz) for three values of the secondary electron emission coefficient ($\gamma = 0.0$, 0.08, and 0.2). The experimental results of Godyak and Kanneh (Ref. 7) at 3.2 MHz are also represented.

Ph. Belenguer and J.-P. Boeuf, Phys. Rev. A 41, 4447 (1990)

Alpha / gamma modes in a single-frequency discharge: fluid model



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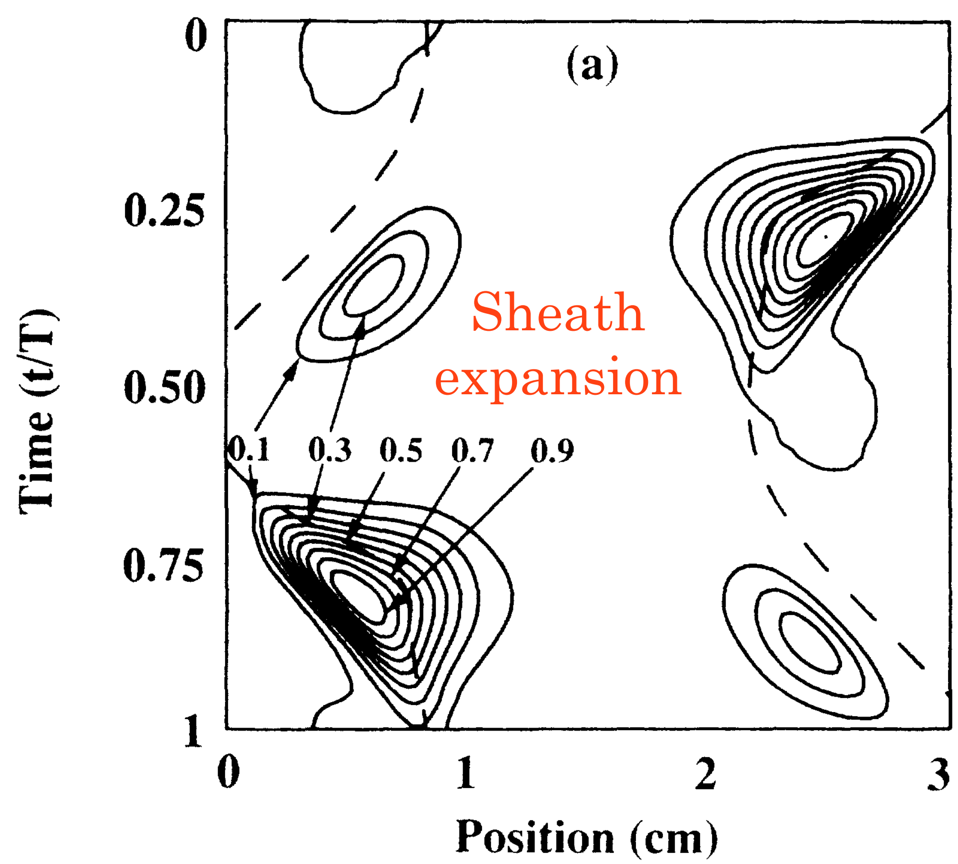
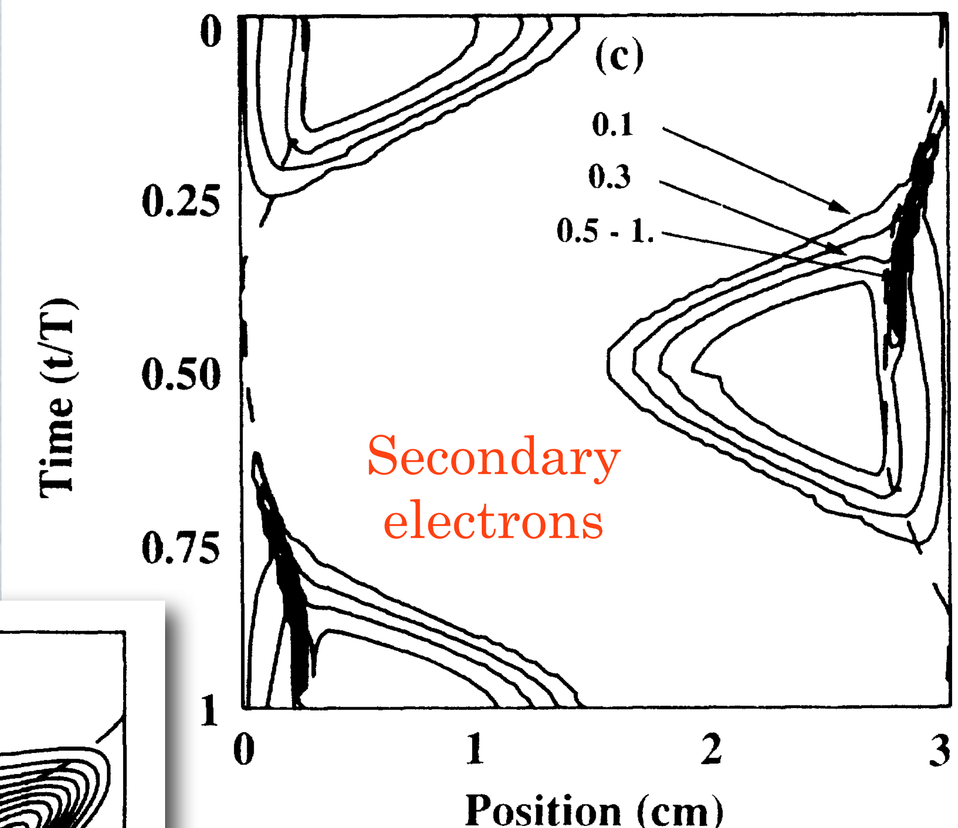
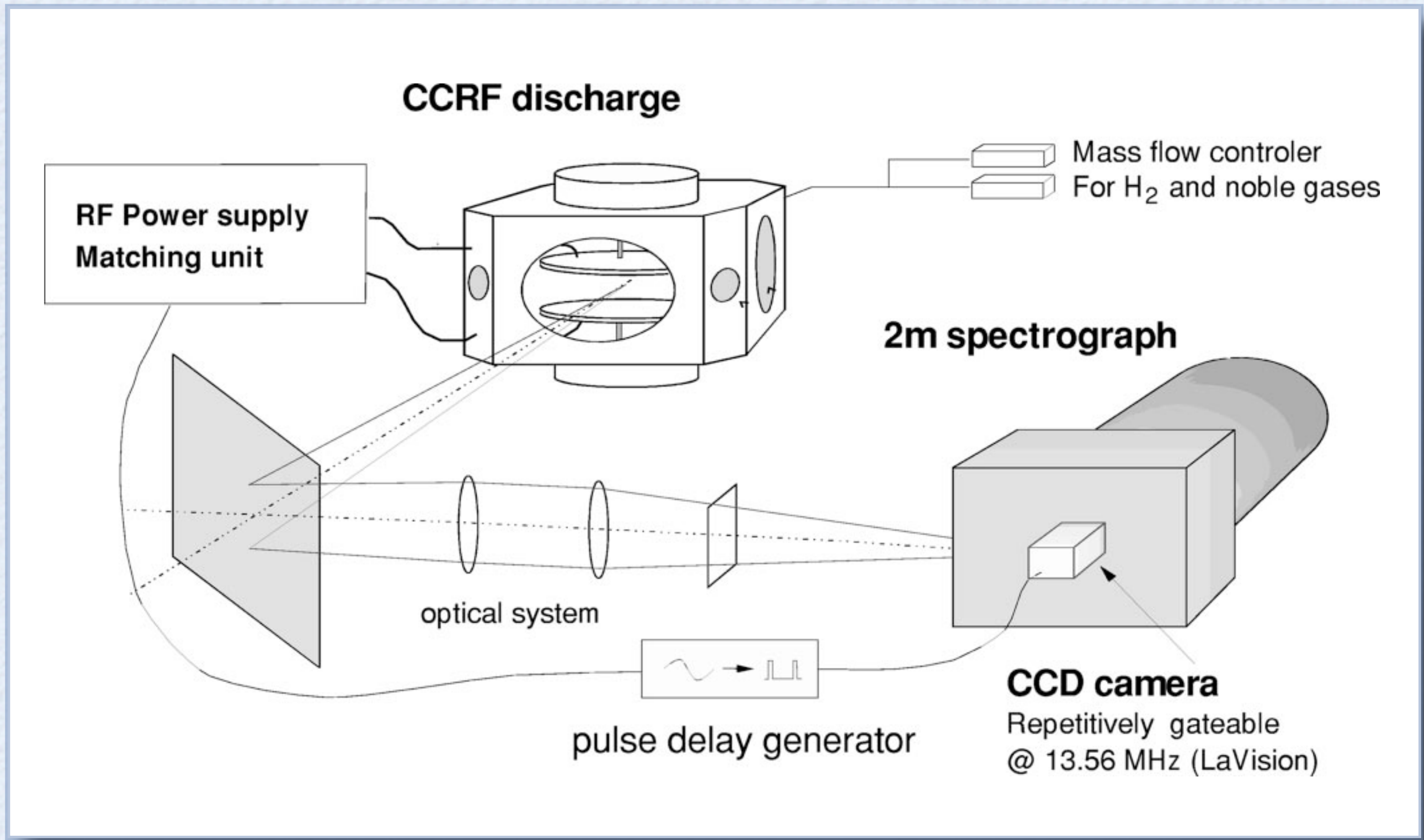


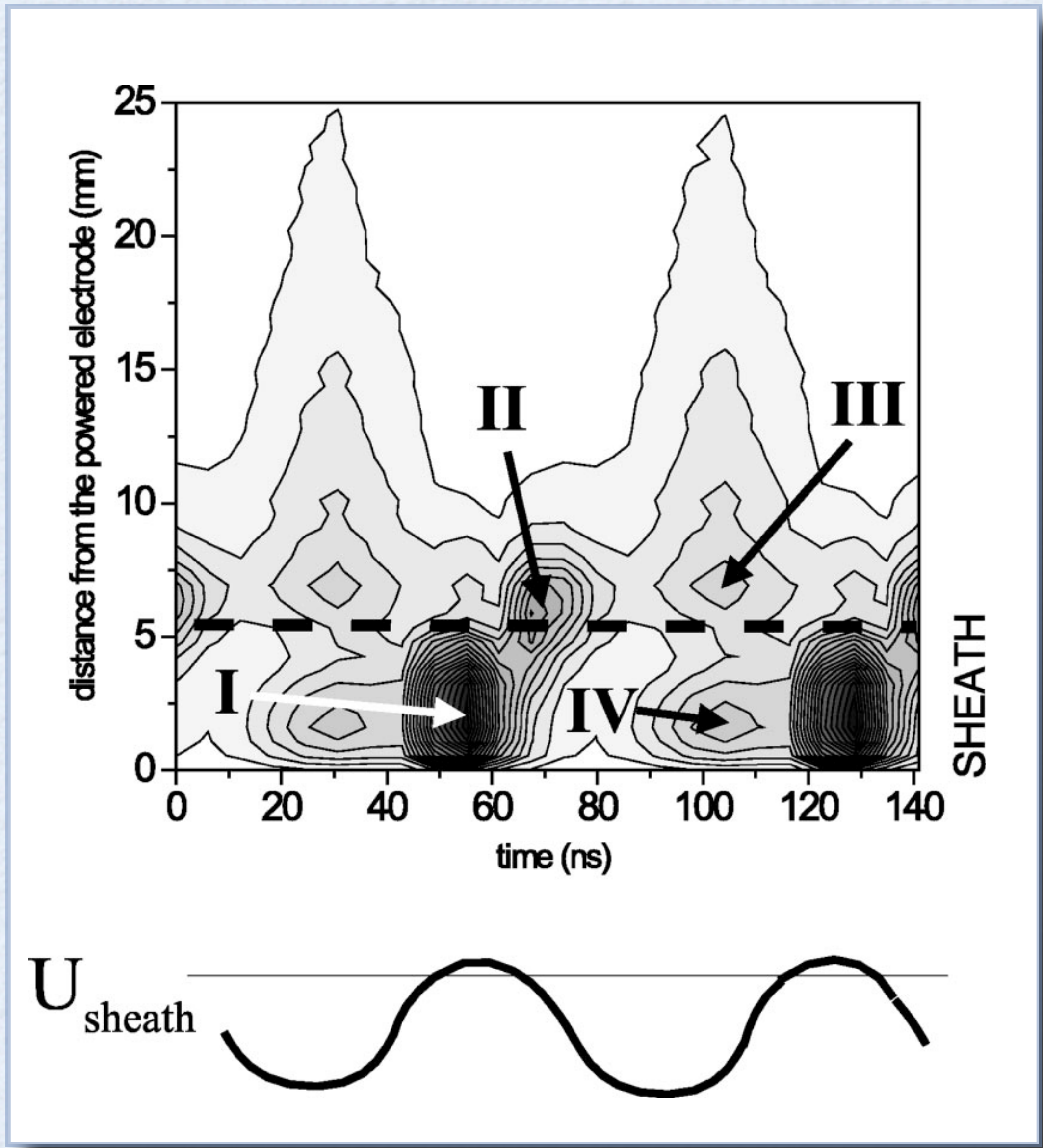


Ph. Belenguer and J.-P. Boeuf,
Phys. Rev. A 41, 4447 (1990)

Alpha / gamma modes in a single-frequency discharge: experiments

Gans T, Lin C C, Schulz-von der Gathen V and Döbele H F 2003 Phys. Rev. A 67 012707

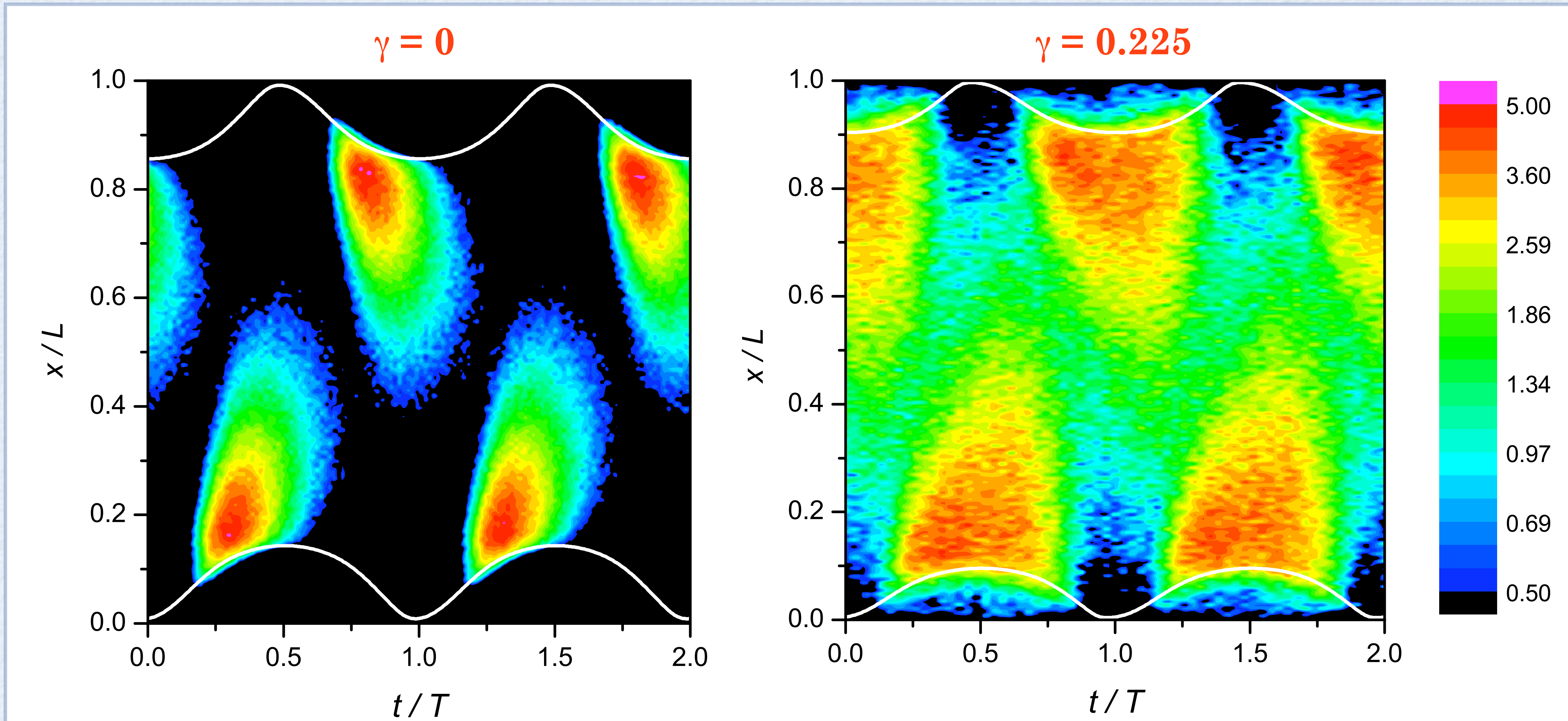




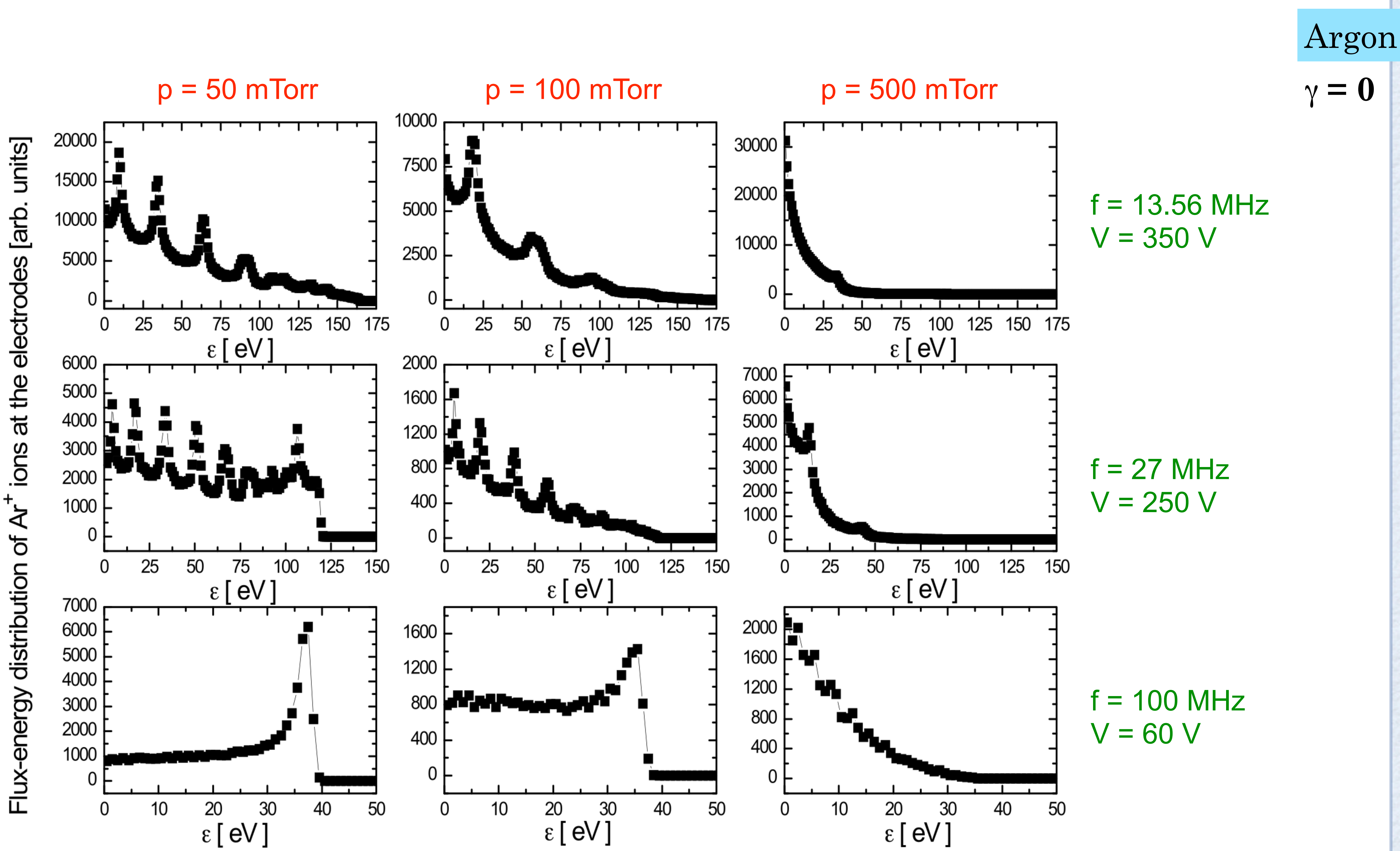
Phase Resolved Optical Emission Spectroscopy

Alpha / gamma modes in a single-frequency discharge: PIC simulation

$f = 27.12$ MHz, $p = 20$ Pa, $V = 300$ V, $L = 2.5$ cm



Tailoring the flux-energy distributions of ions reaching the electrodes



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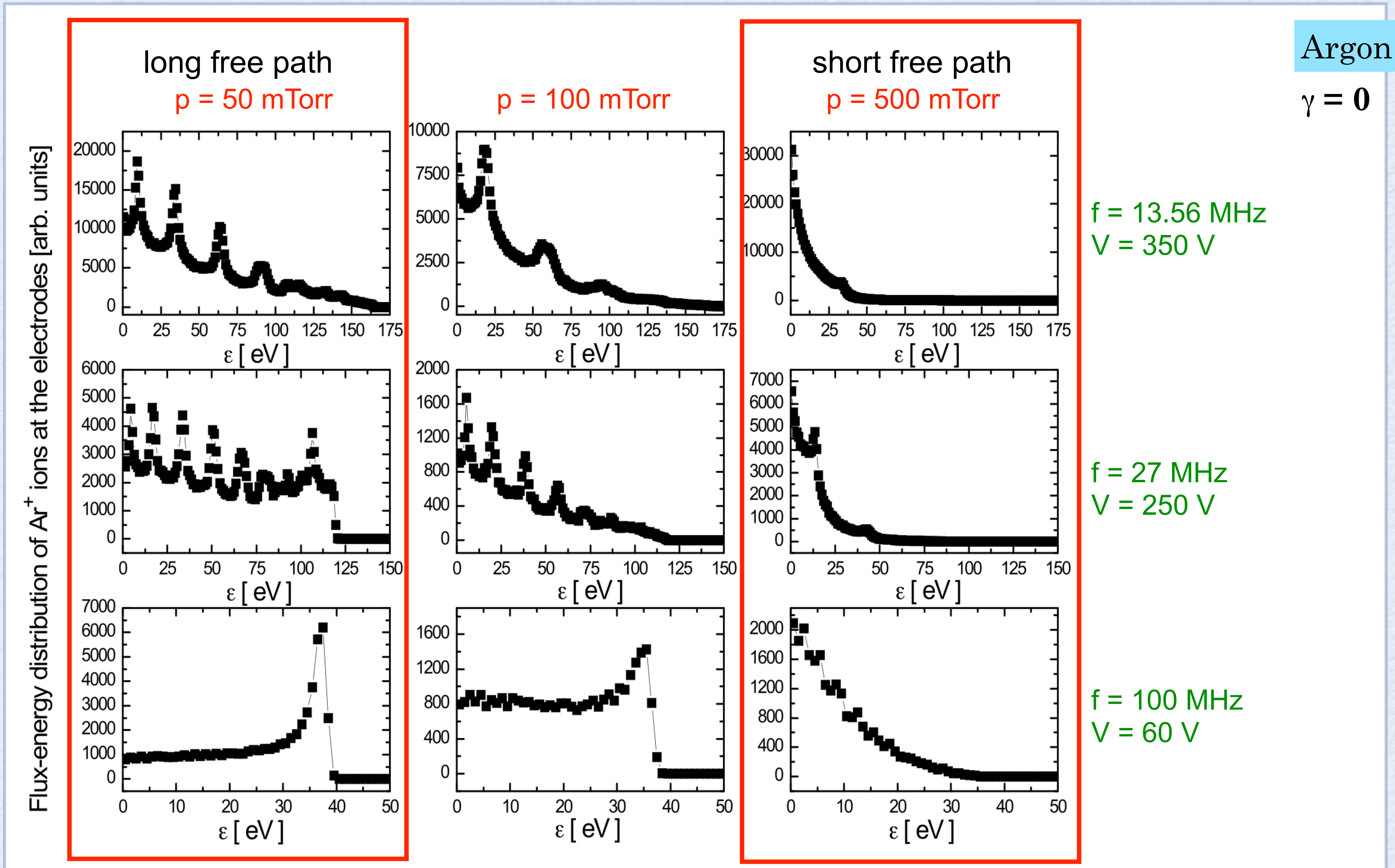

