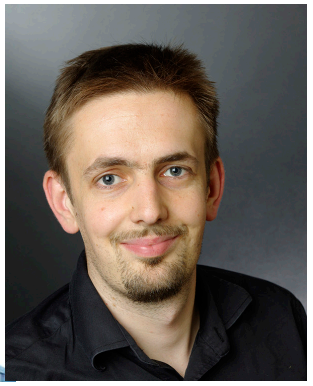
Fundamental investigations of capacitive radio frequency plasmas: simulations and experiments

Z. Donkó[1], J. Schulze[2], U. Czarnetzki[2], A. Derzsi[1], P. Hartmann[1], I. Korolov[1], E. Schüngel[2]

[1] Electrical Discharges & Plasma Physics Research Group
Institute for Solid State Physics and Optics, Wigner Research Centre for Physics
H-1121 Budapest, Konkoly-Thege M. str. 29-33, Hungary

[2] Institute for Plasma and Atomic Physics
Ruhr University Bochum
44780 Bochum, Germany

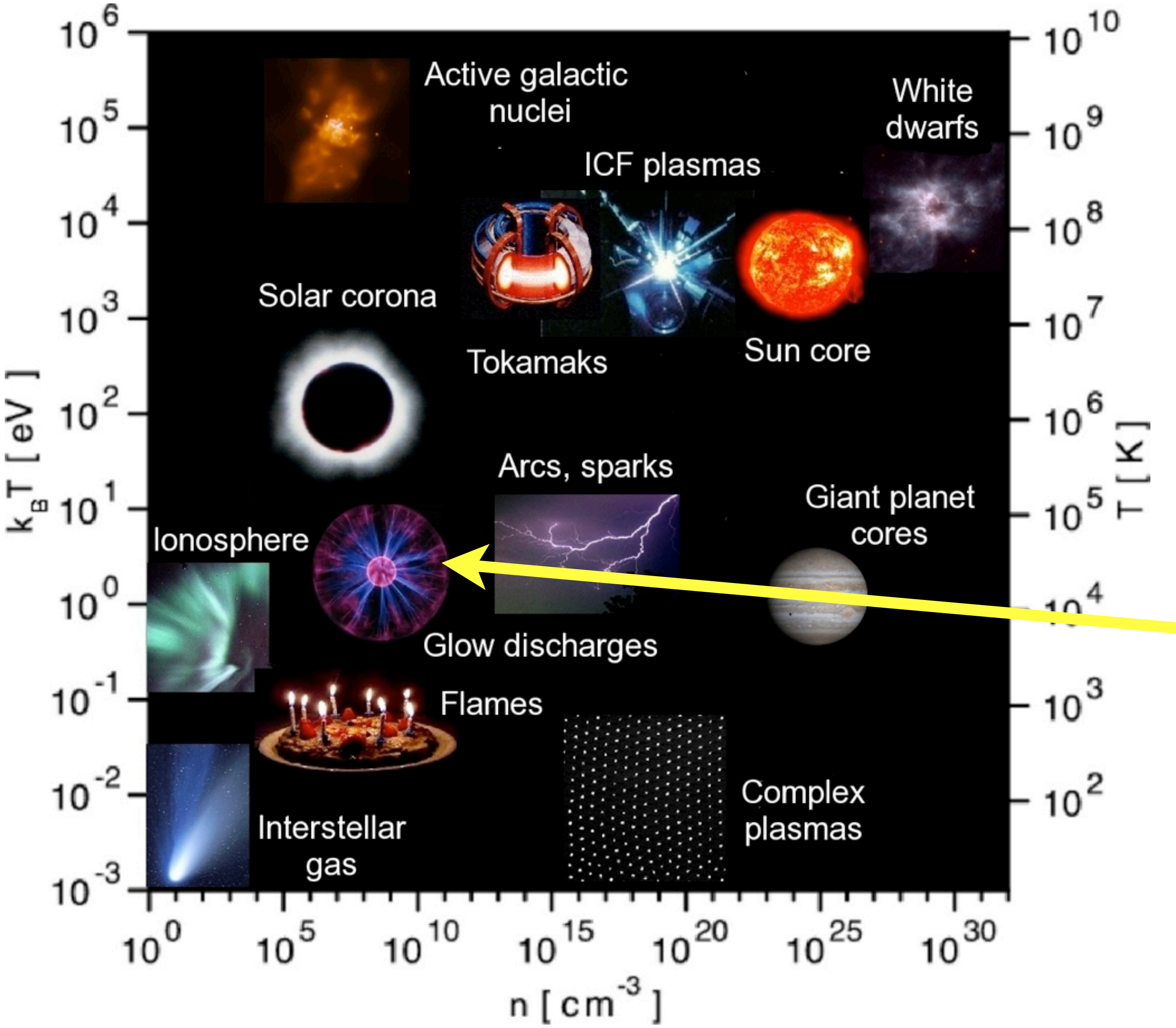
Outline

- Why capacitive radiofrequency discharges?
- Methods (models, simulation technique, diagnostics)
- Different modes of electron heating
- Control of ion properties:
  - “Classical” dual-frequency discharges
  - Effects of secondary electrons in dual-frequency discharges
  - The Electrical Asymmetry Effect
- The Plasma Series Resonance effect

Why capacitive radiofrequency discharges?