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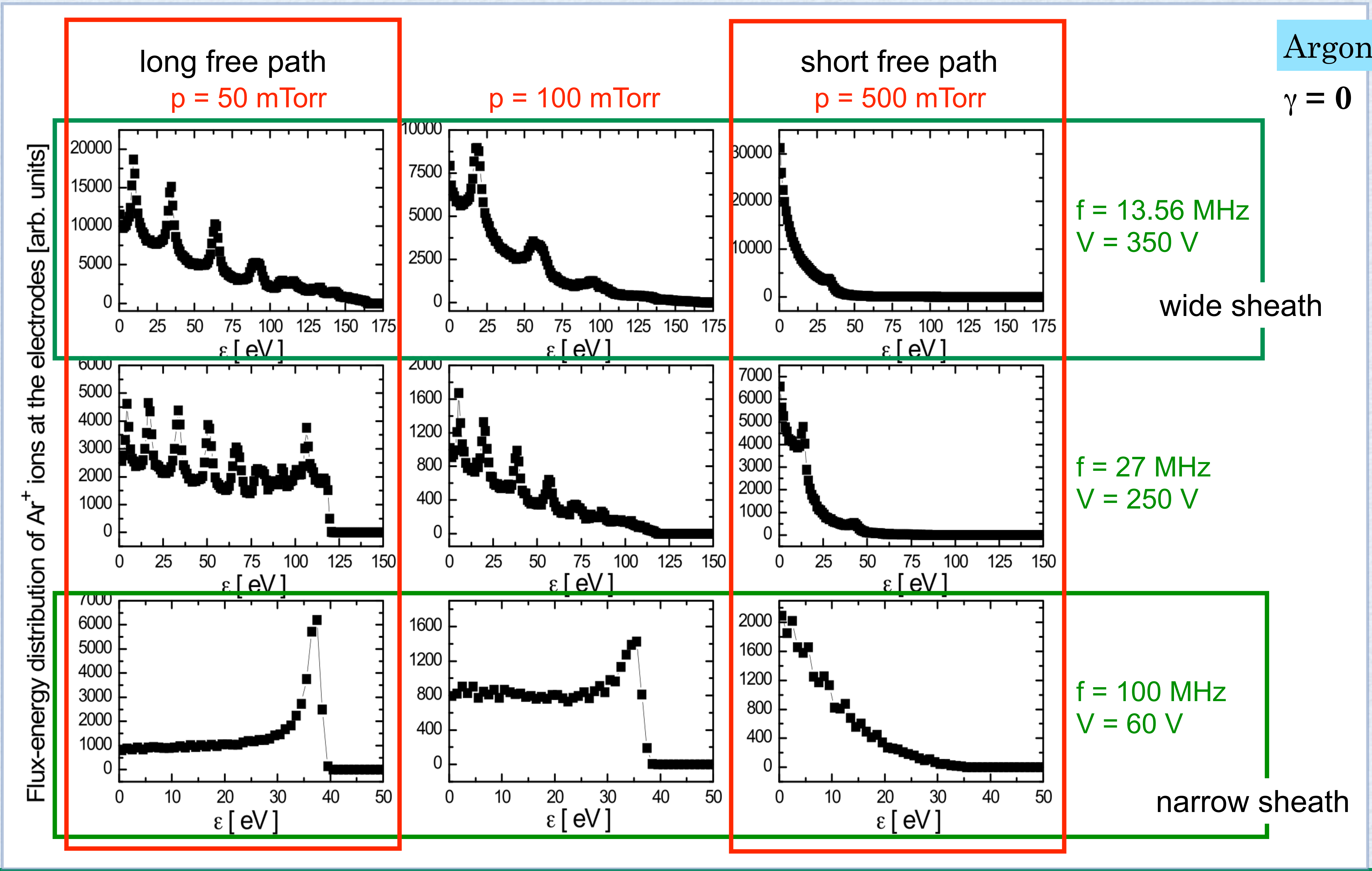

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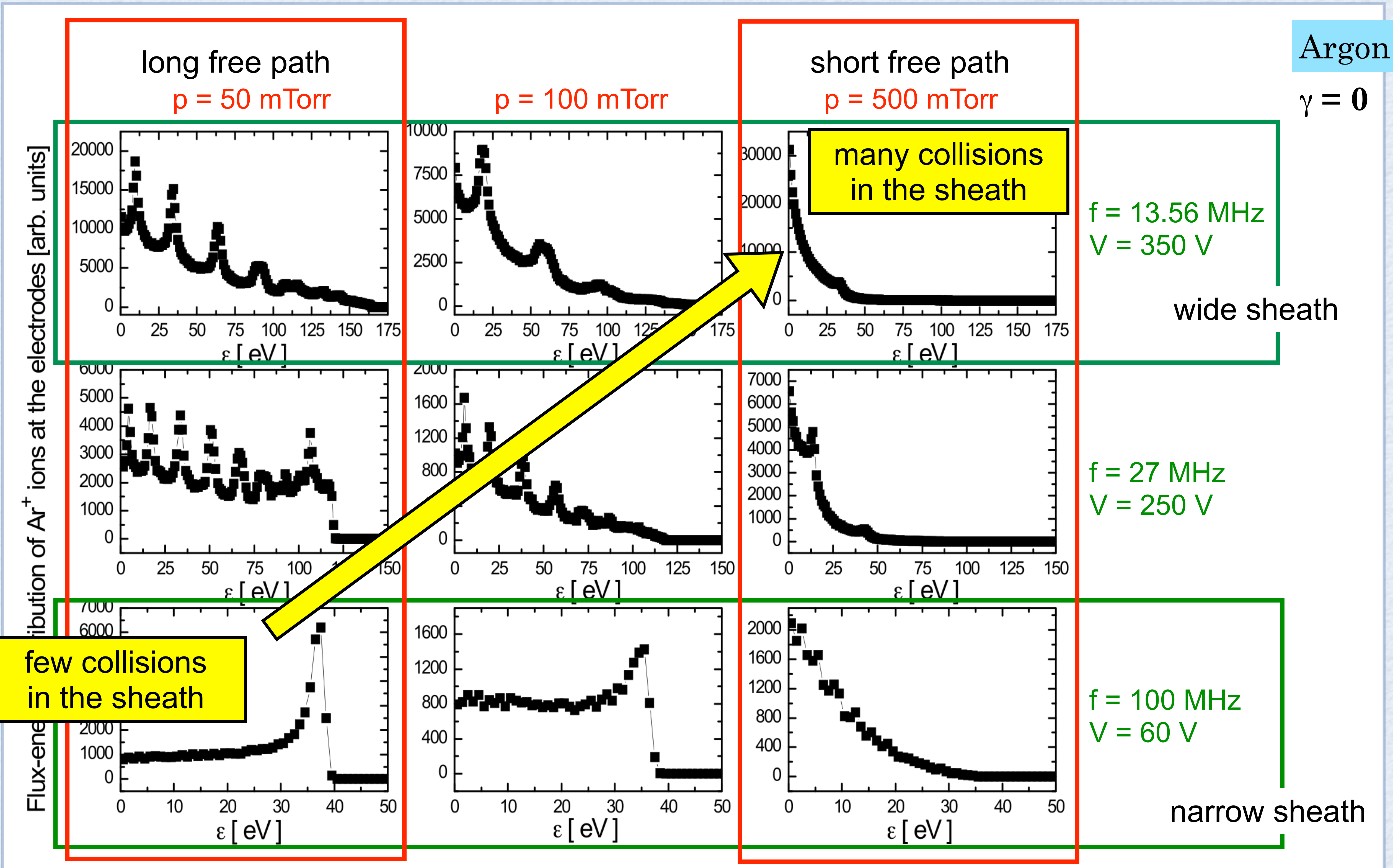
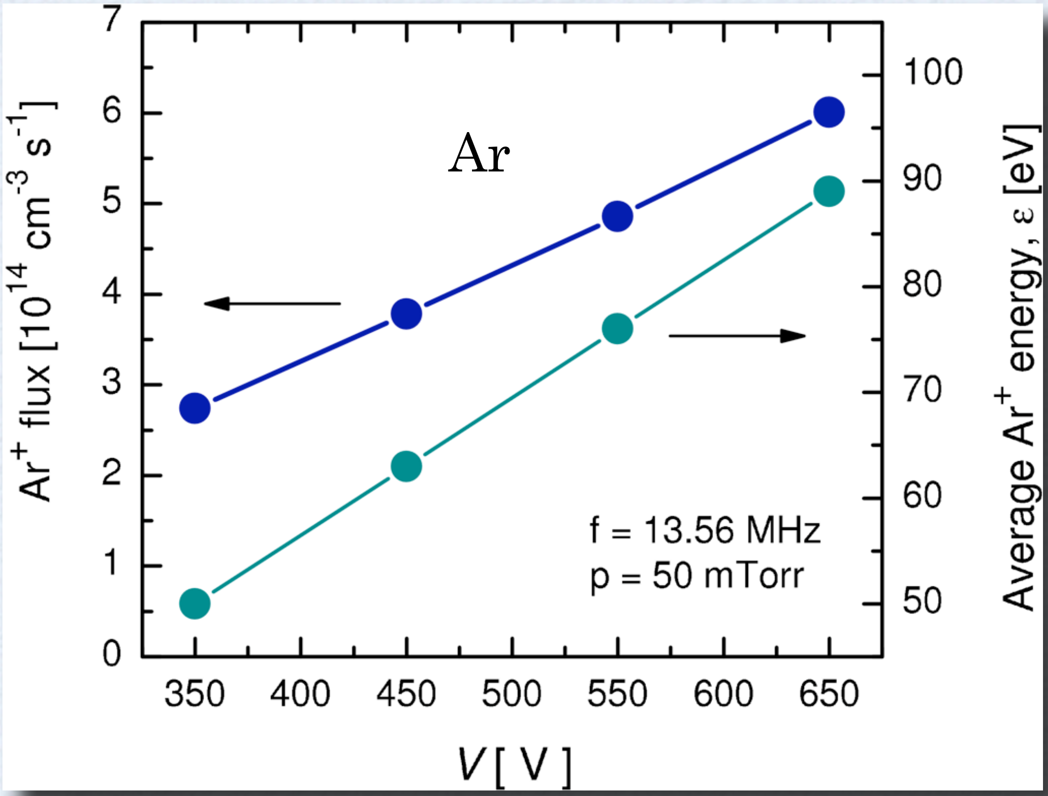


Ion properties vs. excitation voltage

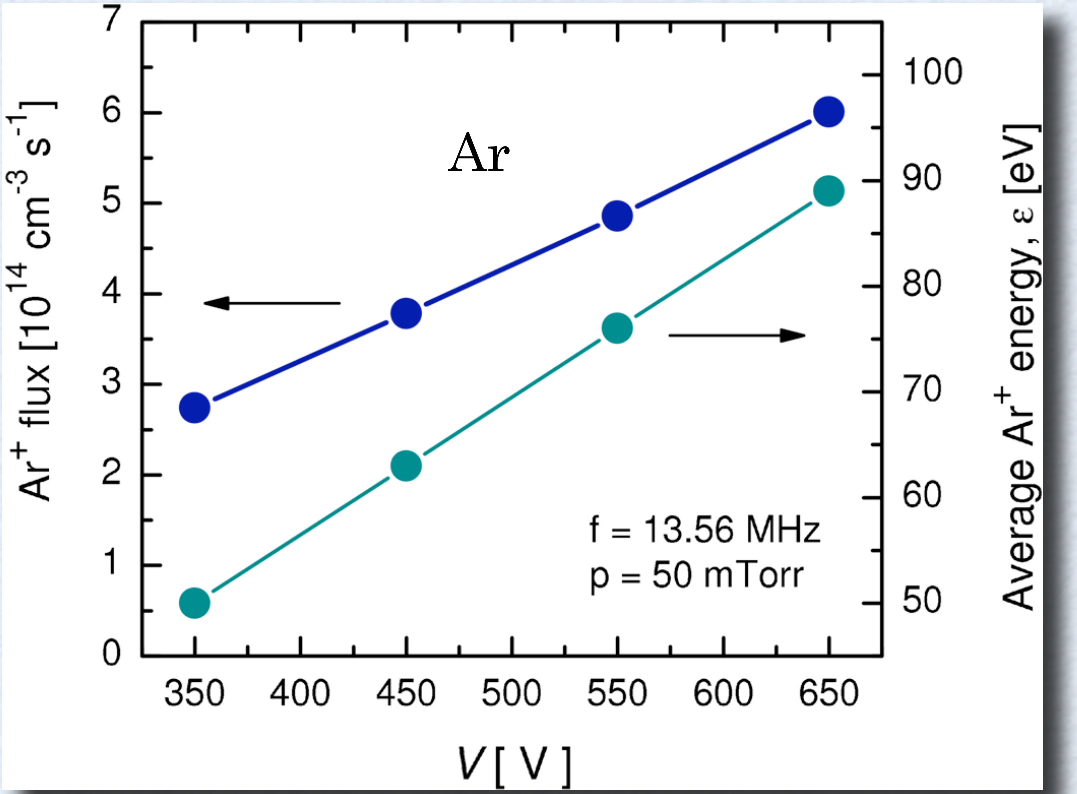
Single-frequency RF discharge



The RF voltage influences both the flux of ions and the average energy of the ions

Ion properties vs. excitation voltage

Single-frequency RF discharge



The RF voltage influences both the flux of ions and the average energy of the ions

Is there a way to control these ion properties (nearly) independently from each other?

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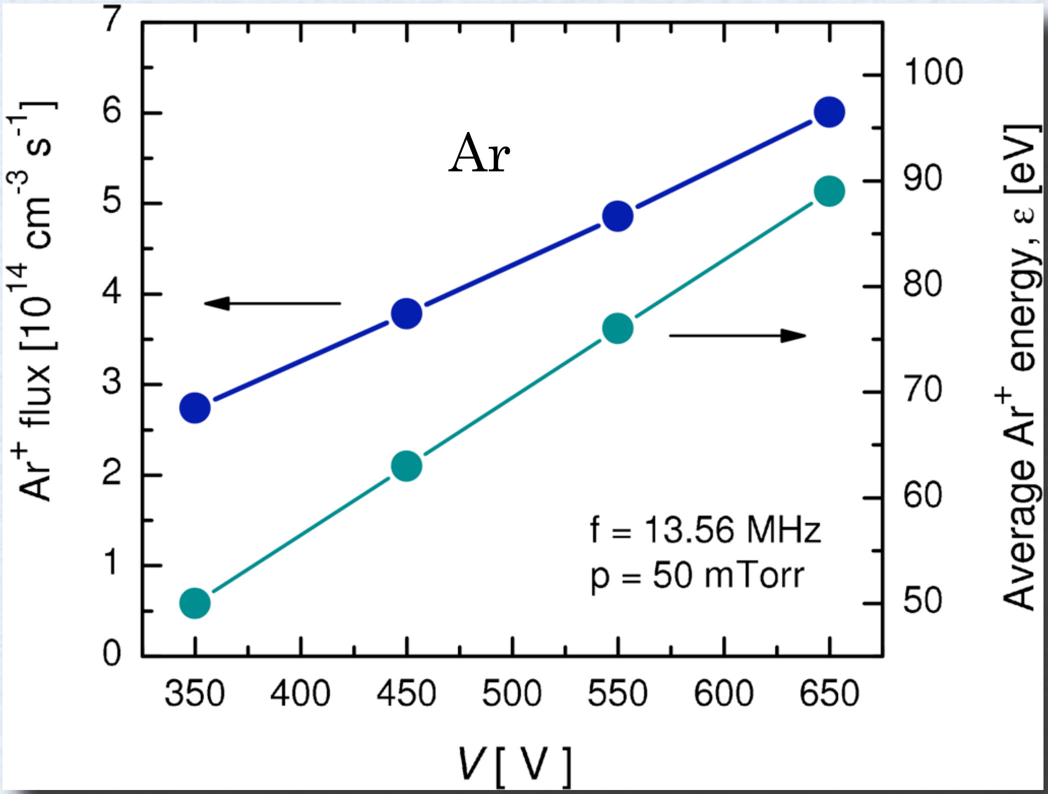

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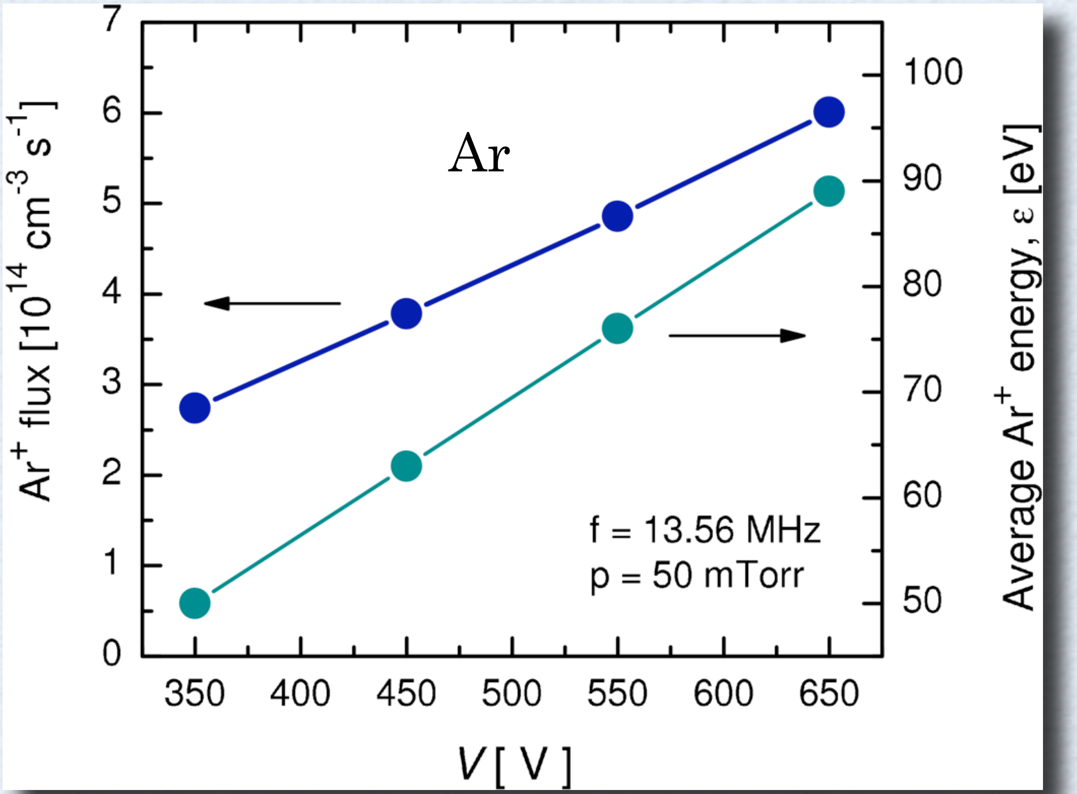
Dual frequency excitation

$$V(t) = V_{\mathrm{HF}} \sin(2\pi f_{\mathrm{HF}} t) + V_{\mathrm{LF}} \sin(2\pi f_{\mathrm{LF}} t)$$

Ion properties vs. excitation voltage

Single-frequency RF discharge



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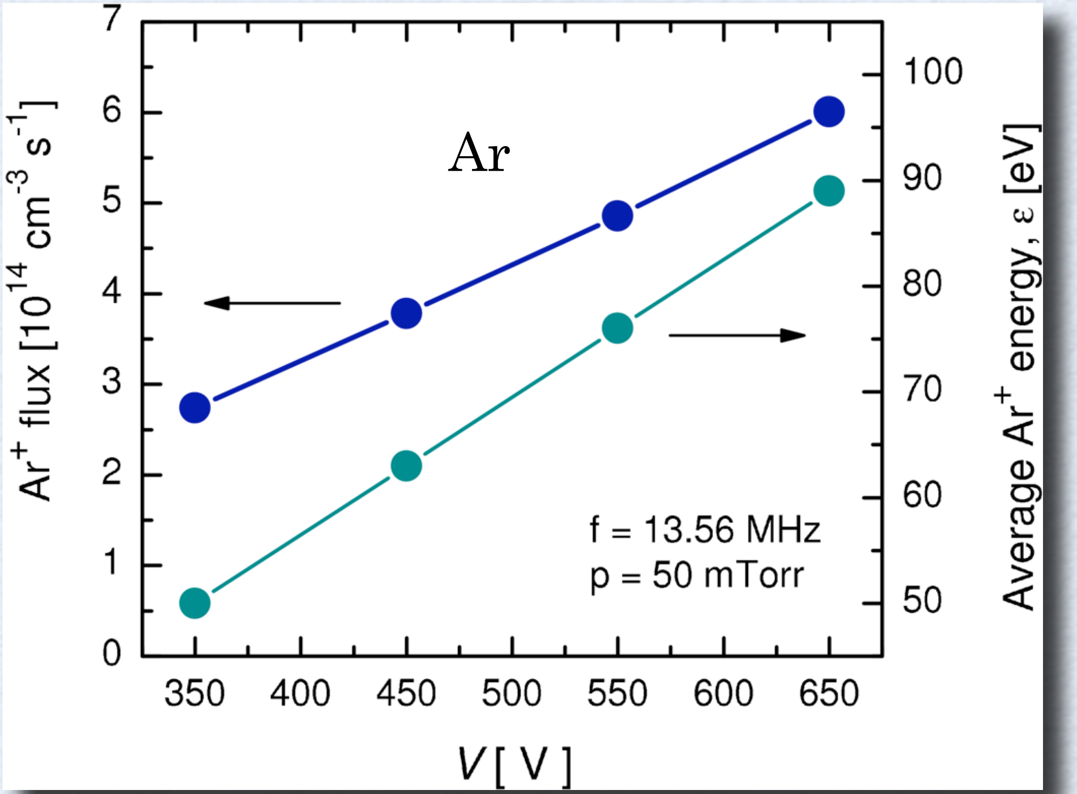
Heating & ion flux

Ion energy

Ion properties vs. excitation voltage

Single-frequency RF discharge



The RF voltage influences both the flux of ions and the average energy of the ions

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$$V(t) = V_{\mathrm{HF}} \sin(2\pi f_{\mathrm{HF}} t) + V_{\mathrm{LF}} \sin(2\pi f_{\mathrm{LF}} t)$$

Heating & ion flux — **Ion energy**

Goto H H, Löwe H D, and Ohmi T **1992**, J. Vac. Sci. Technol. A 10, 3048
Kitajima T, Takeo Y, Petrovic Z L and Makabe T 2000 Appl. Phys. Lett. 77 489
Boyle P C , Ellingboe A R and Turner M M 2004 Plasma Sources Sci. Technol. 13 493-503, J. Phys. D. 37 697
Denda T, Miyoshi Y, Komukai Y, Goto T, Petrovic Z L and Makabe T 2004 J. Appl. Phys. 95 870
Lee J K, Manuilenko O V, Babaeva N Yu, Kim H C and Shon J W 2005 Plasma Sources Sci. Technol. 14 89
Kawamura E, Lieberman M A and Lichtenberg A J 2006 Phys. Plasmas 13 053506
Turner M M and Chabert P 2006 Phys. Rev. Lett. 96 205001
Gans T, Schulze J, O'Connell D, Czarnetzki U, Faulkner R, Ellingboe A R and Turner M M 2006 Appl. Phys. Lett. 89 261502
Schulze J, Gans T, O'Connell D, Czarnetzki U, Ellingboe A R and Turner M M 2007 J. Phys. D 40 7008-7018
Schulze J, Donko Z, Luggenhölscher D and Czarnetzki U 2009 Plasma Sources Sci. Technol. 18, 034011
Semmler E, Awakowicz P and von Keudell A 2007 Plasma Sources Sci. Technol. 16 839
Salabas A and Brinkmann R P 2005 Plasma Sources Sci. Technol. 14 2 53-59
Georgieva V and Bogaerts A 2006 Plasma Sources Sci. Technol., 15, 368-377

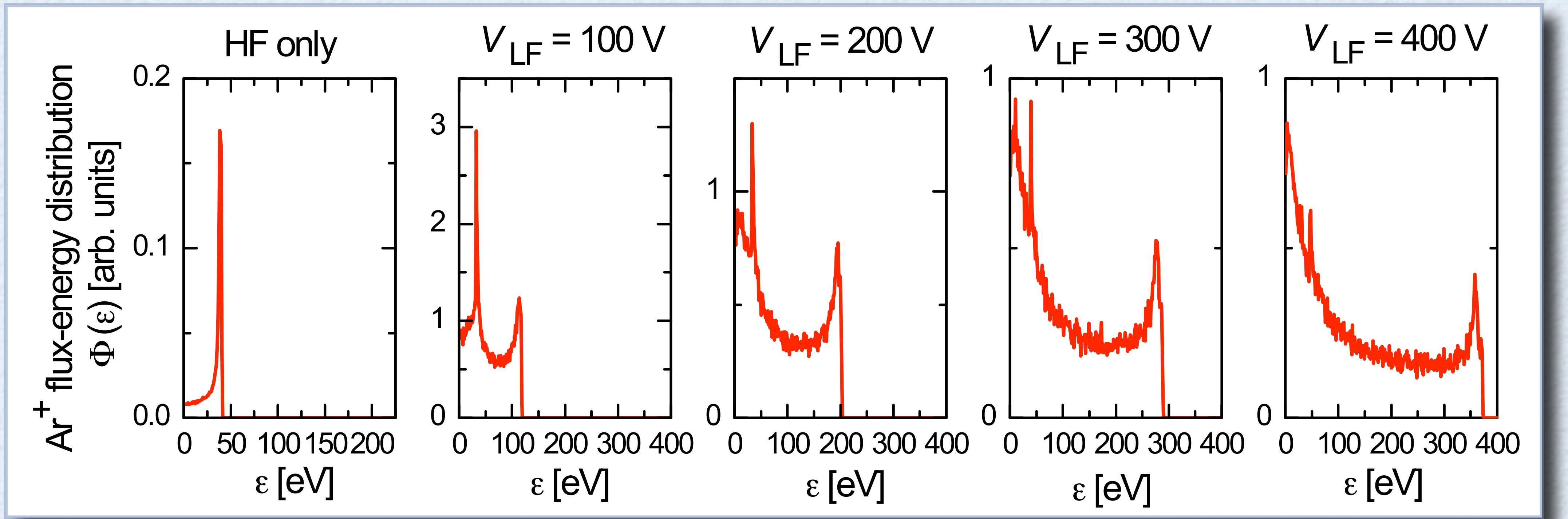
"Classical" dual-frequency discharges

and the influence of secondary electrons on the control of ion properties

Control of ion properties in "classical" dual-frequency discharges

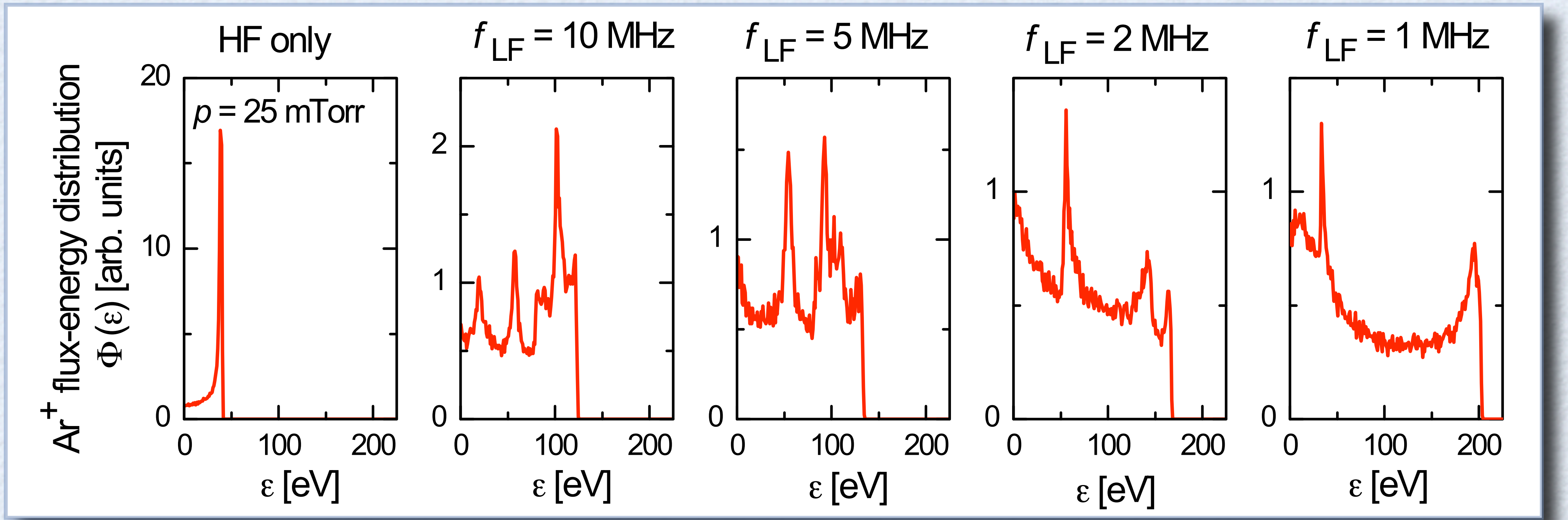
- Ar discharge
- p = 25 mTorr
- L = 2 cm
- V_{HF} = 60 V
- f_{HF} =100 MHz

Effect of LF voltage at f_{LF} =1 MHZ



γ = 0

Effect of LOW frequency at V_{LF} = 200V



Functional separation works best at **low pressures** and at $f_{HF} >> f_{LF}$

Control of ion properties in "classical" dual-frequency discharges

P. C. Boyle, A. R. Ellingboe and M. M. Turner, J. Phys. D 37, 697 (2004)

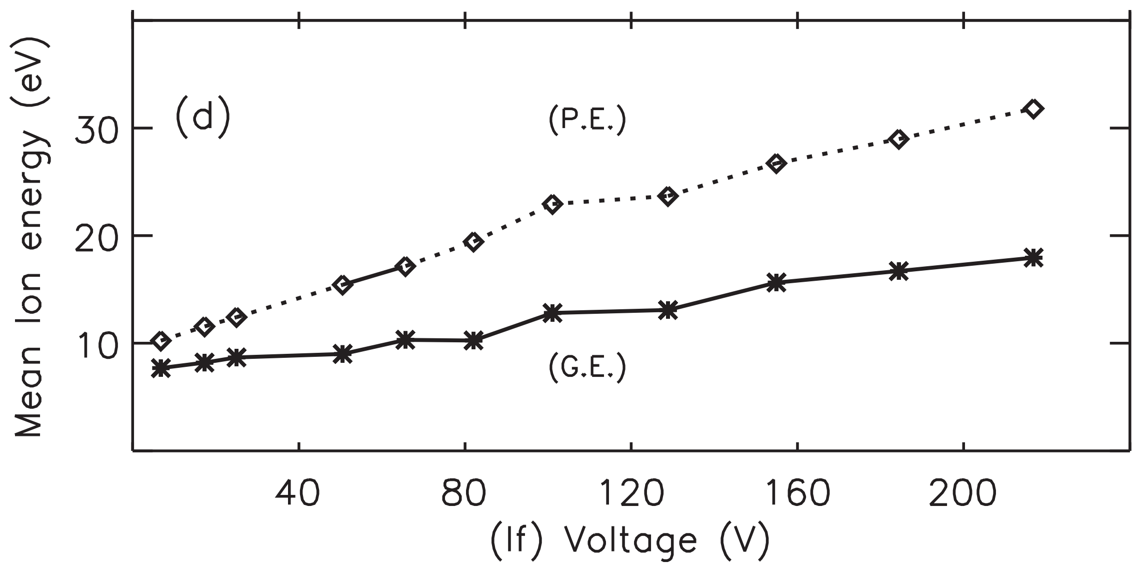


Figure 1. Dependence of plasma parameters on the low frequency voltage amplitude in a dual frequency device. The high frequency power is held constant at ≈1200 W m^{-2}. P.E. and G.E. refer to the powered and grounded electrodes, respectively.

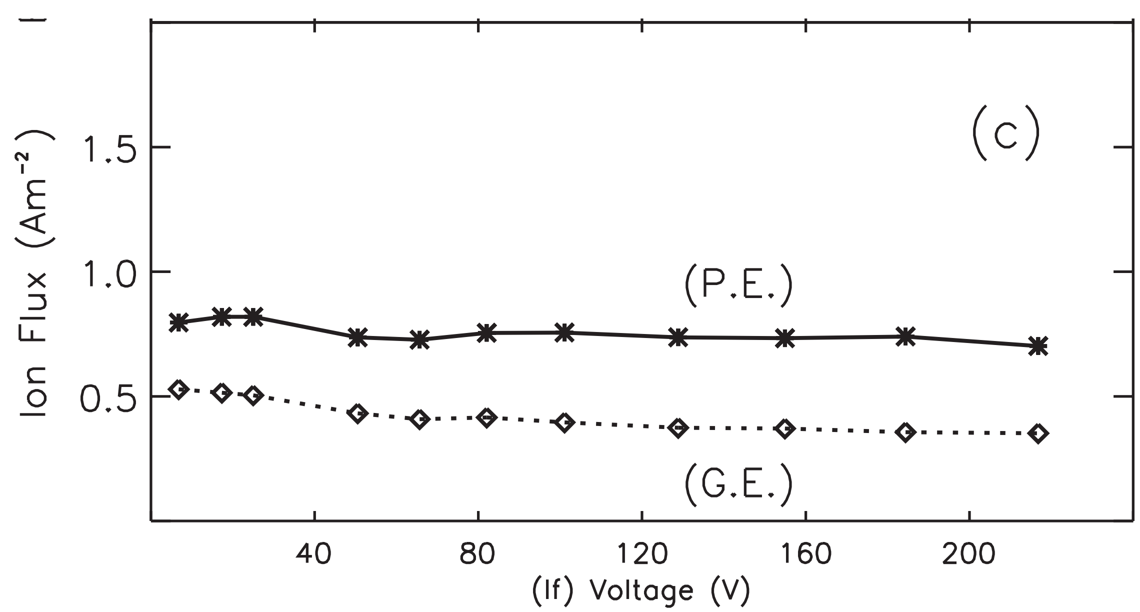
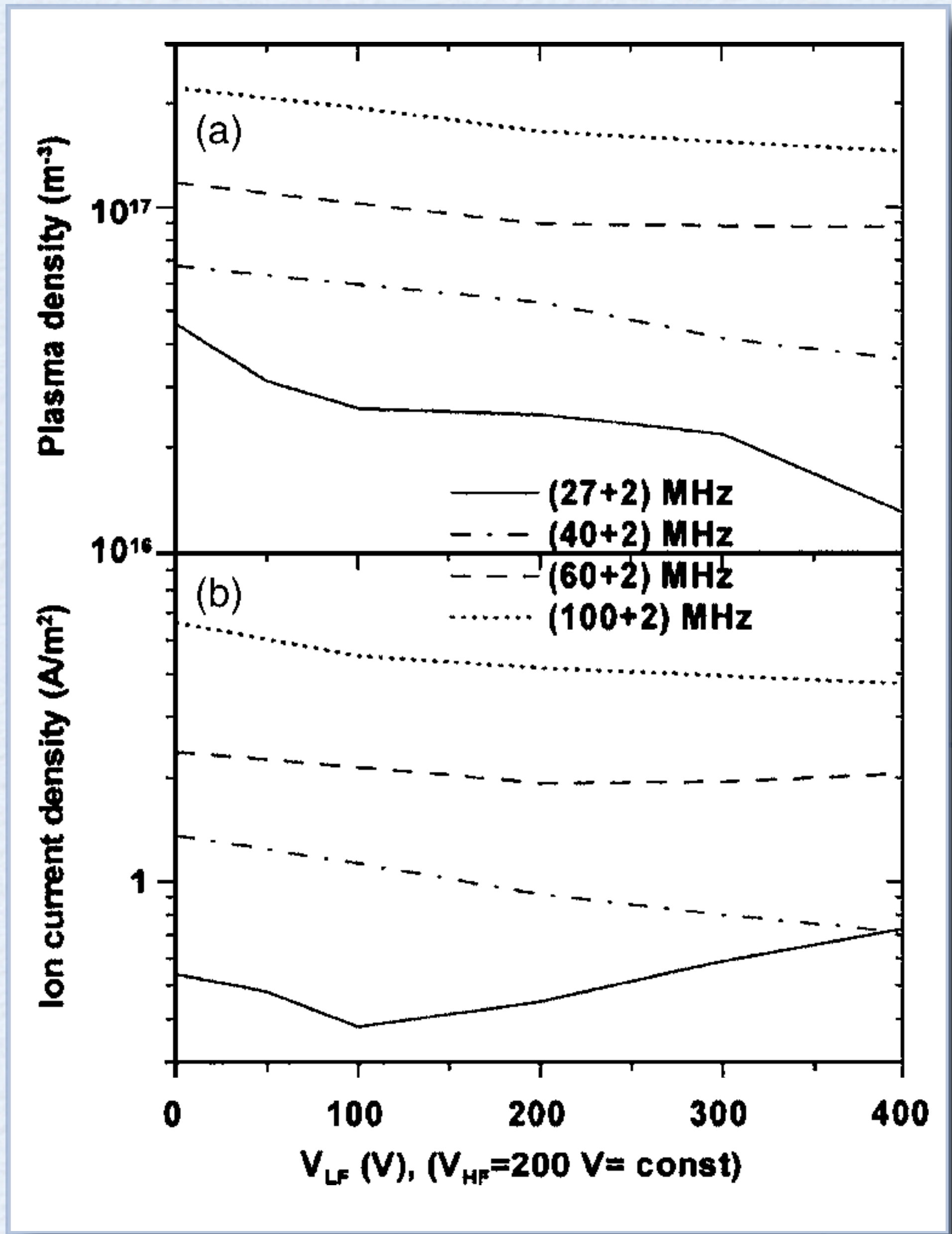


Figure 2. Plasma density, electron temperature, and ion current density onto electrodes as functions of the low frequency voltage amplitude. Conditions same as in figure 1.

100 MHz / 1MHz
p = 50 mTorr
L = 1.5 cm
ARGON

$\gamma = 0$

Control of ion properties in "classical" dual-frequency discharges



part of Fig. 2.

V. Georgieva and A. Bogaerts, J. Appl. Phys. 98, 023308 (2005)

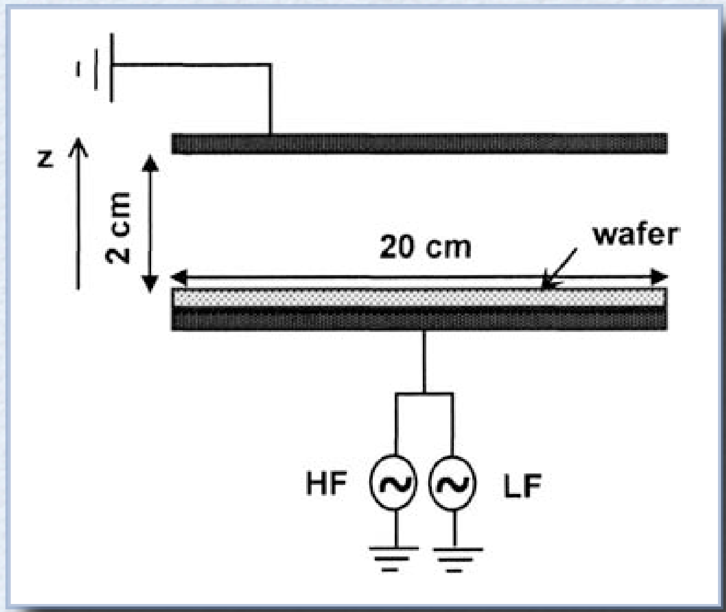
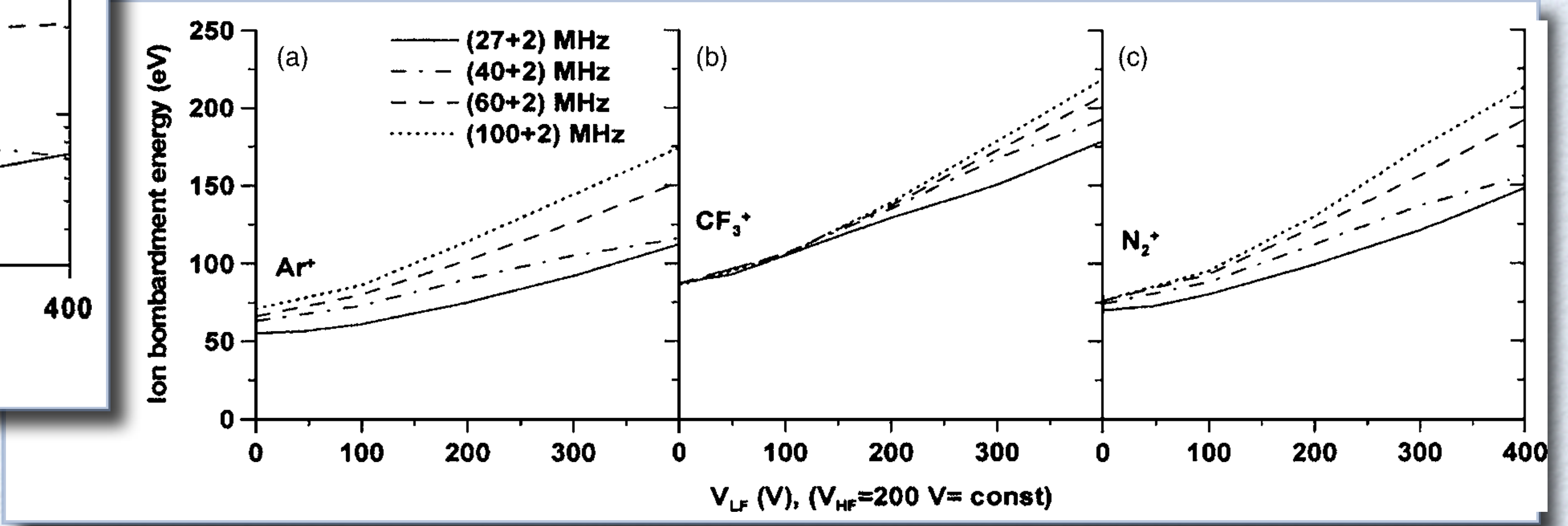
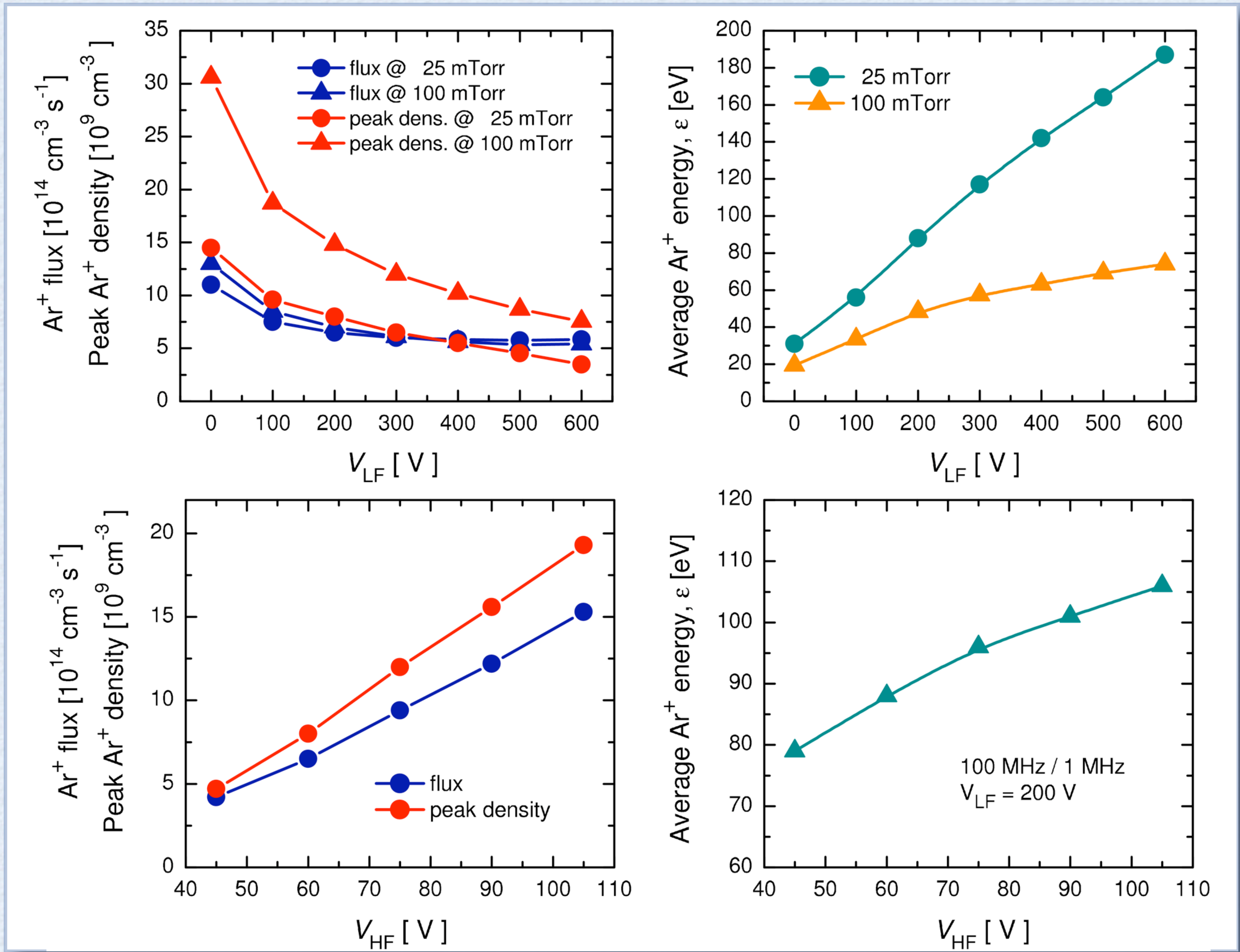


Fig. 3.



Ar / CF_4 / N_2 mixture at a ratio of 0.8 / 0.1 / 0.1, p = 30 mTorr.

Control of ion properties in "classical" dual-frequency discharges



The GOAL:

- Ion flux is controlled by V_{HF}
- Ion energy is controlled by V_{LF}

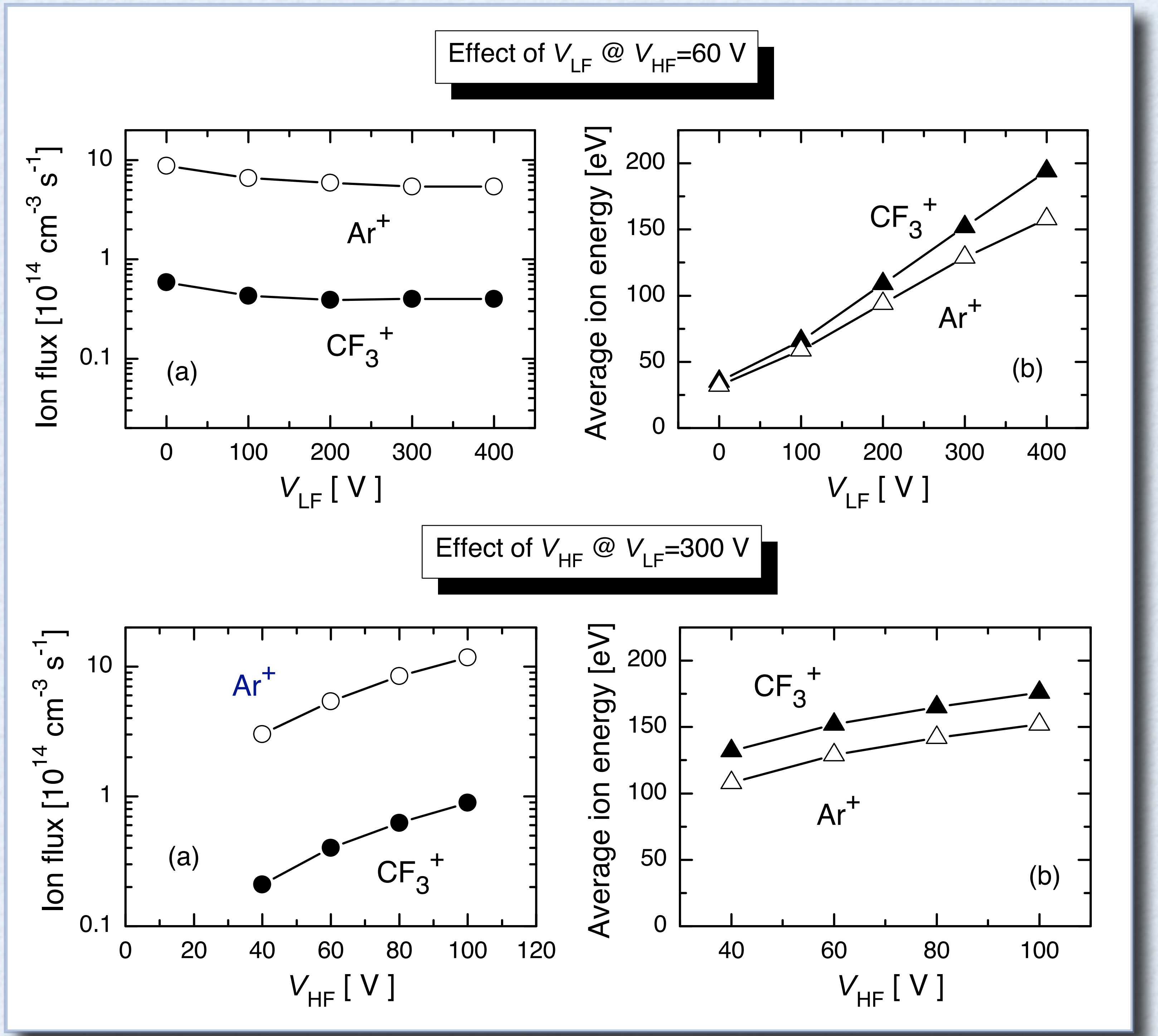
REALITY:

- Ion flux depends on some extent on V_{LF}
- Ion energy is somewhat influenced by V_{HF}

100 MHz & 1 MHz
γ = 0

Z. Donkó, Proc. Symp. Application of Plasma Processes, Podbanske, Slovakia, January 20–25, 2007, IL02, pp. 21–24

Control of ion properties in "classical" dual-frequency discharges

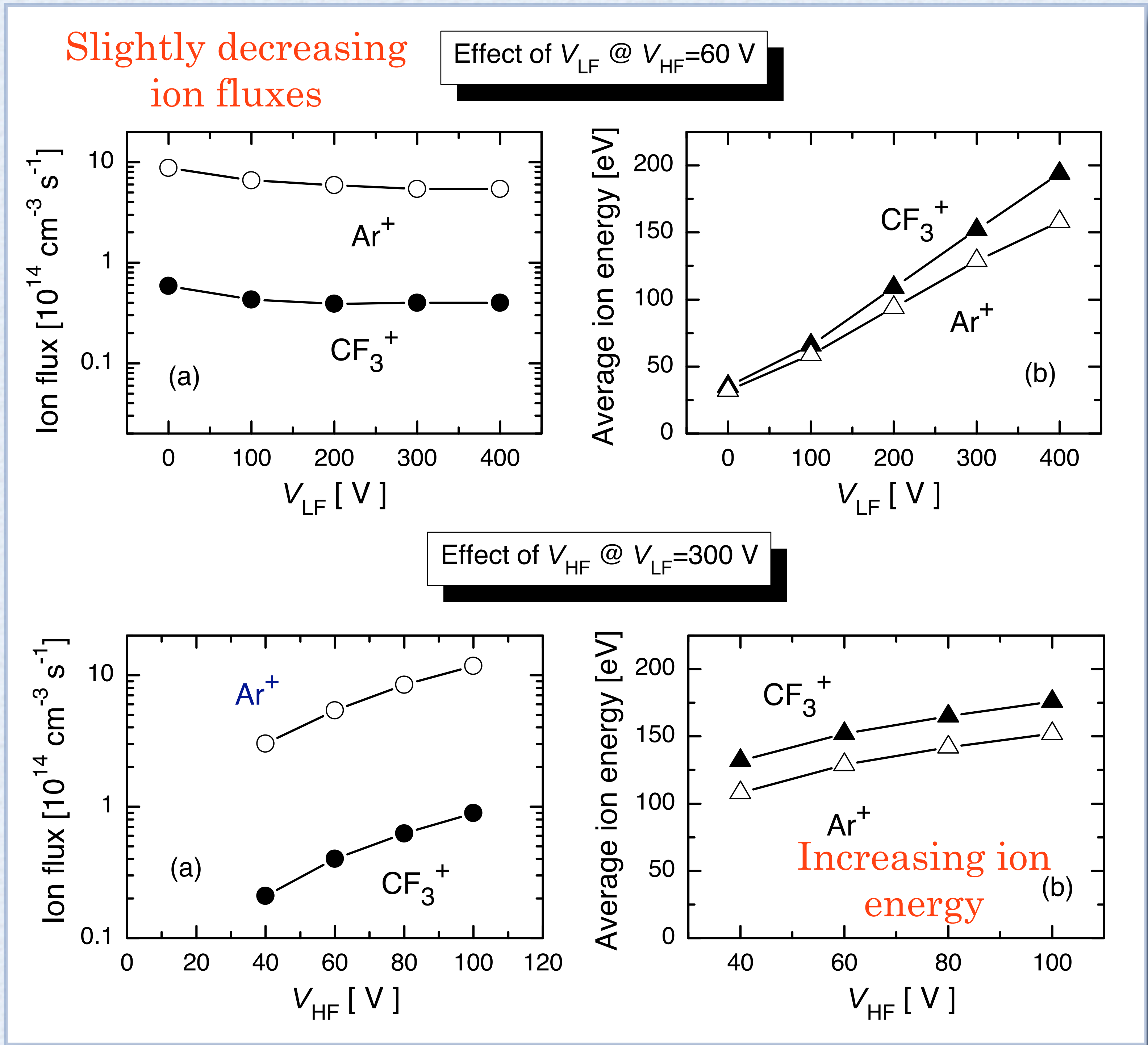


Z. Donkó and Z. Lj. Petrović,
J. Phys. Conf. Series 86, 012011 (2007)
[5th EU-Japan Joint Symp. on
Plasma Processing]

100 MHz / 1 MHz
10%CF_4 +90%Ar discharge
p = 20 mTorr

$\gamma = 0$

Control of ion properties in "classical" dual-frequency discharges



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Control of ion properties in "classical" dual-frequency discharges

J. P. Booth, G. Curley, D. Marić and P. Chabert, Plasma Sources Sci. Technol. 19, 015005 (2010)

Experiment:
Ar/O_2 mixture, oxidized Si electrodes, **γ up to 0.5**

Plasma density and ion current increases with increasing low-frequency power

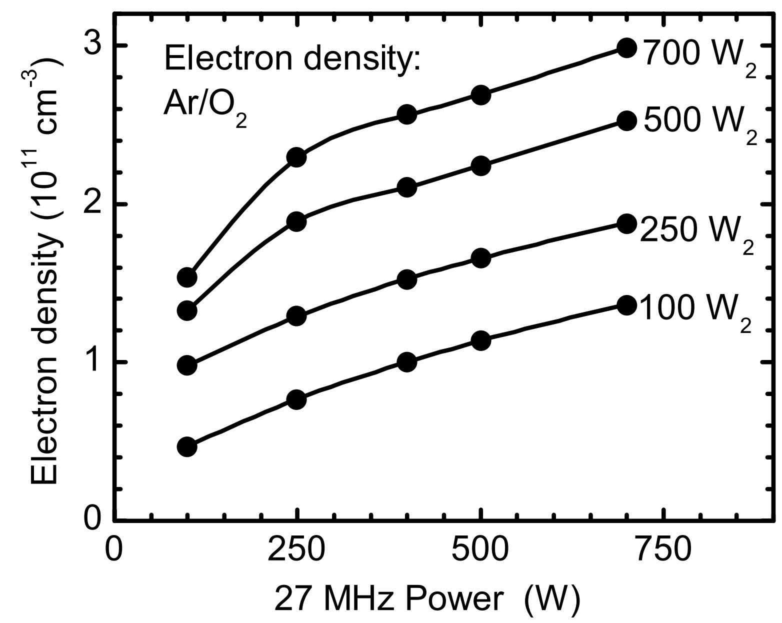

Figure 1. Electron density in an Ar/O_2 plasma as a function of 2 and 27 MHz power.

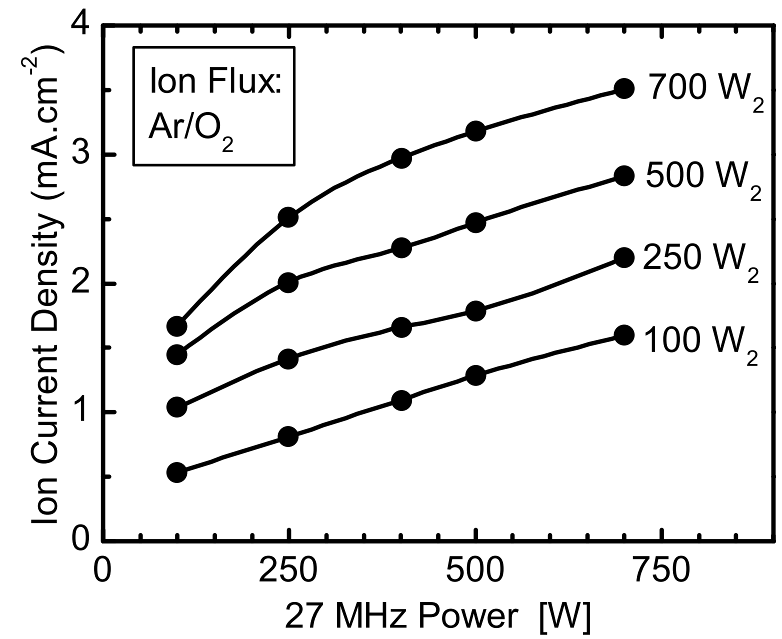

Figure 2. Ion flux in an Ar/O_2 plasma as a function of 2 and 27 MHz power.

Control of ion properties in "classical" dual-frequency discharges

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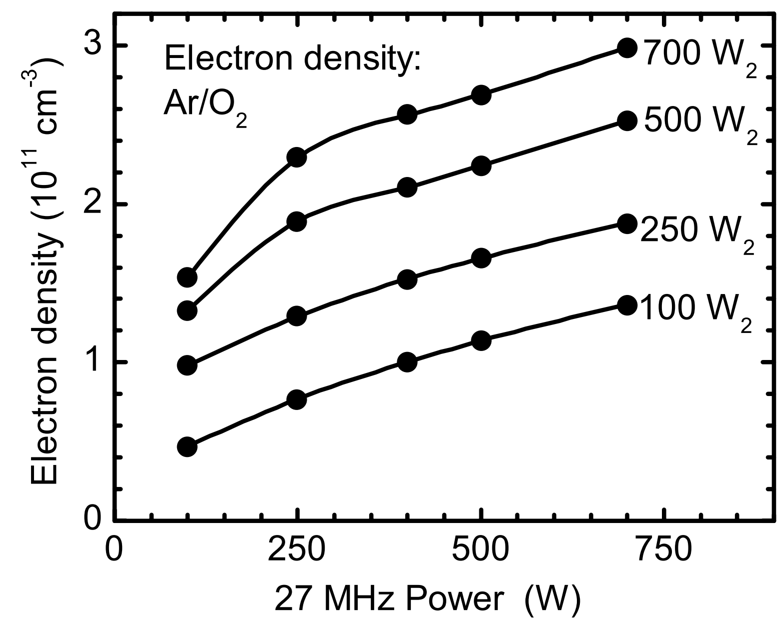

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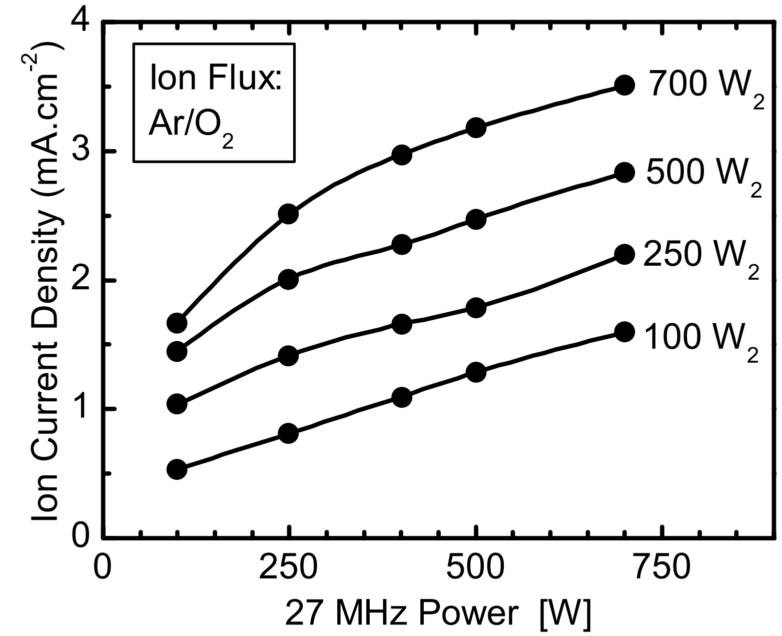
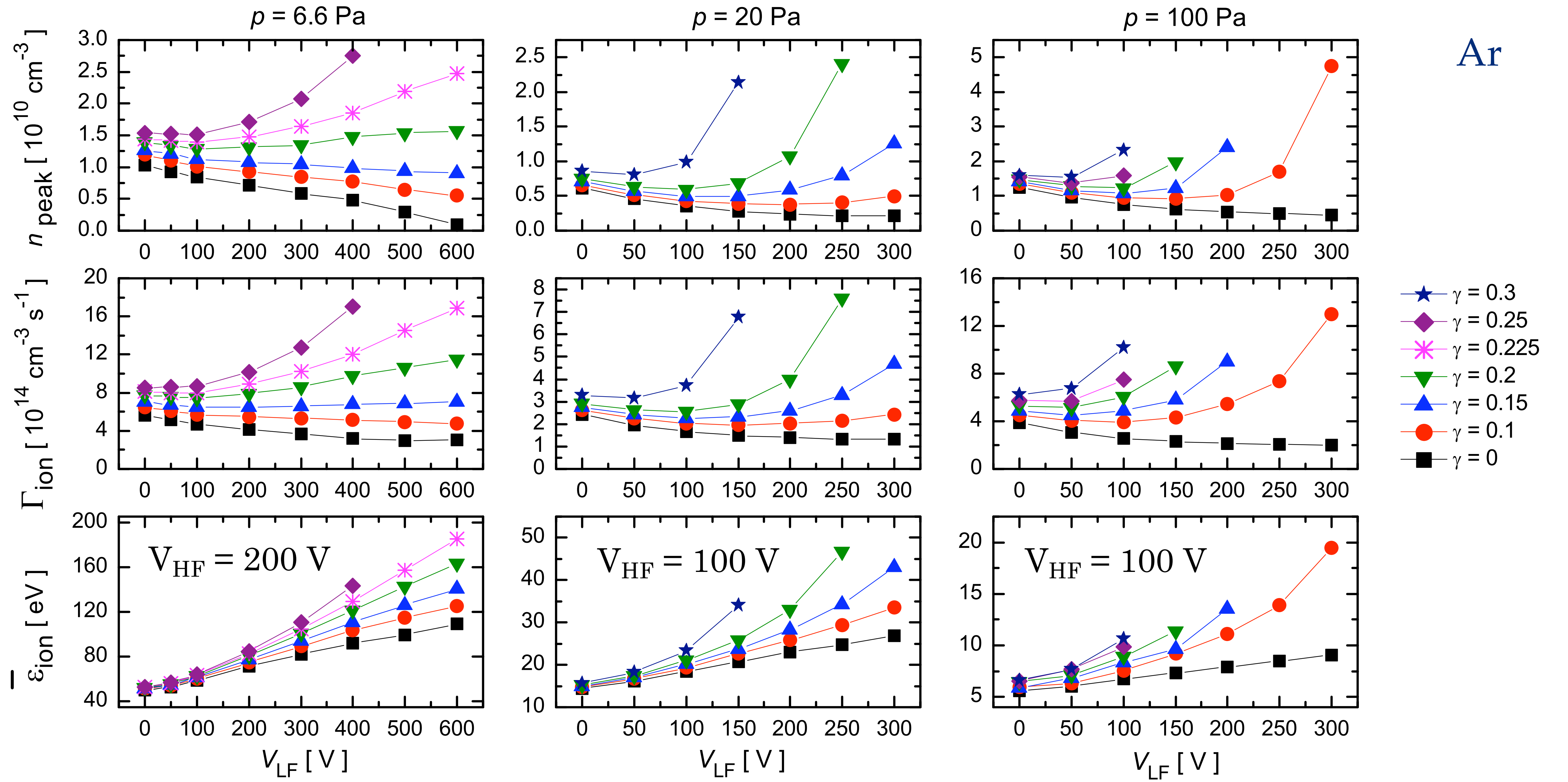


Figure 2. Ion flux in an Ar/O_2 plasma as a function of 2 and 27 MHz power.

A systematic study, covering a wide range of discharge conditions and secondary yield would be desirable...

Ion properties in classical DF discharges (27 + 2 MHz): results for the effect of secondary electrons



Z. Donkó, J. Schulze, P. Hartmann, I. Korolov, U. Czarnetzki, and E. Schüngel, Appl. Phys. Lett. 97, 081501 (2010).

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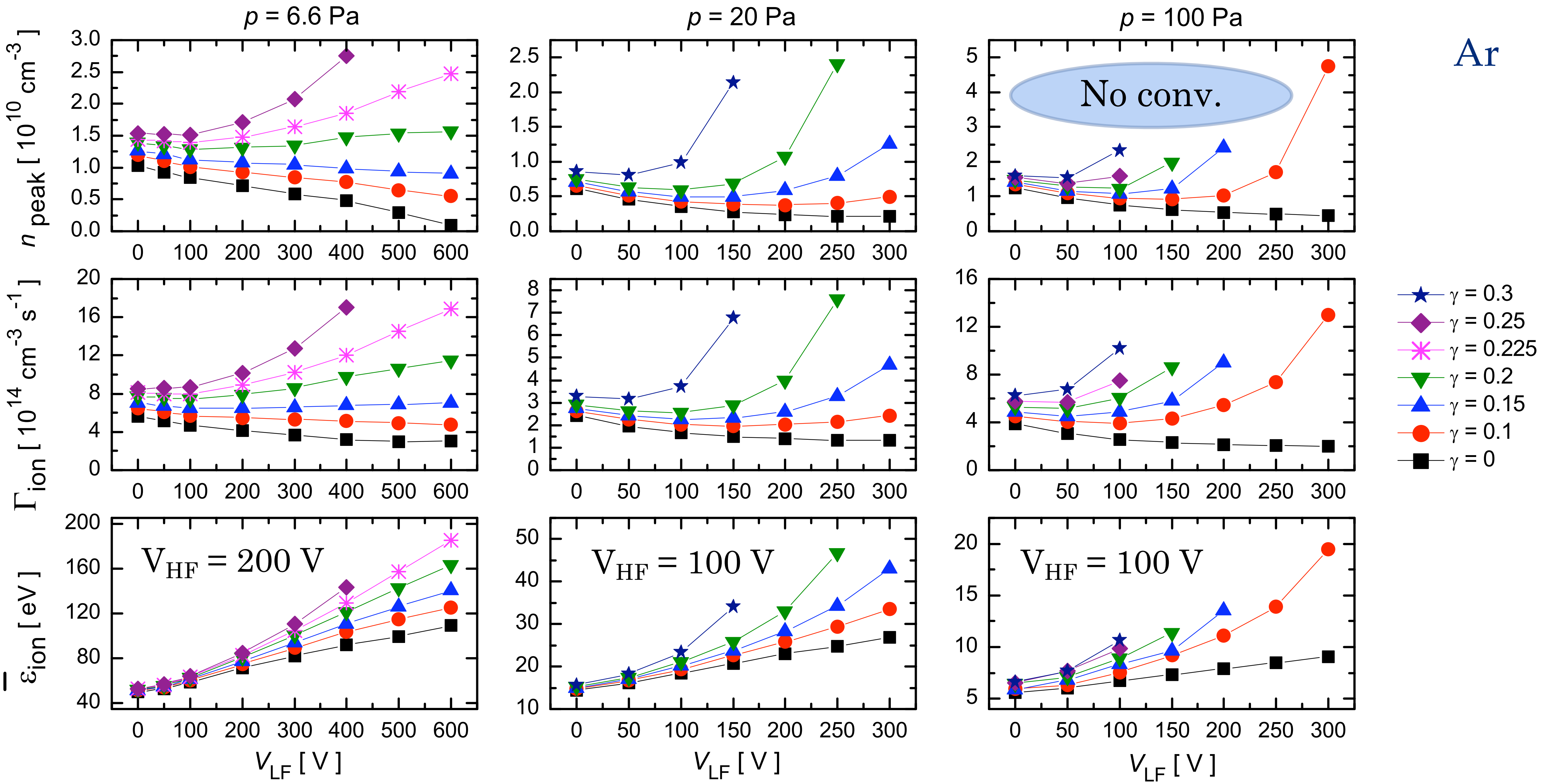

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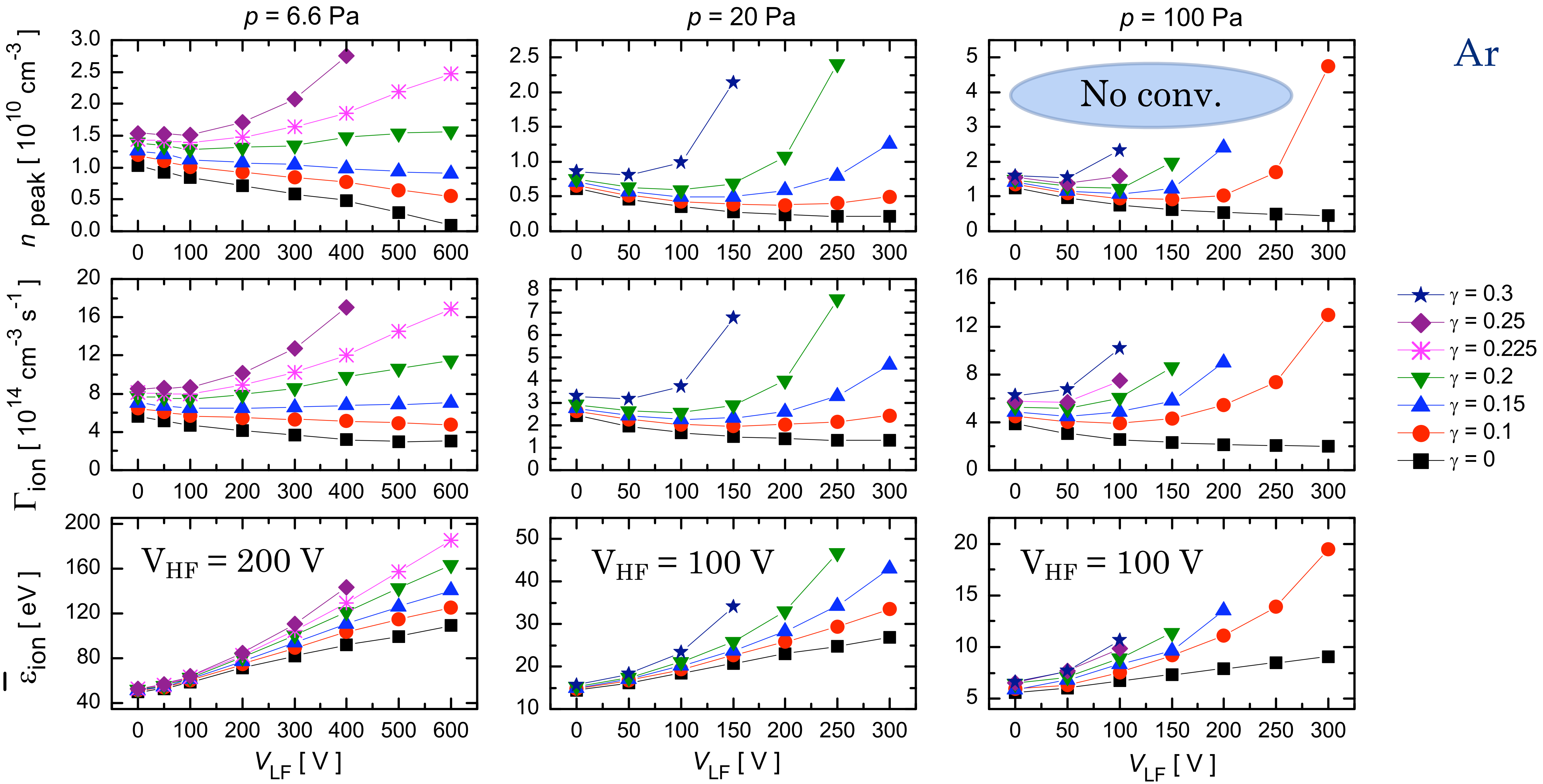
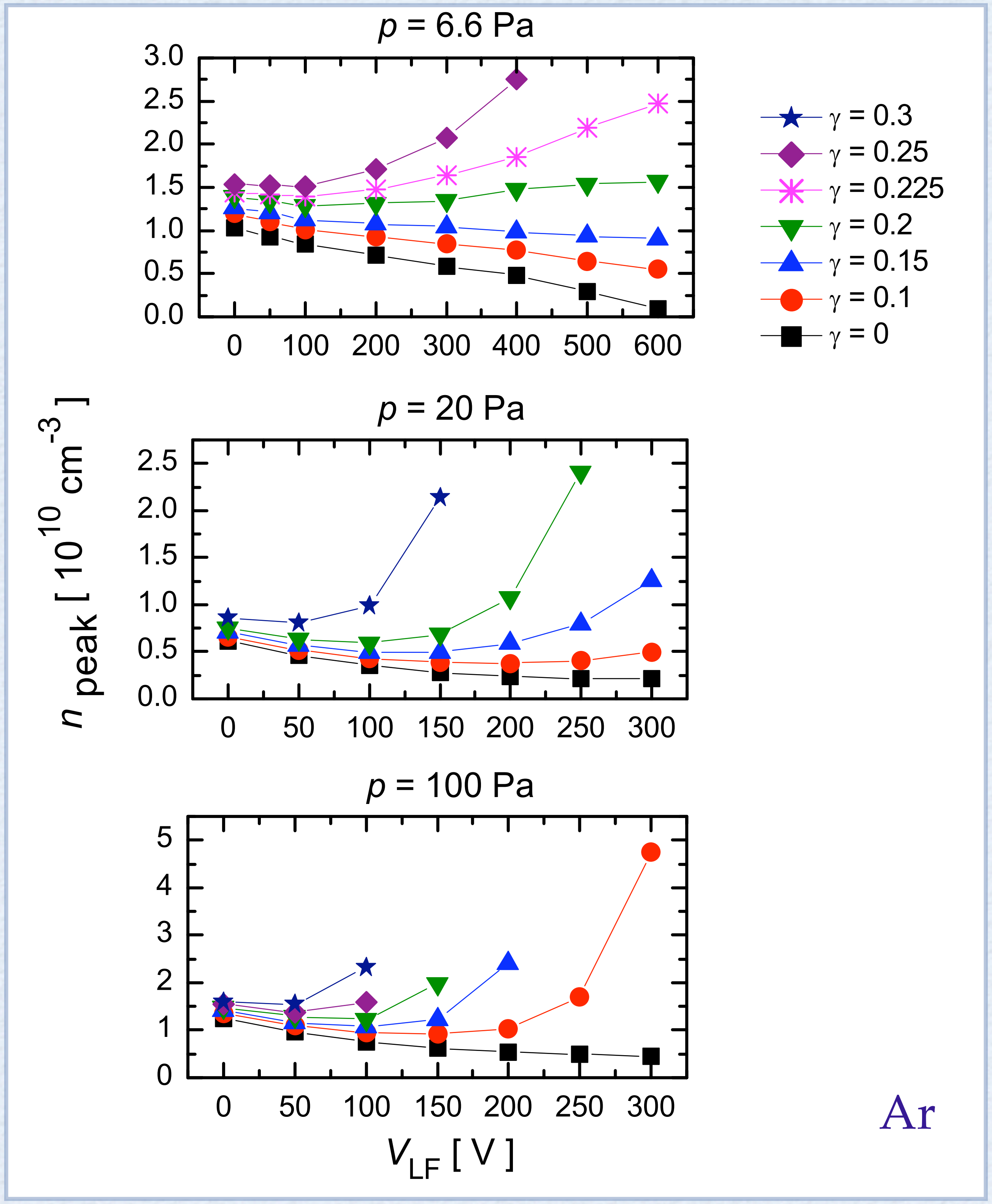


Ion energy increases with low-frequency voltage: OK ✓
Peak ion density and ion flux behave in a very similar way } → analyze the ion density

Z. Donkó, J. Schulze, P. Hartmann, I. Korolov, U. Czarnetzki, and E. Schüngel, Appl. Phys. Lett. 97, 081501 (2010).

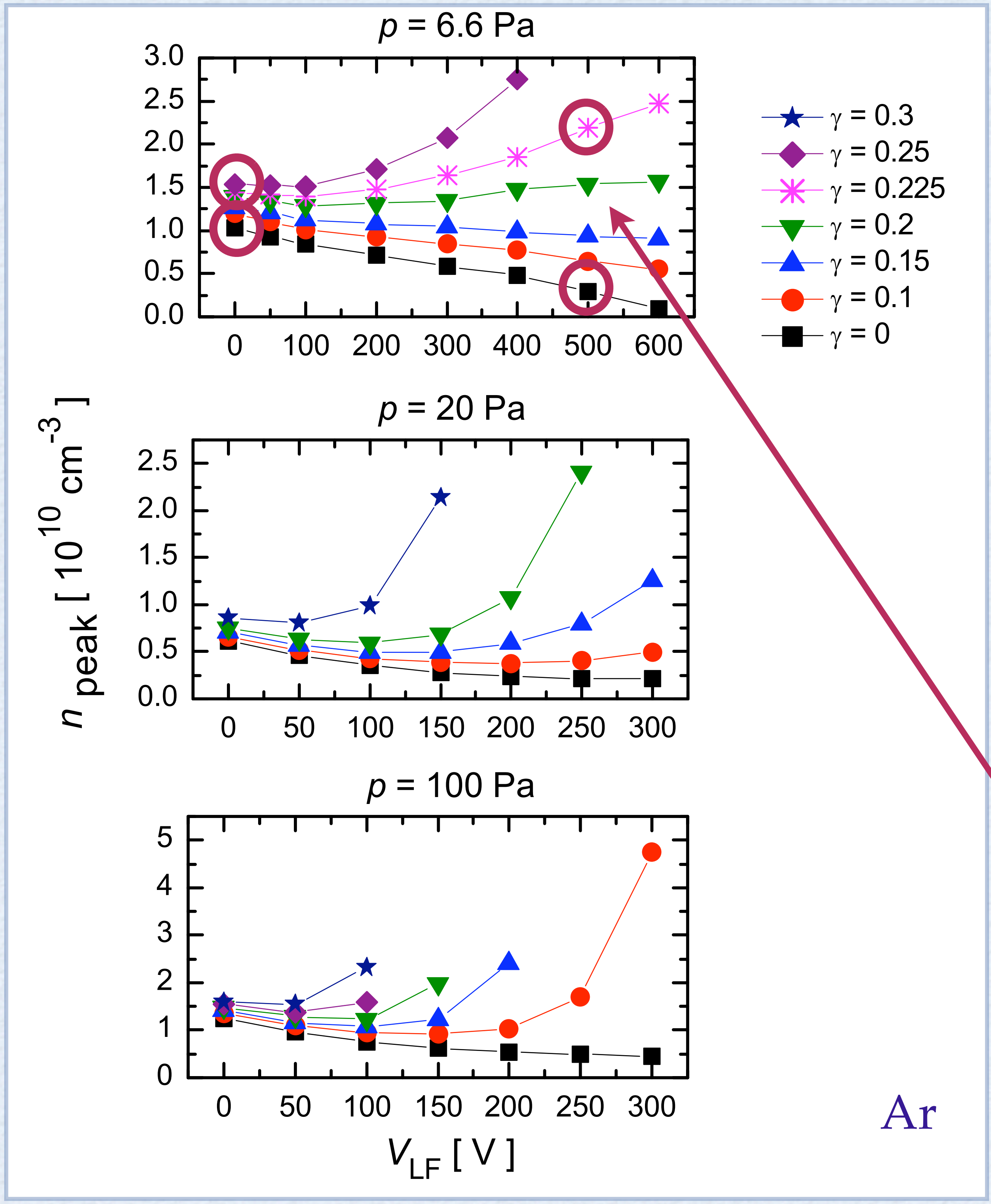
Ion properties in classical DF discharges (27 & 2 MHz)



- At low γ the ion density decreases with increasing LF voltage
- At intermediate γ a nearly constant ion density can be realized, with increasing pressure (when ionization by secondaries start to dominate) this becomes more difficult due to the rapid increase of the ion density with increasing LF voltage
- At high γ the ion density increases with increasing LF voltage at all pressures

Separate control of ion flux and energy is generally not possible, only for certain parameter domain

Ion properties in classical DF discharges (27 & 2 MHz)



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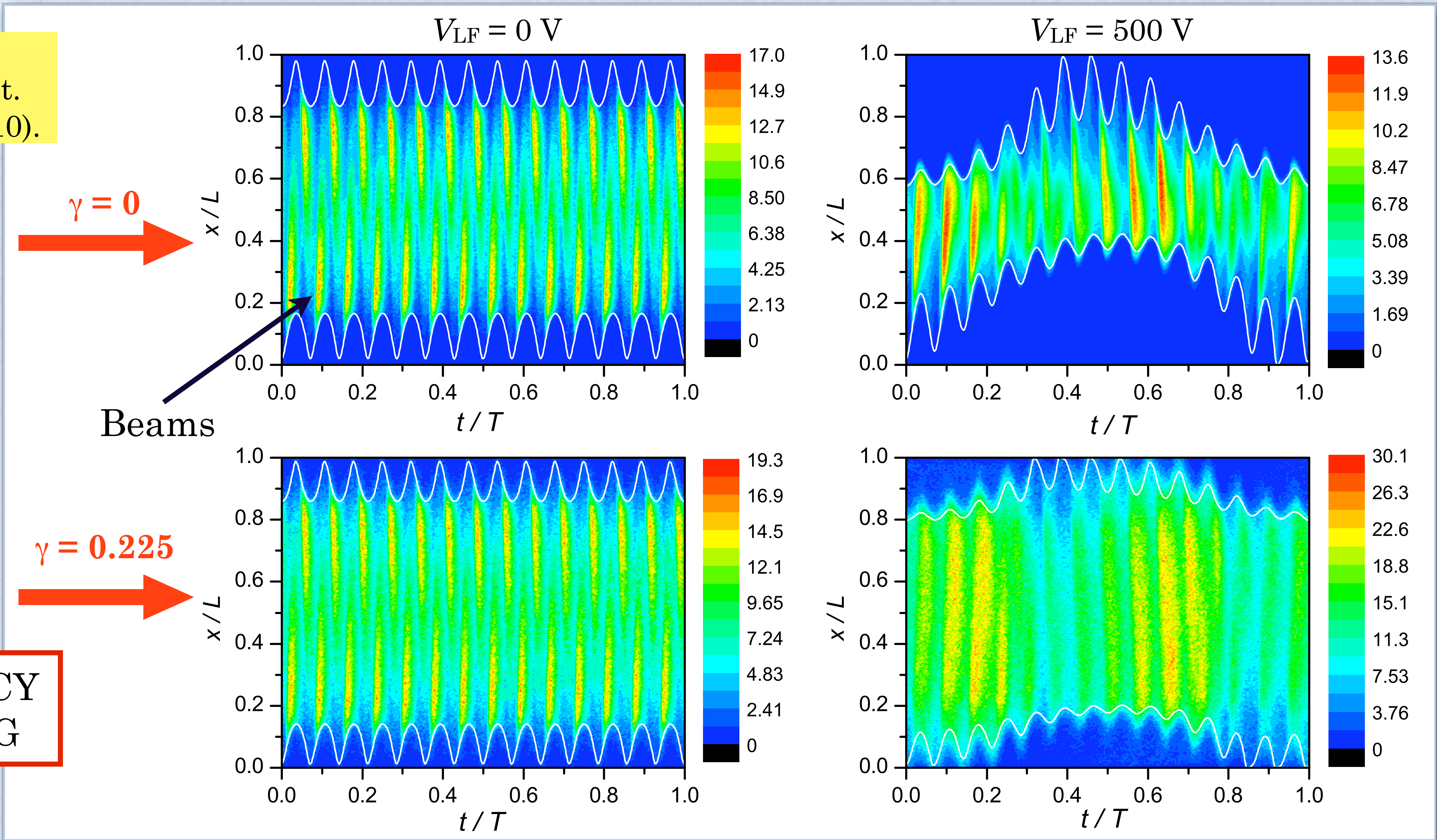
Why does this happen?

Classical DF discharges (27 & 2 MHz): frequency coupling & effect of secondary electrons

Z. Donkó, et al, Appl. Phys. Lett. 97, 081501 (2010).

Argon
$V_{HF} = 200$ V
$p = 6.6$ Pa
$L = 2.5$ cm
$T = 400$ K



FREQUENCY COUPLING

See also:

Turner M M and Chabert P 2006 Phys. Rev. Lett. **96** 205001
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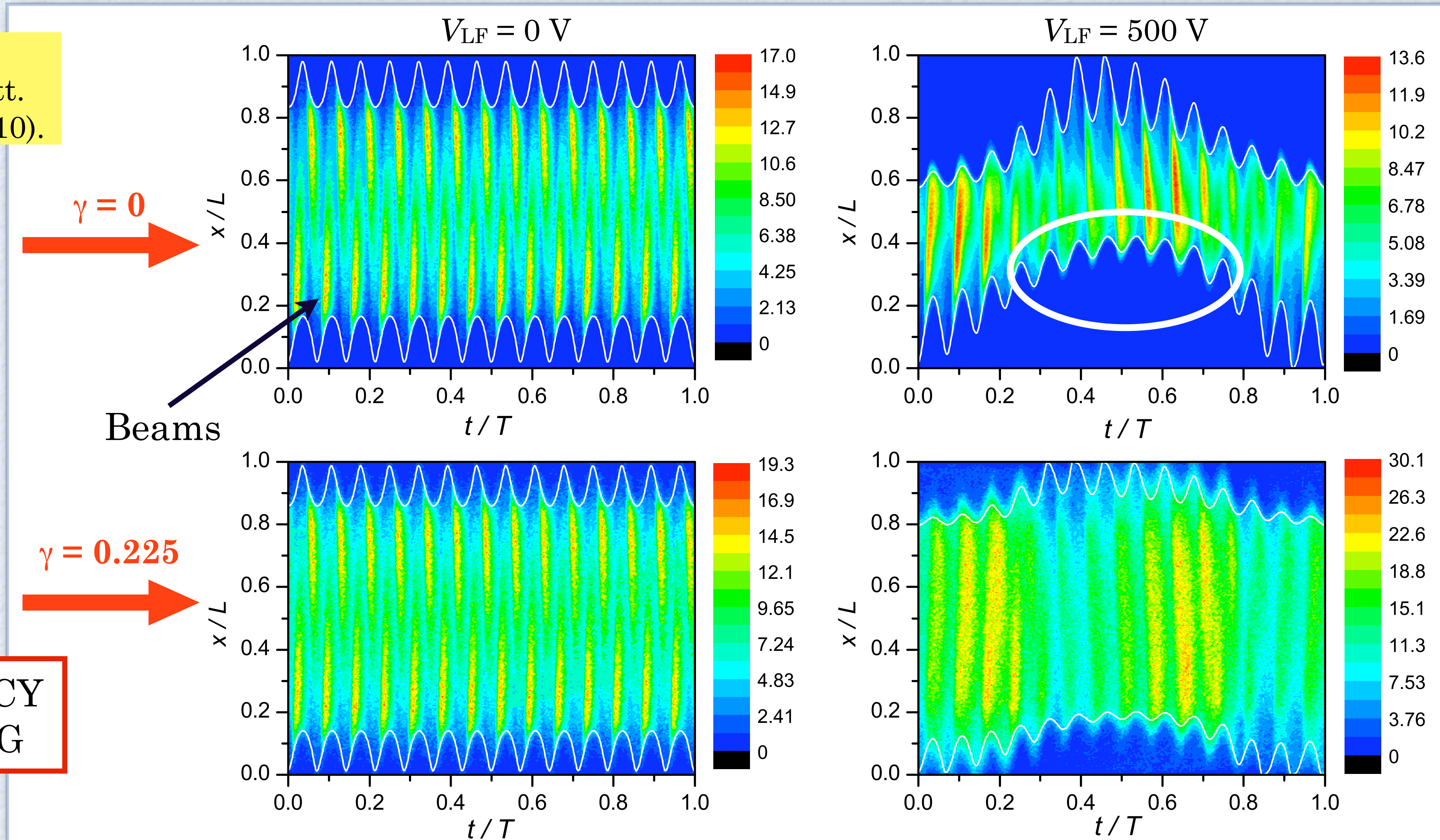
Classical DF discharges (27 & 2 MHz): frequency coupling & effect of secondary electrons

Z. Donkó, et al, Appl. Phys. Lett. 97, 081501 (2010).

Argon
V_{HF} = 200 V
p = 6.6 Pa
L = 2.5 cm
T = 400 K



FREQUENCY COUPLING

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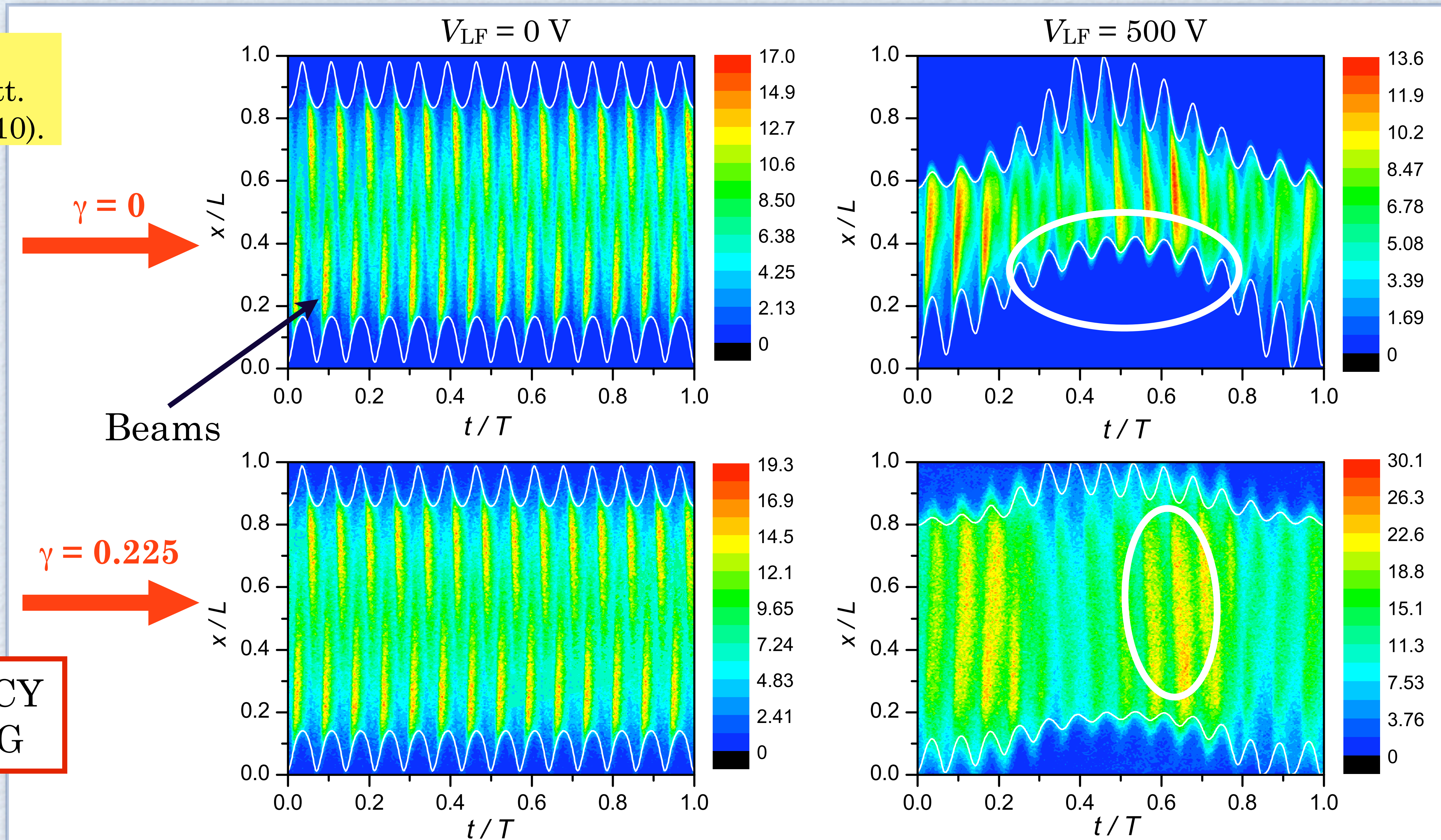

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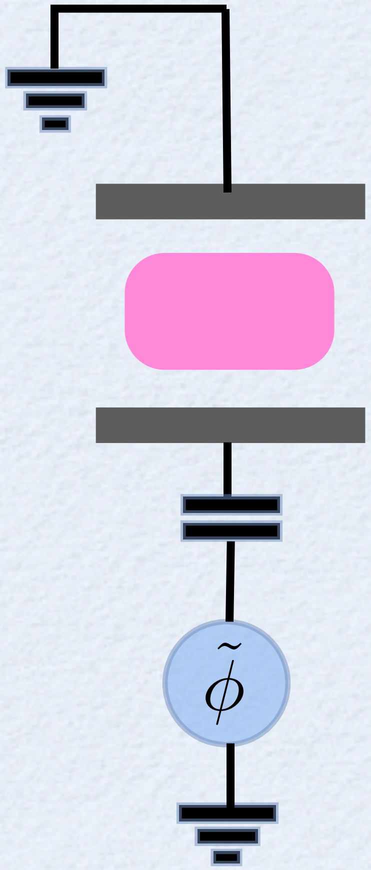
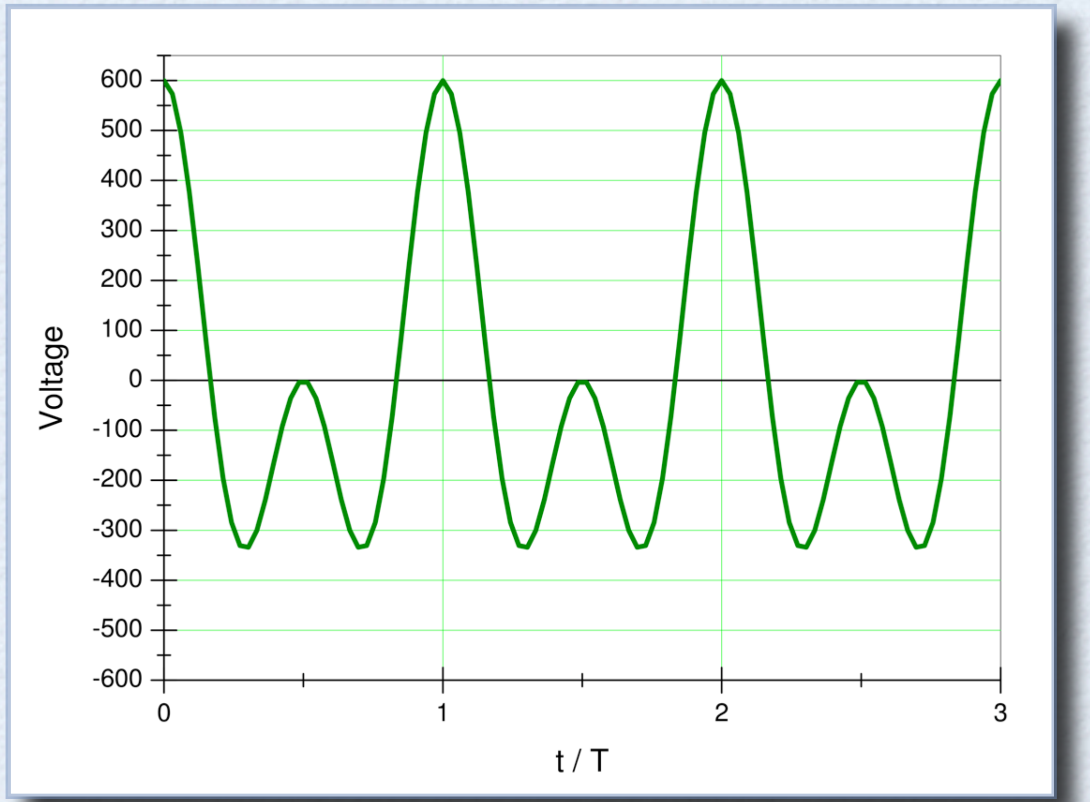
The Electrical Asymmetry Effect

and the influence of secondary electrons on the control of ion properties

The Electrical Asymmetry Effect





$$\tilde{\phi} = V_0[\cos(\omega t + \theta) + \cos(2\omega t)]$$

↑ control parameter

Heil B G, Schulze J, Mussenbrock T, Brinkmann R P and Czarnetzki U 2008 IEEE Trans. on Plasma Sci. 36 1404
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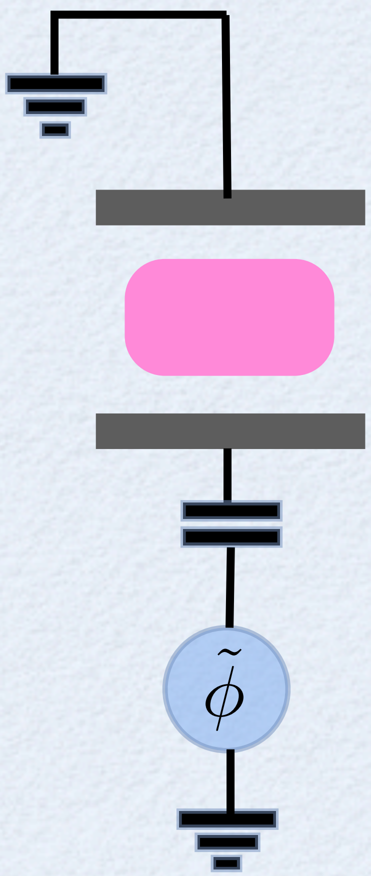
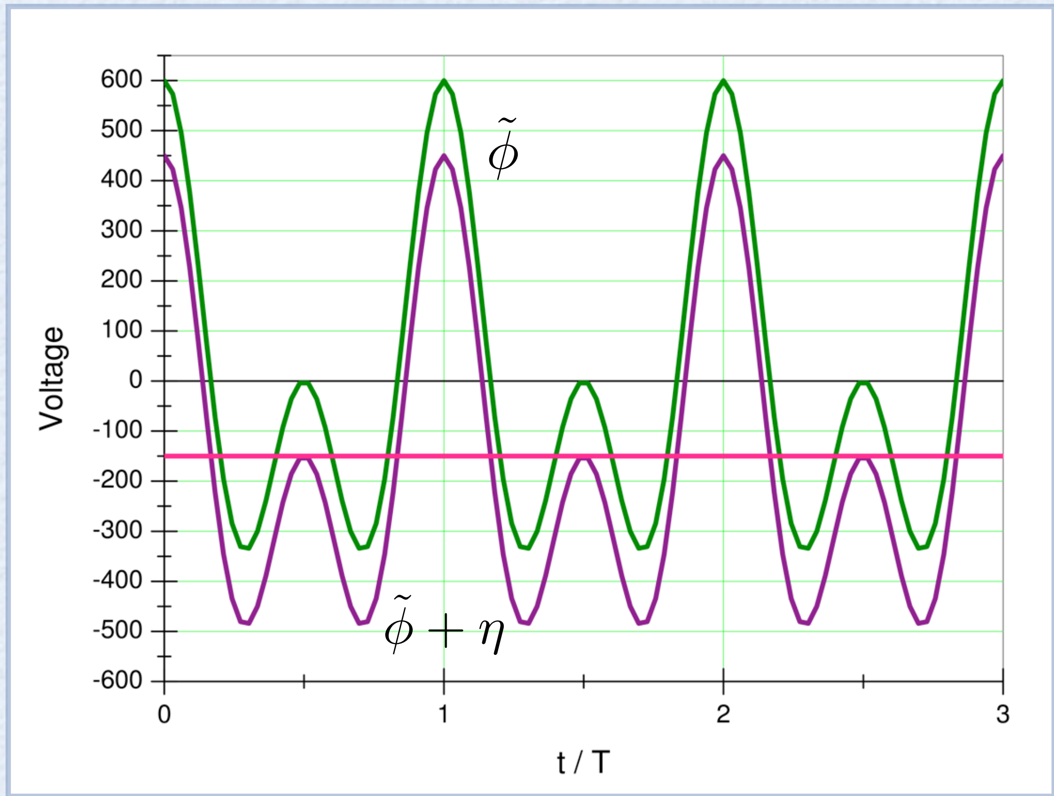
The Electrical Asymmetry Effect





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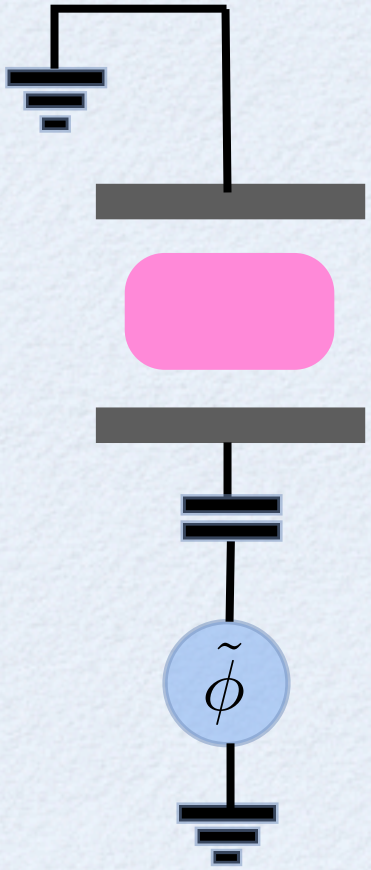

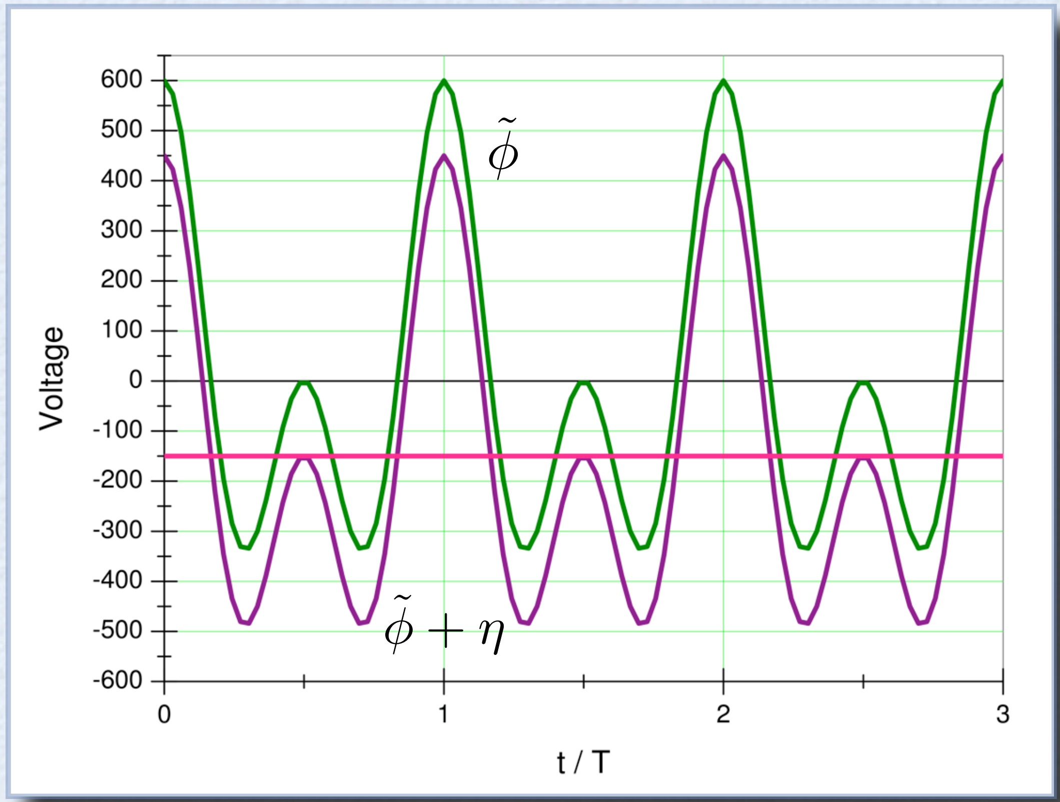

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In geometrically symmetrical reactors a DC self bias builds up, if the discharge is driven by a waveform which contains an even harmonic of the fundamental frequency.

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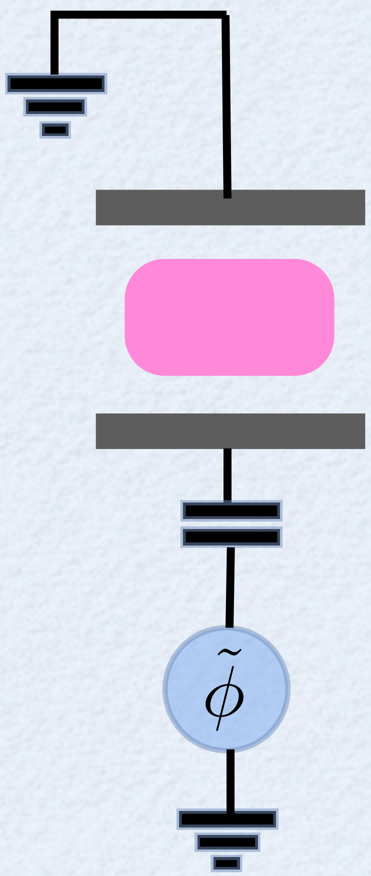

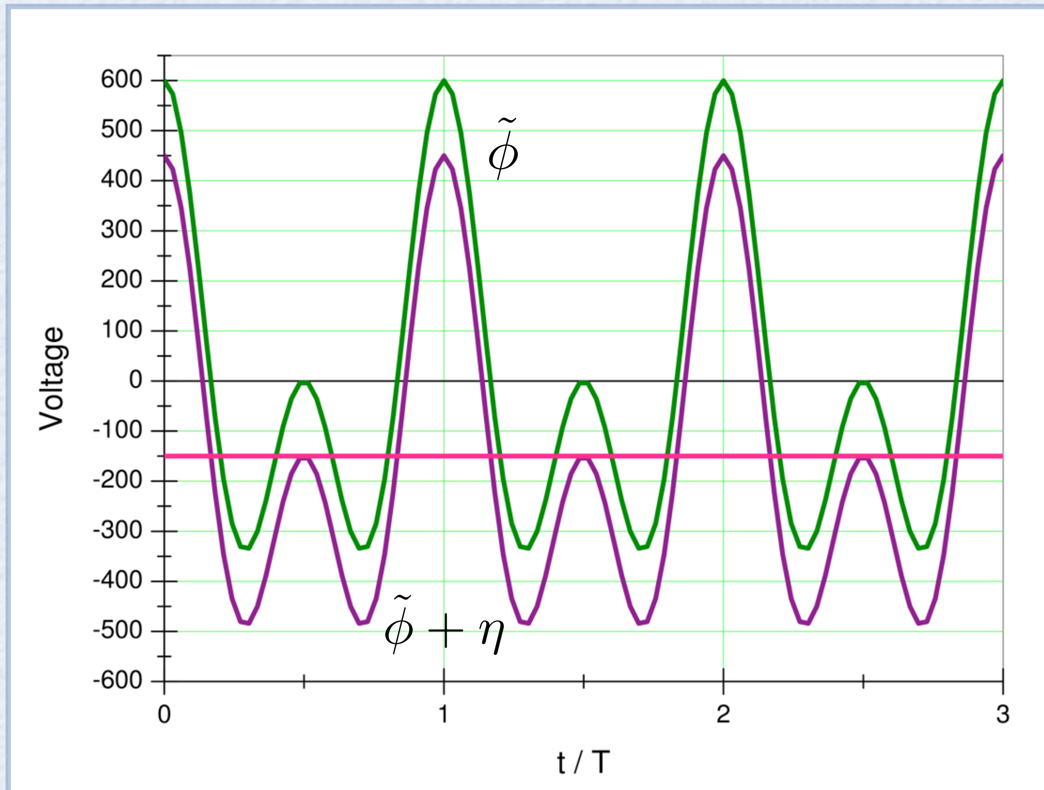

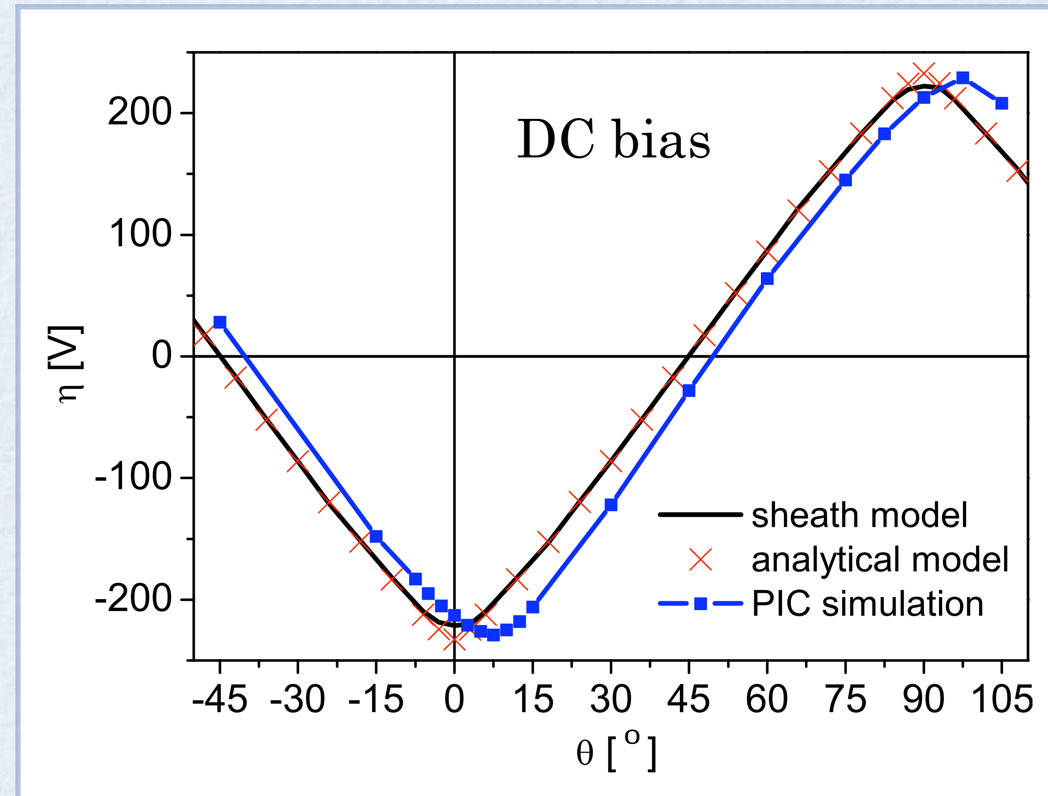

$$\tilde{\phi} = V_0[\cos(\omega t + \theta) + \cos(2\omega t)]$$

↑ control parameter

f = 13.56 MHz & 27.12 MHz,
p = 20 mTorr, d = 6.7 cm
V_0 = 315 V (both freq. !!)

In geometrically symmetrical reactors a DC self bias builds up, if the discharge is driven by a waveform which contains an even harmonic of the fundamental frequency.

Z. Donkó, J. Schulze, B. G. Heil and U. Czarnetzki,
J. Phys. D 42, 025205 (2009)

Heil B G, Schulze J, Mussenbrock T, Brinkmann R P and Czarnetzki U 2008 IEEE Trans. on Plasma Sci. 36 1404
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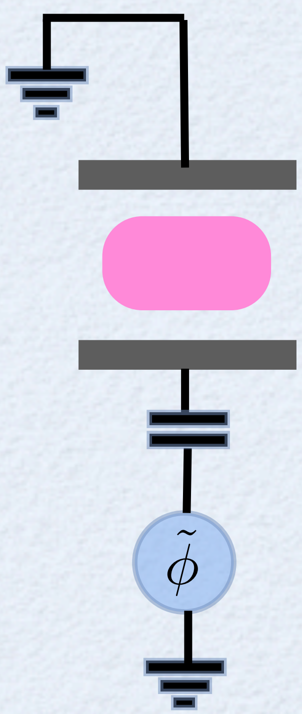
The Electrical Asymmetry Effect

- Ar discharge
- $p = 5$ Pa
- $L = 2.5$ cm
- $f = 13.56$ MHz
- $V_0 = 200$ V



PIC results

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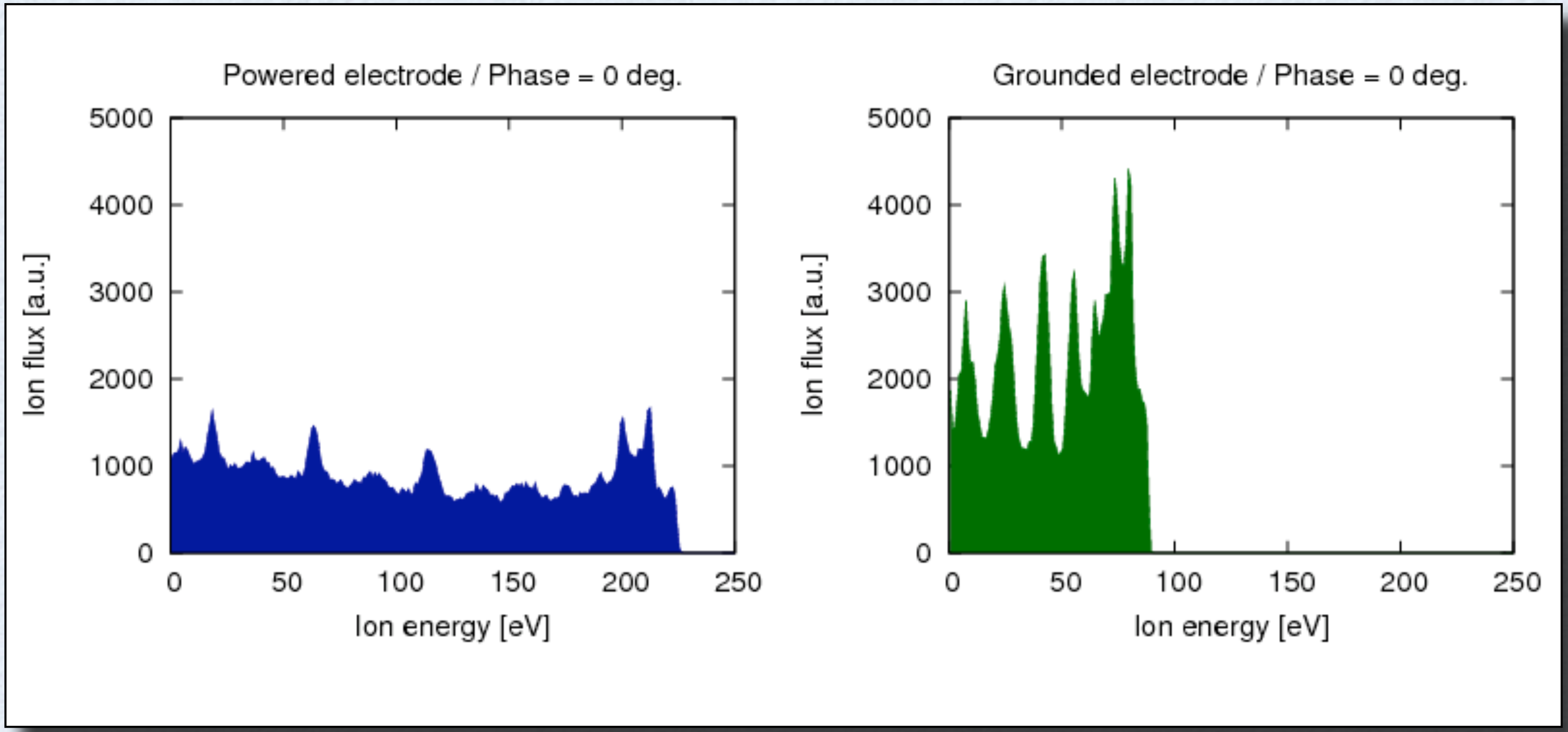

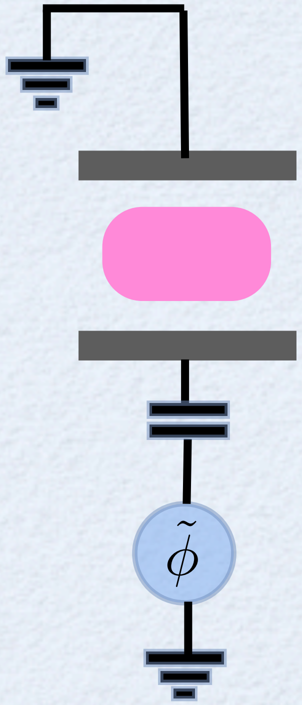

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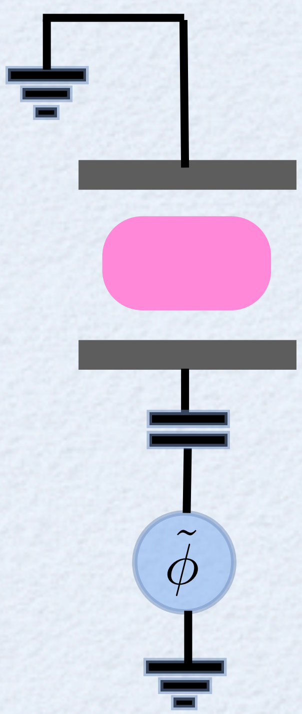

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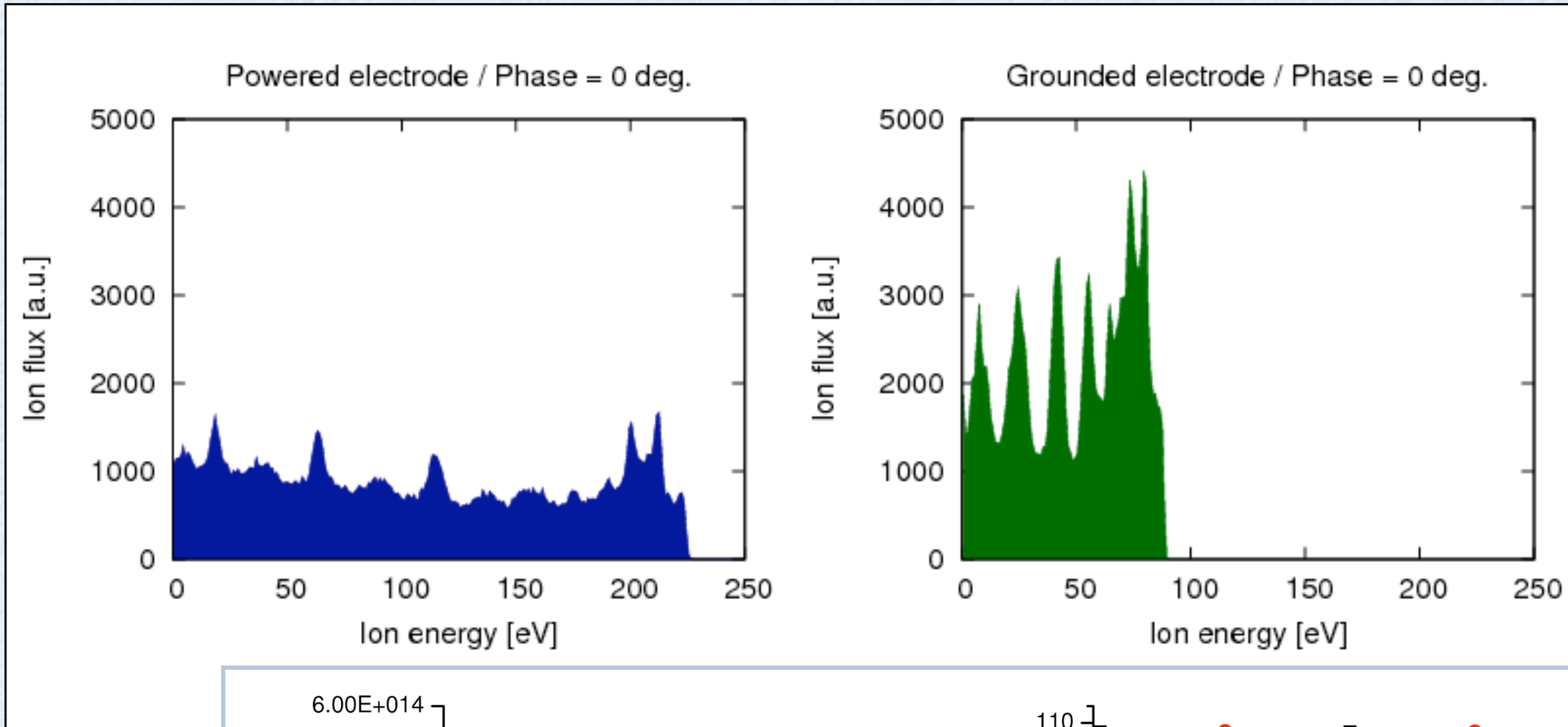

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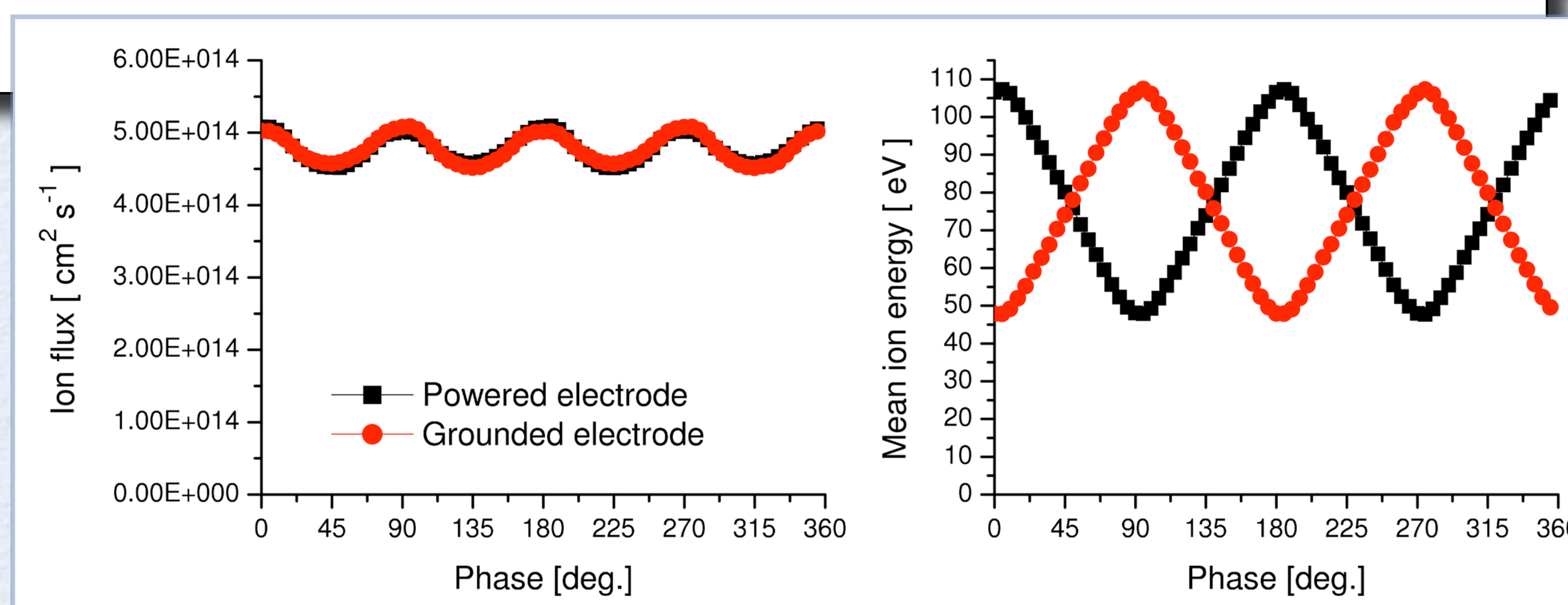
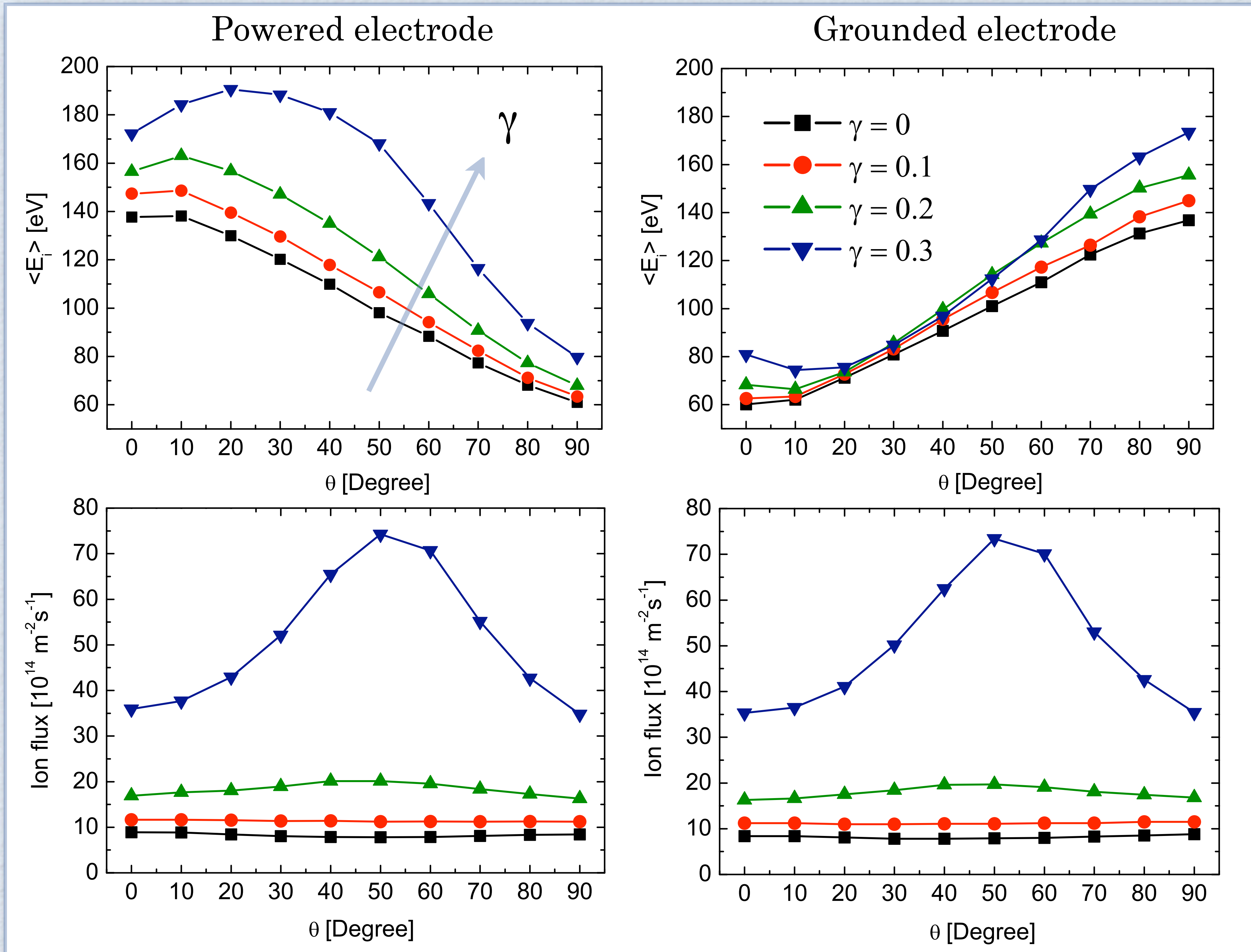


Mean ion energy and ion flux : effect of γ on EAE operation



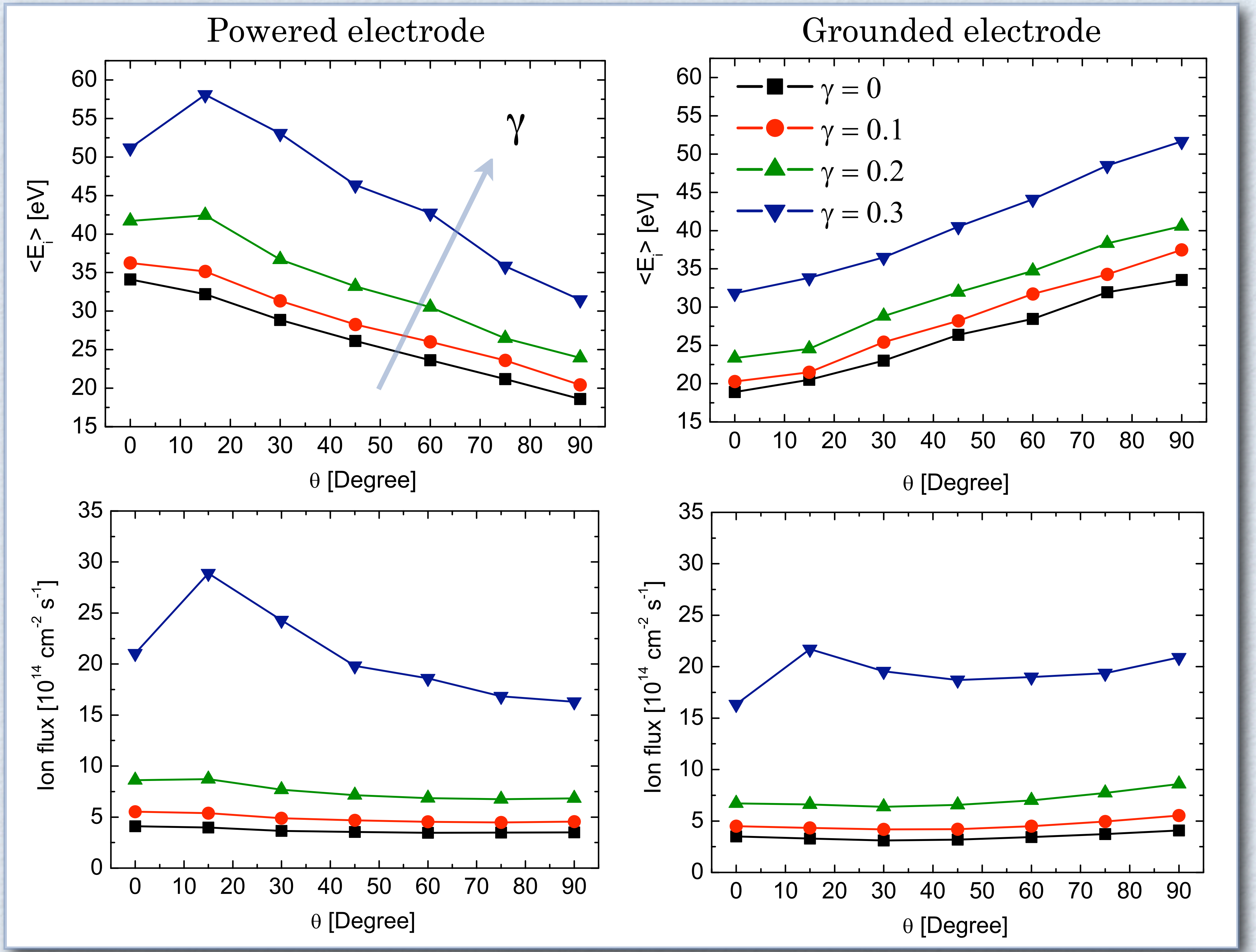
6.6 Pa
$V_{HF} = V_{HF} = 300$ V

Mean ion energy range: 2.4 (at low γ), 3.2 (at high γ); varies in an opposite manner at the two electrodes

The peak at the powered electrode at high γ results from the charge dynamics

The ion flux is approximately constant at at low γ, the peak at high γ is the result of the sheath/ionization dynamics (discussed later)

Mean ion energy and ion flux : effect of γ on EAE operation



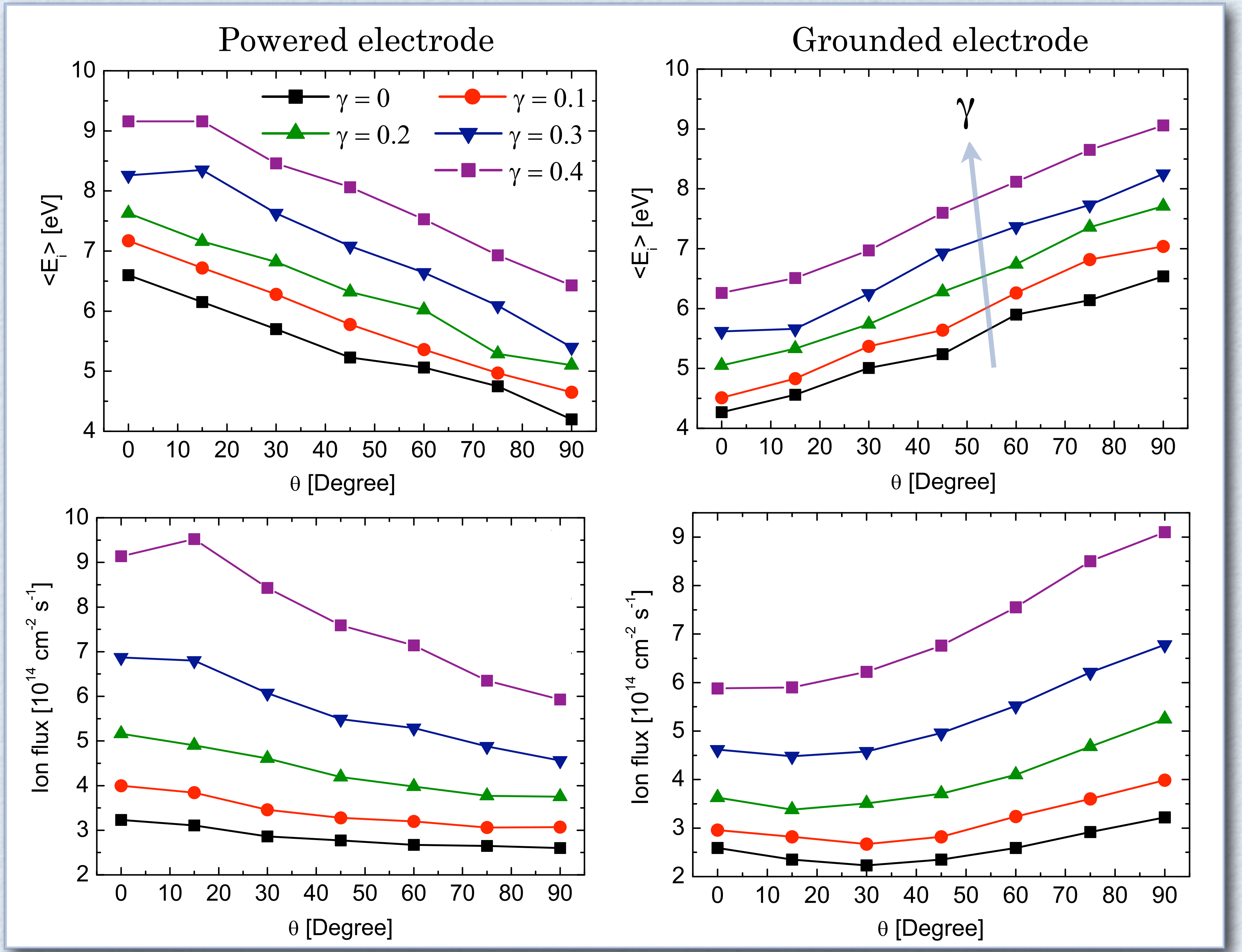
20 Pa
$V_{HF} = V_{HF} = 150$ V

Mean ion energy range: 1.6-1.7

The peak at the powered electrode at high γ results from the charge dynamics

The ion flux is approximately constant at at low γ, the peak at high γ is the result of the sheath/ionization dynamics

Mean ion energy and ion flux : effect of γ on EAE operation

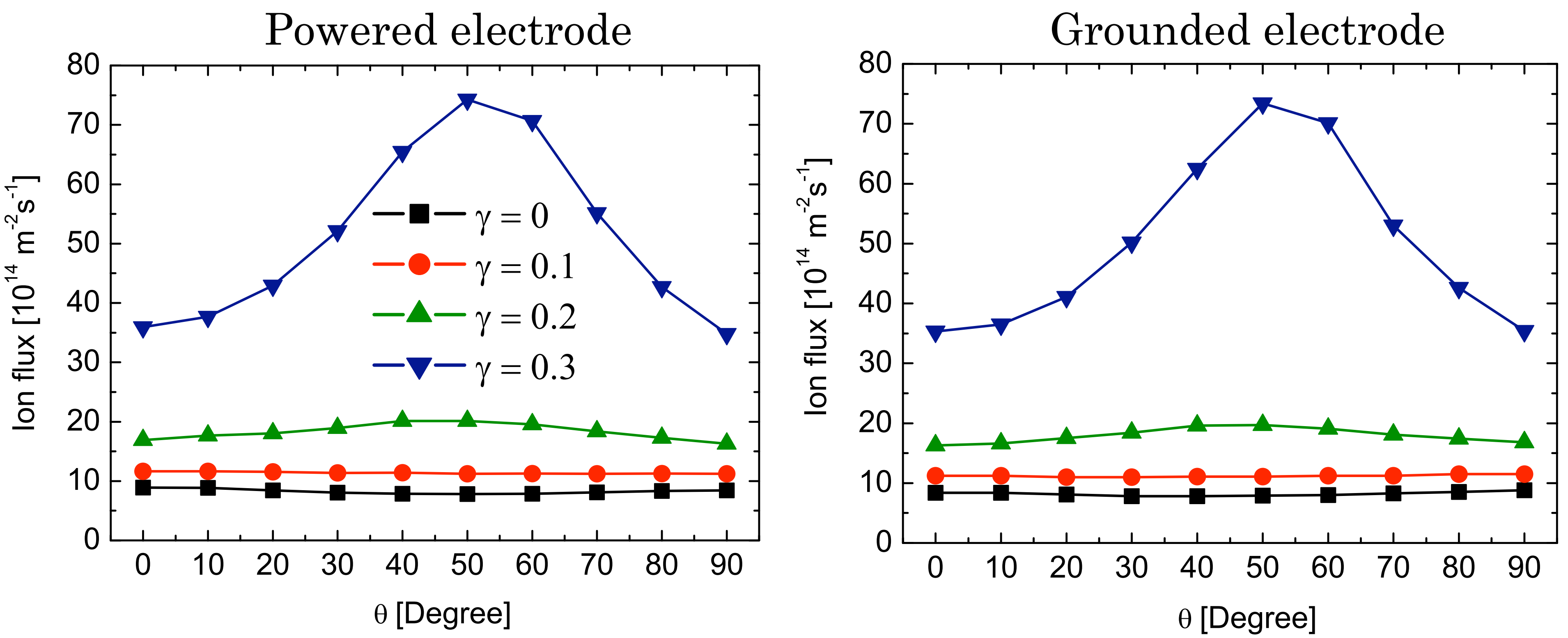


100 Pa
$V_{HF} = V_{HF} = 75$ V

Mean ion energy range ≈1.6

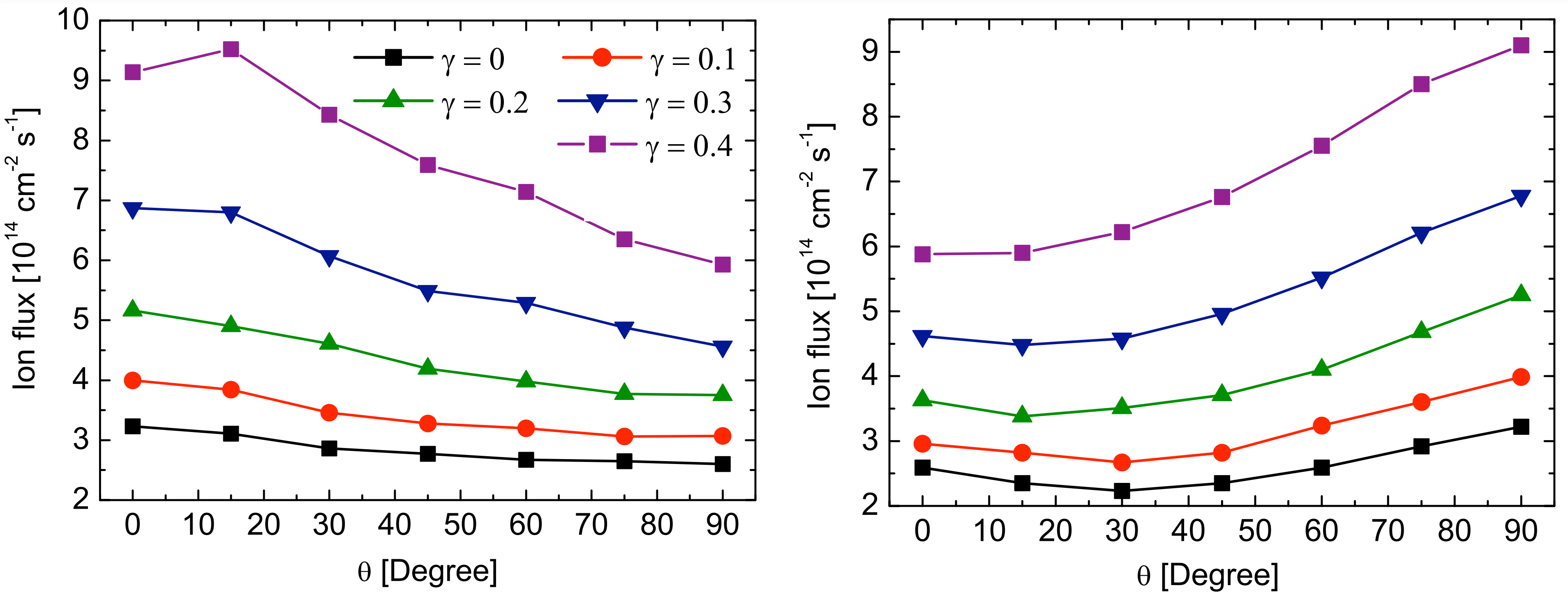
The ion flux at the powered electrode decreases as a function of θ. The opposite is observed at the grounded electrode.

Ion flux vs. phase angle : low / high pressure at high γ



6.6 Pa

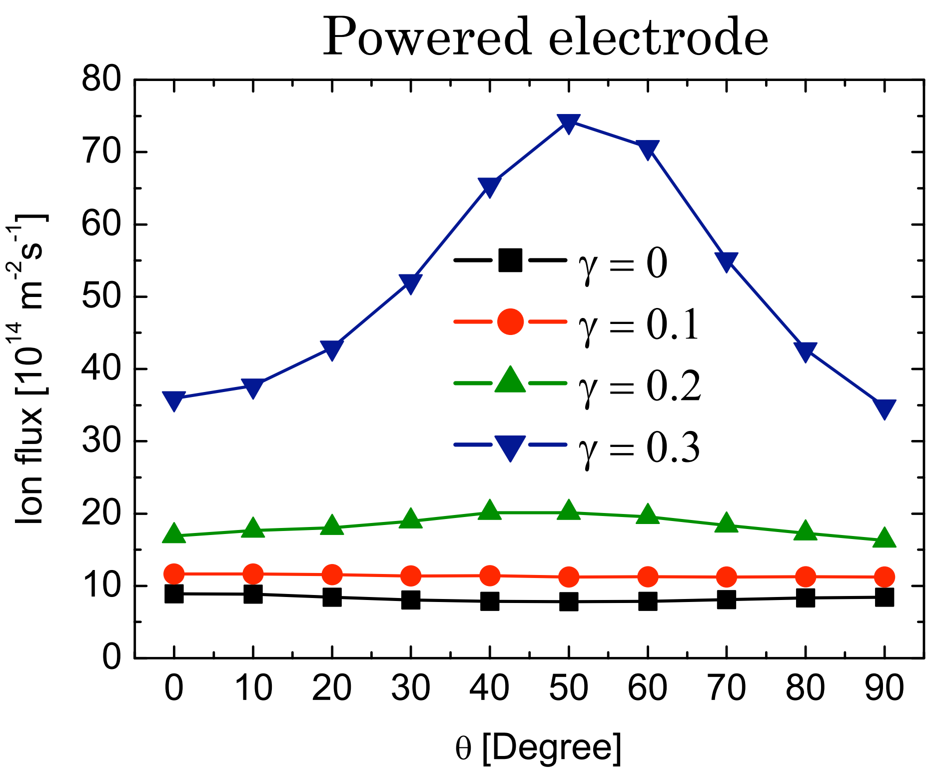
low pressure: nonlocal ionization → *similar* dependence of ion flux on phase angle at both electrodes

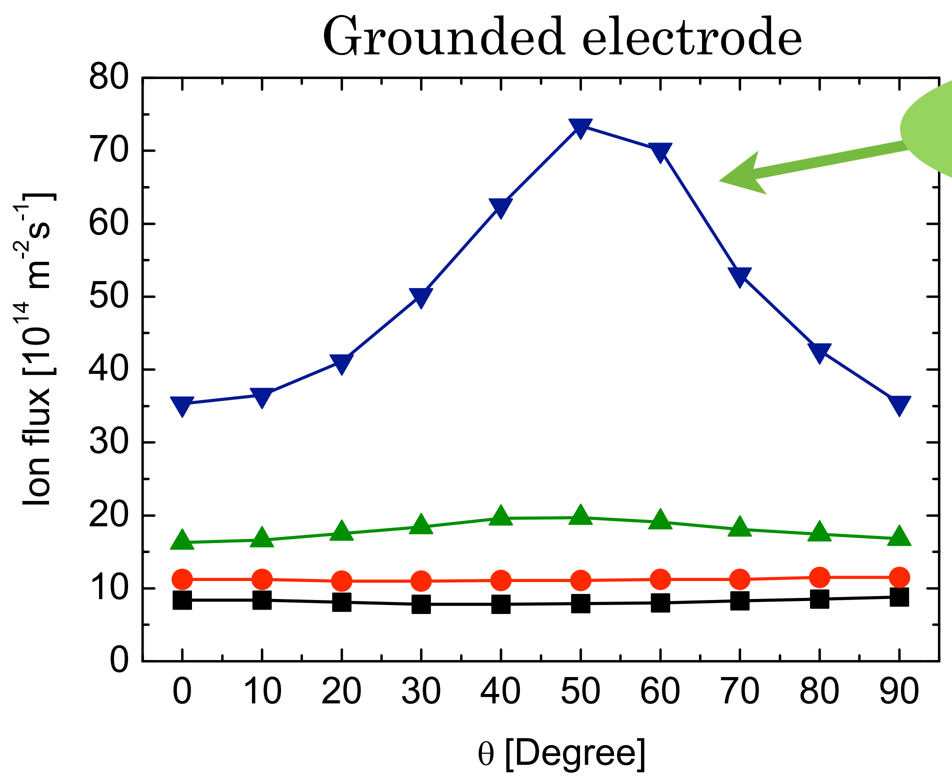


100 Pa

high pressure: ionization source due to secondary electrons is localized at the sheath edges → *opposite* dependence of ion flux on phase angle at both electrodes

Ion flux vs. phase angle : low / high pressure at high γ

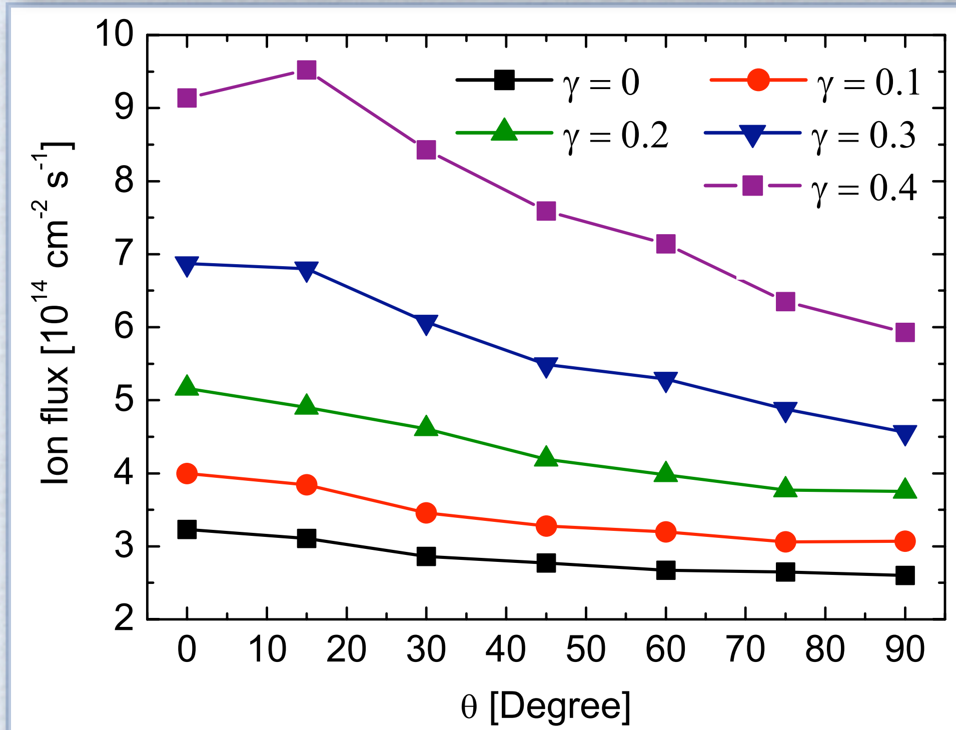


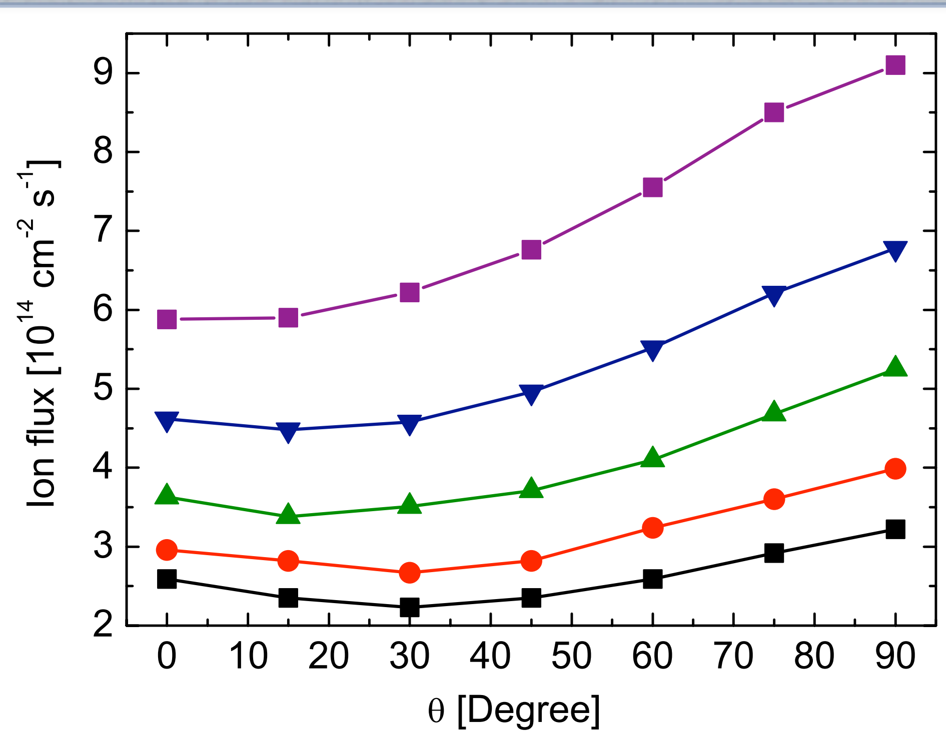


How is this peak formed?

6.6 Pa

low pressure: nonlocal ionization → *similar* dependence of ion flux on phase angle at both electrodes





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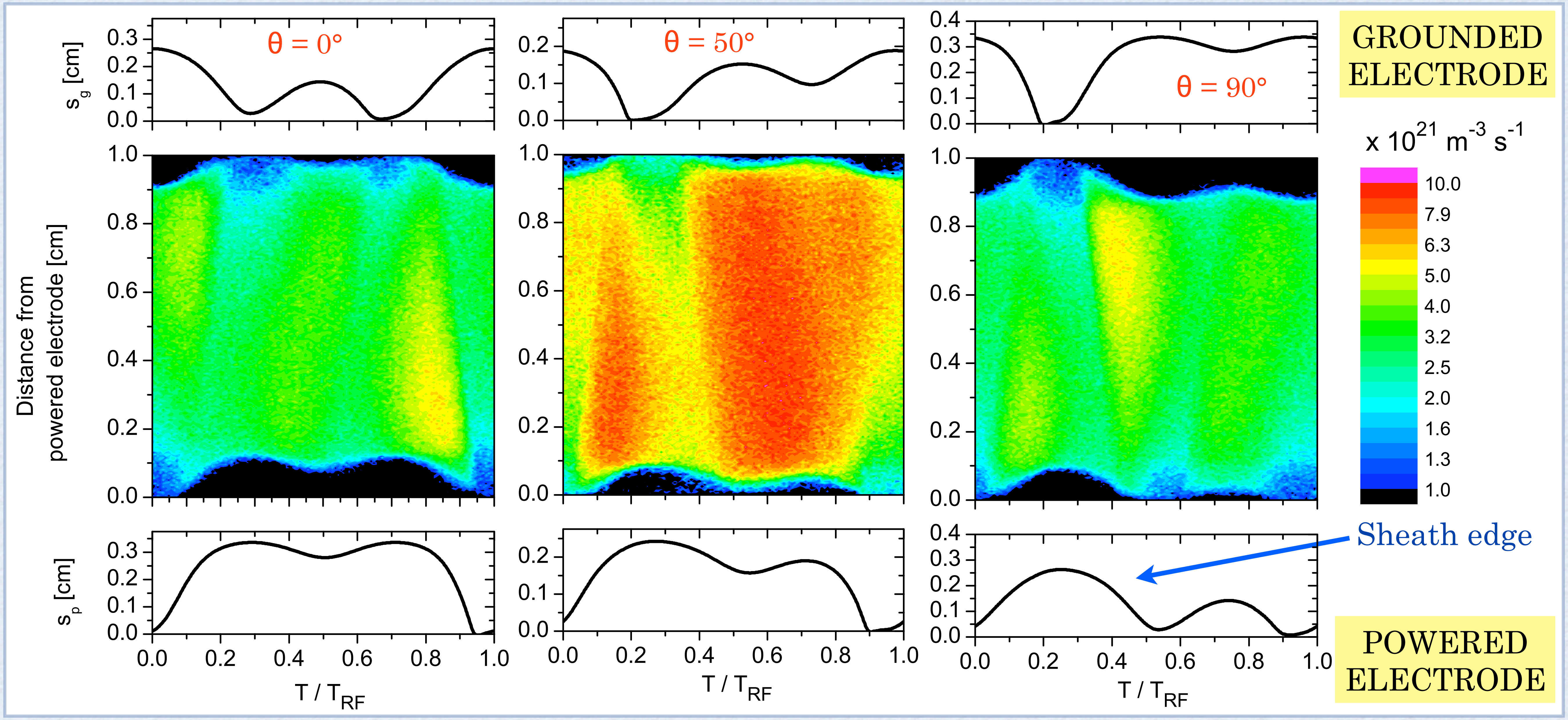
Ionization rate vs. phase angle @ low pressure (6.6 Pa)



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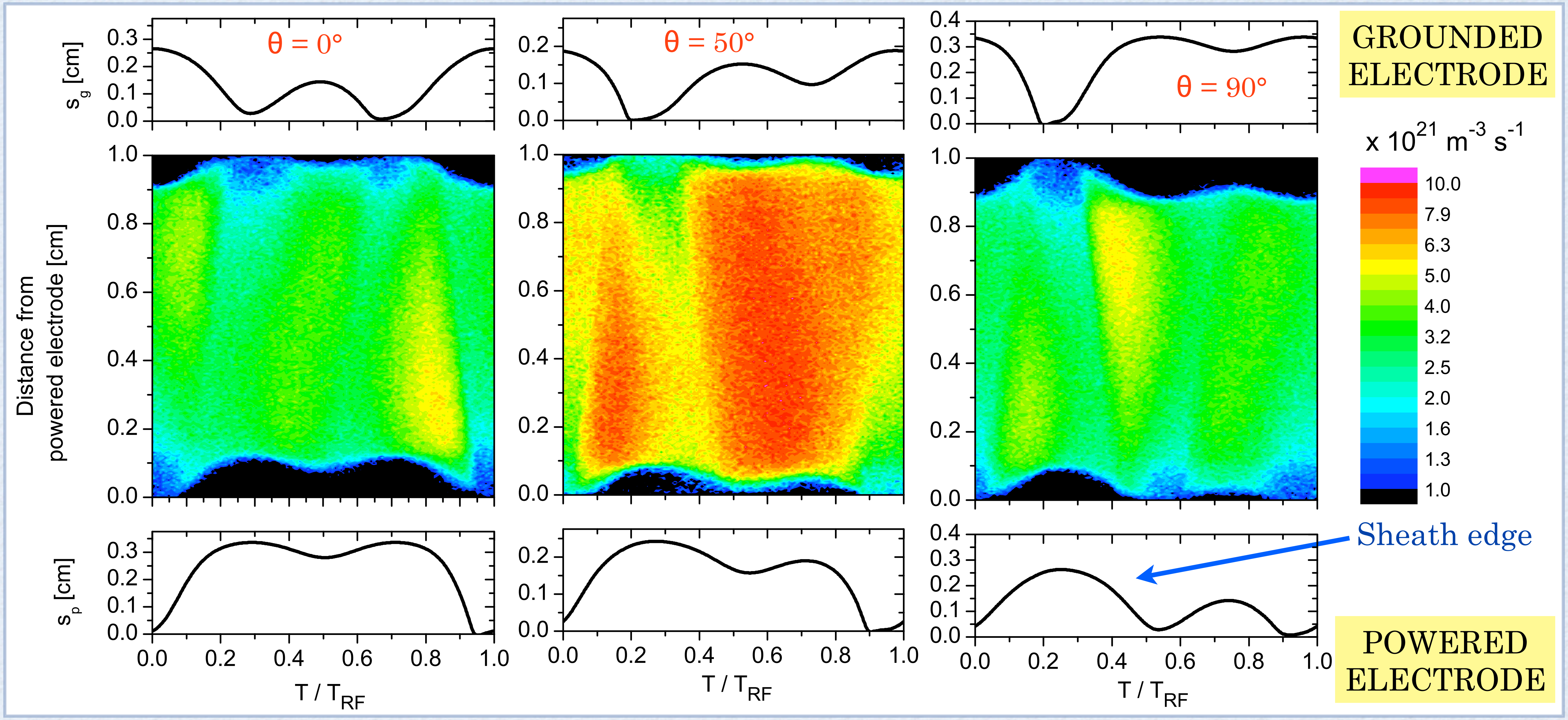

At θ = 0° most γ-electrons originate from the bottom sheath, propagate through the bulk, arrive at the top electrode during sheath collapse, and are lost (vice versa at θ = 90°) → poor confinement of γ-electrons → low ion flux

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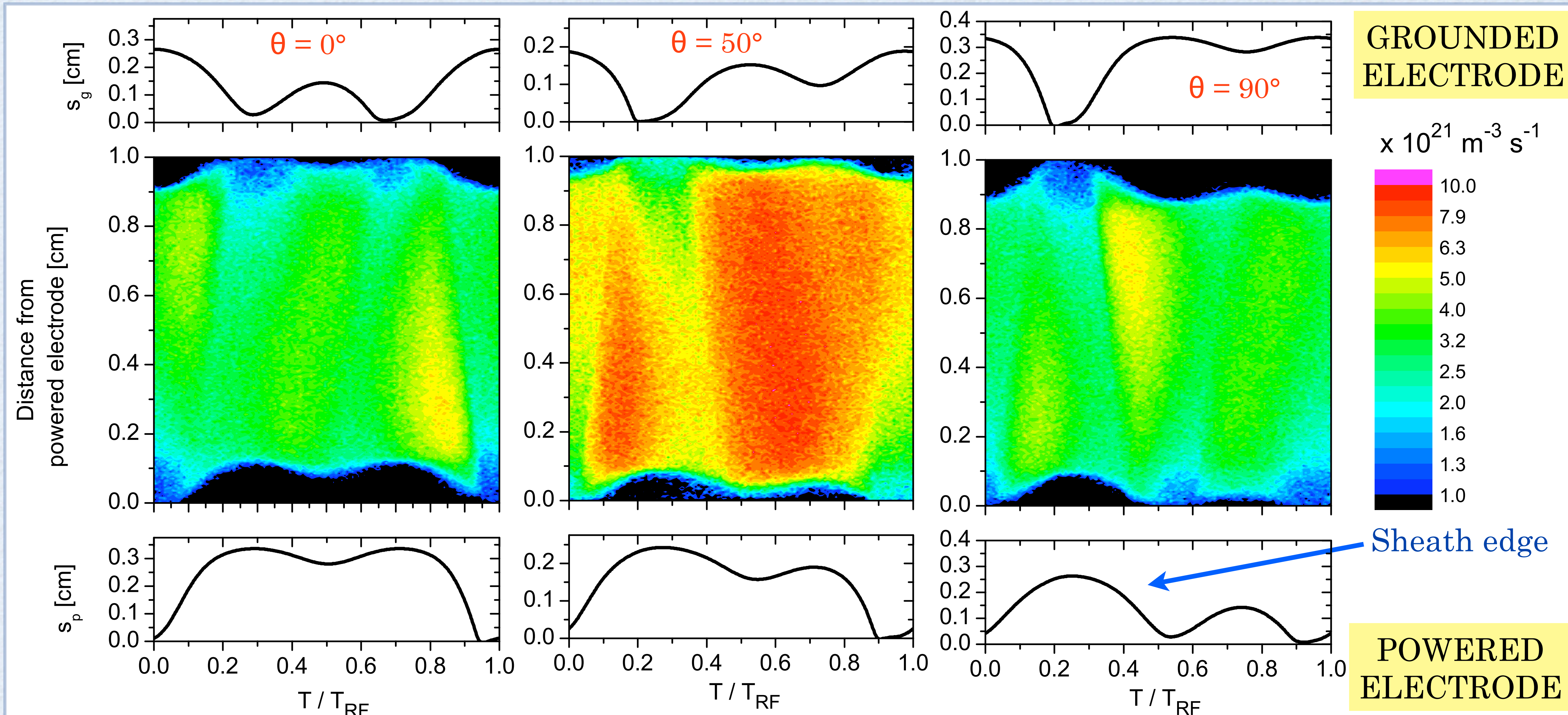

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At θ = 50° γ-electrons originating from one sheath arrive at the other electrode at phases, when the sheath is expanded → γ-electrons are reflected back into the bulk → good confinement of γ-electrons → high ion flux

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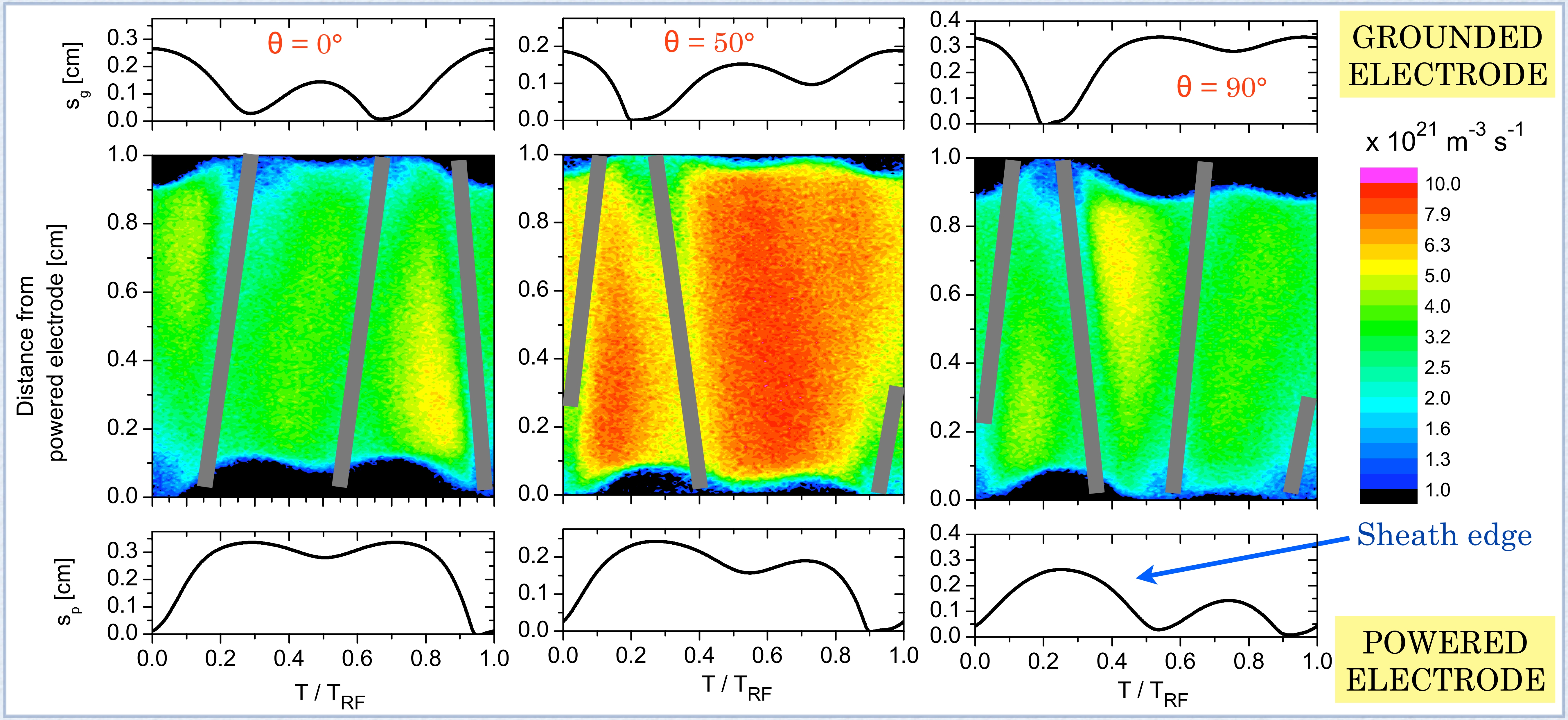

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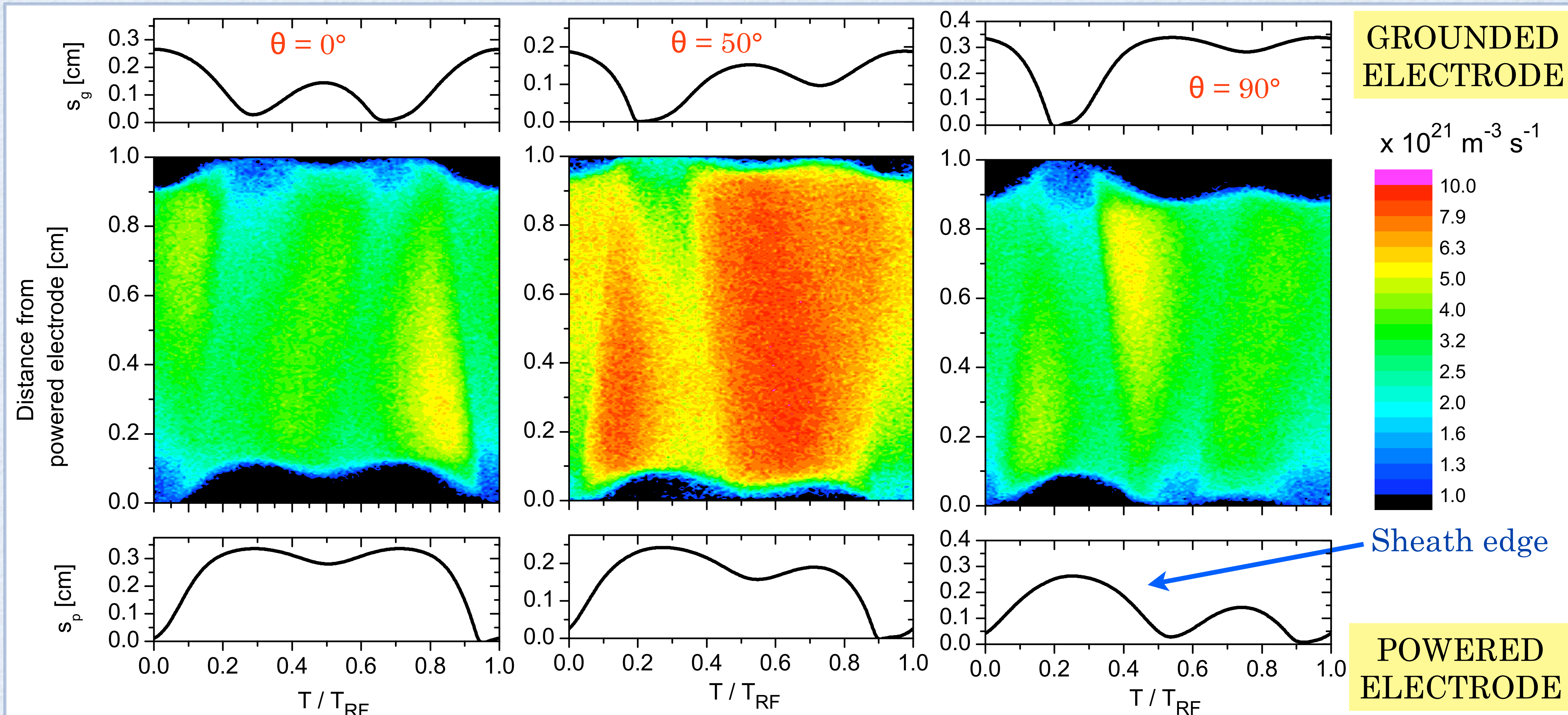

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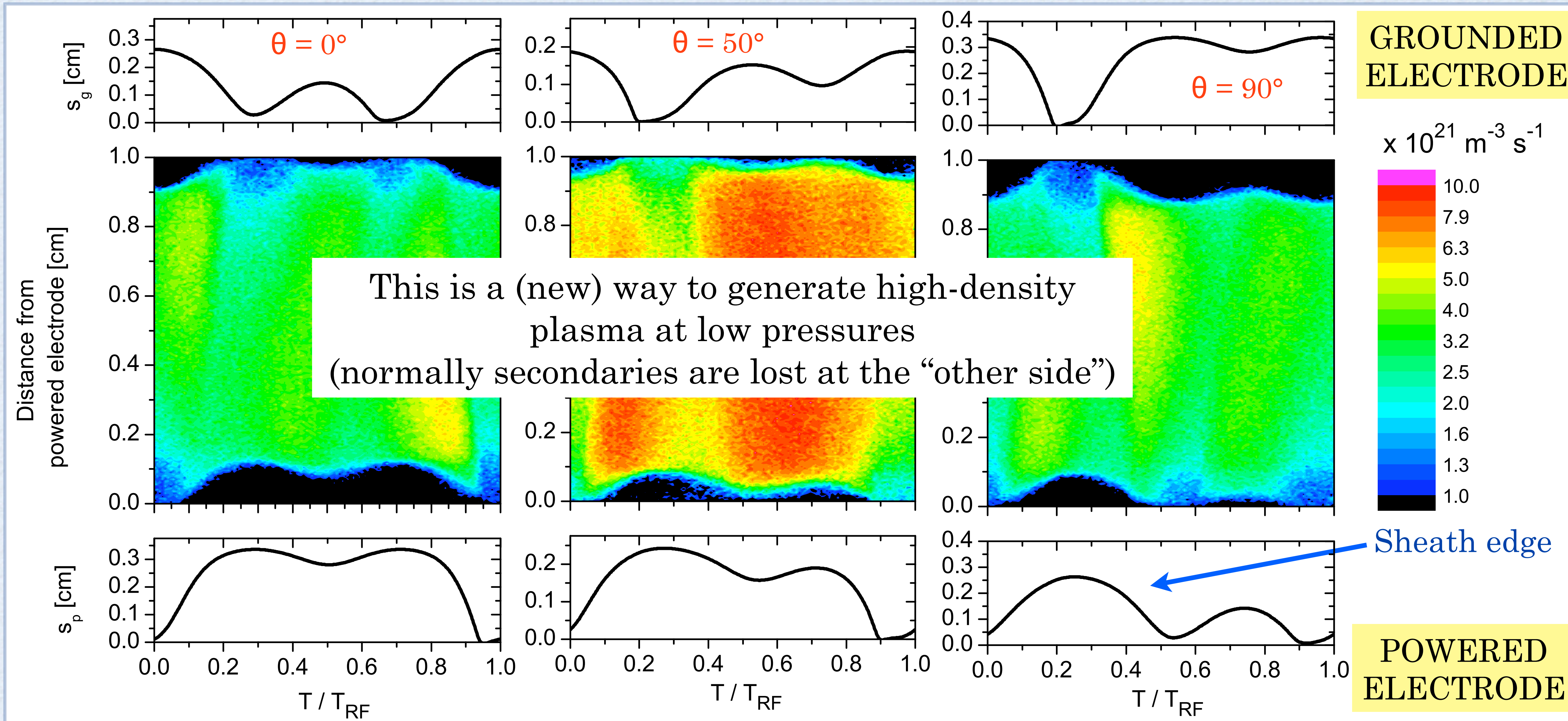

This is a (new) way to generate high-density plasma at low pressures
(normally secondaries are lost at the "other side")

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Summary

- Importance of independent control of ion flux and ion energy
- The effect of secondary electrons:
  - classical dual-frequency discharges: independent control of ion properties is limited to a narrow range of discharge parameters
  - electrical asymmetry effect: good control, compared to classical DF discharges, except at high secondary yield values
- PIC/MCC simulations provided explanations / evidences for these effects

This material is available at : http://plasma.szfki.kfki.hu/~zoli/presentations

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- All of you for your attention

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Progress on simulations of multiple-frequency capacitively coupled discharges - References

- Electric field reversals in the sheath region of capacitively coupled radio frequency discharges at different pressures
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  Z. Donkó, J. Schulze, B. G. Heil and U. Czarnetzki, J. Phys. D 42, 025205 (2009)
- Self-excited nonlinear plasma series resonance oscillations in geometrically symmetric capacitively coupled radio frequency discharges
  Z. Donkó, J. Schulze, U. Czarnetzki and D. Luggenhölscher, Appl. Phys. Lett 94, 131501 (2009)
- Different modes of electron heating in dual-frequency capacitively coupled radio frequency discharges
  J. Schulze, Z. Donkó , D. Luggenhölscher and U. Czarnetzki, PSST 18, 034011 (2009)
- The Electrical Asymmetry Effect - A novel and simple method for separate control of ion energy and flux in capacitively coupled RF discharges
  U. Czarnetzki, B. G. Heil, J. Schulze, Z. Donkó, T. Mussenbrock and R. P. Brinkmann, J. Phys.: Conf. Ser. 162, 012010 (2009)
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  J. Schulze, E. Schungel, Z. Donko, U. Czarnetzki, J. Phys. D 43, 225201 (2010)
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