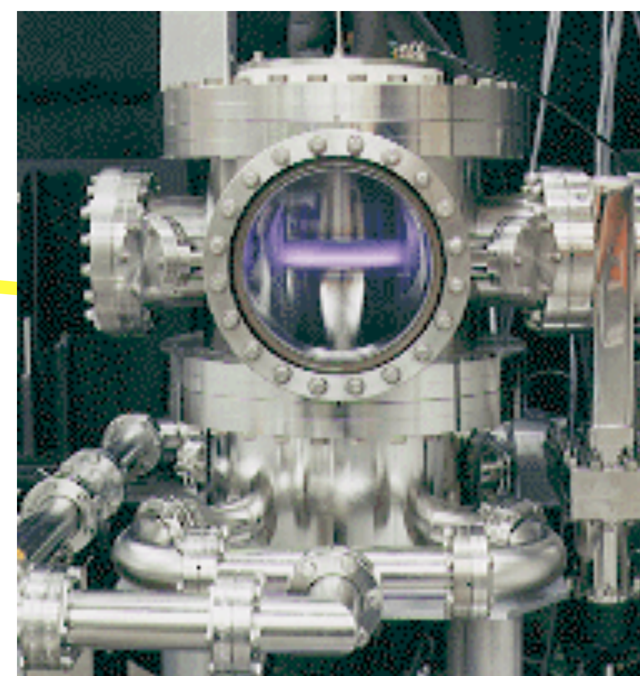
(useful? interesting? both!)

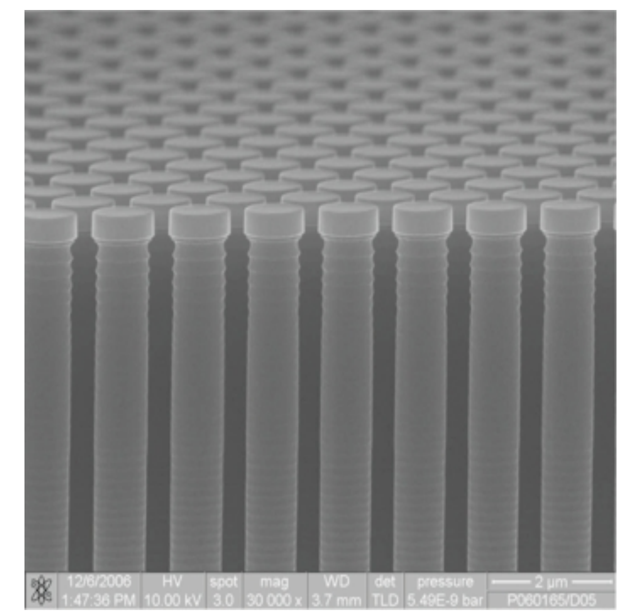
Capacitively coupled radiofrequency discharges

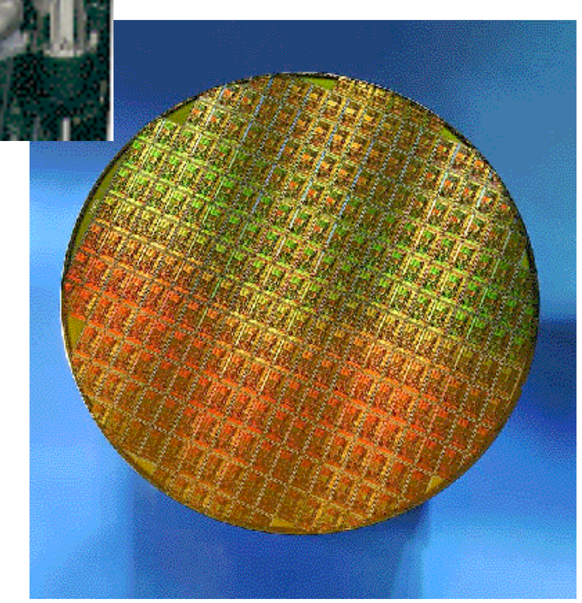


CCRF discharges are used in etching and deposition processes for production of:

- integrated circuits
- solar cells
- biocompatible surfaces

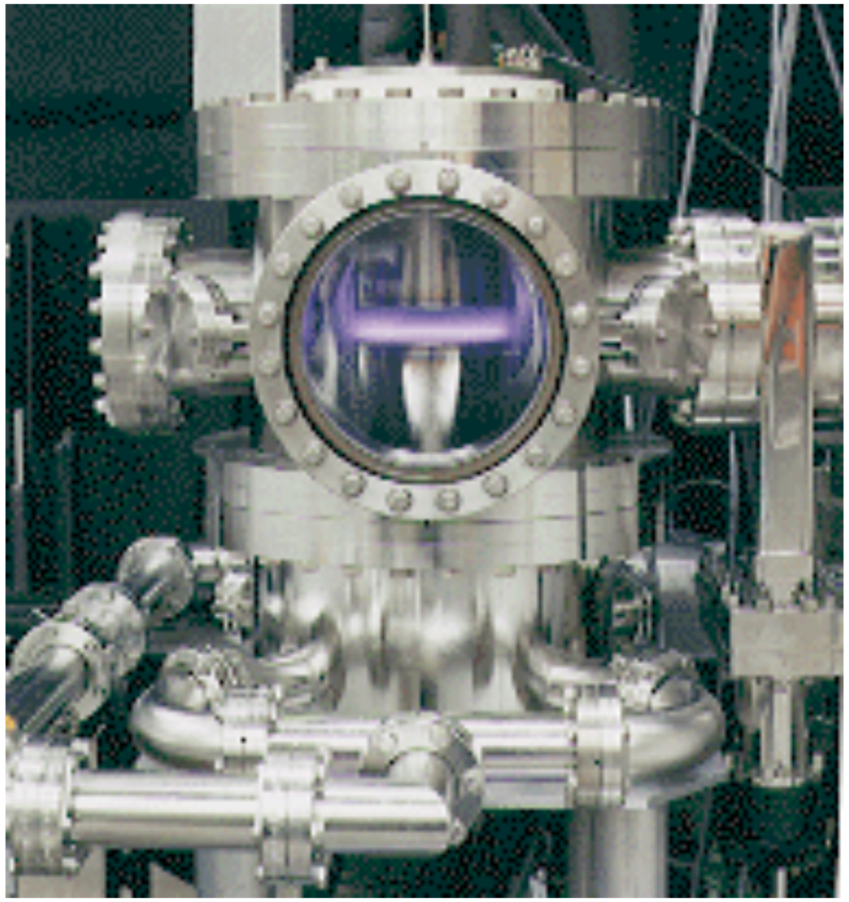






- T_g < 1000 K, ionization by electrons (nonequilibrium systems)
- Low ionization degree
- Electron energy in bulk plasma: 0.1 -10 eV
- Ion energy in bulk plasma: kT_g , 0.05 - 0.1 eV
- Ion energy at electrodes : 1 – 1000 eV

Capacitively coupled radiofrequency discharges



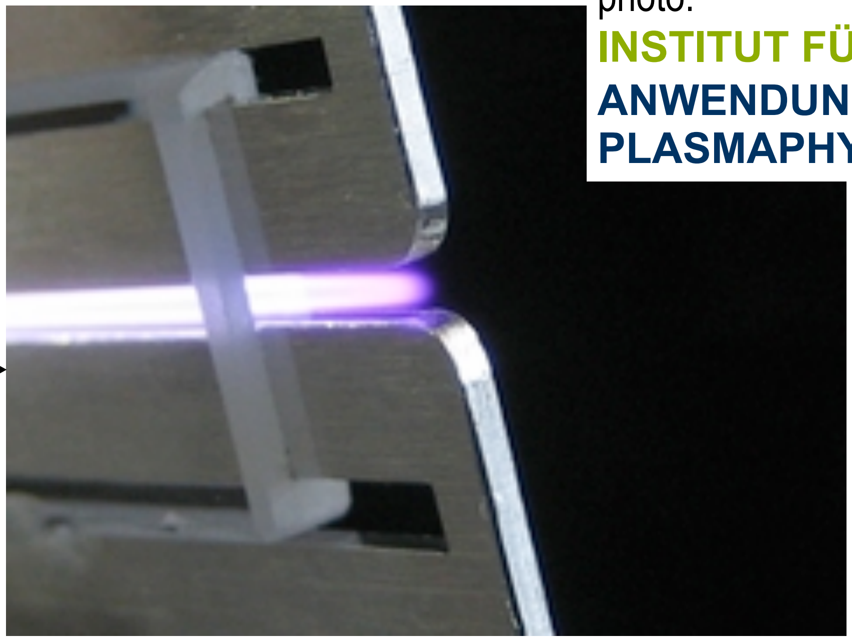


photo:
INSTITUT FÜR EXPERIMENTALPHYSIK II
ANWENDUNGS-ORIENTIERTE PLASMAPHYSIK

Atmospheric Pressure Plasma Jets

- homogeneous plasma (↔ DBD)
- low temperature

Applications:

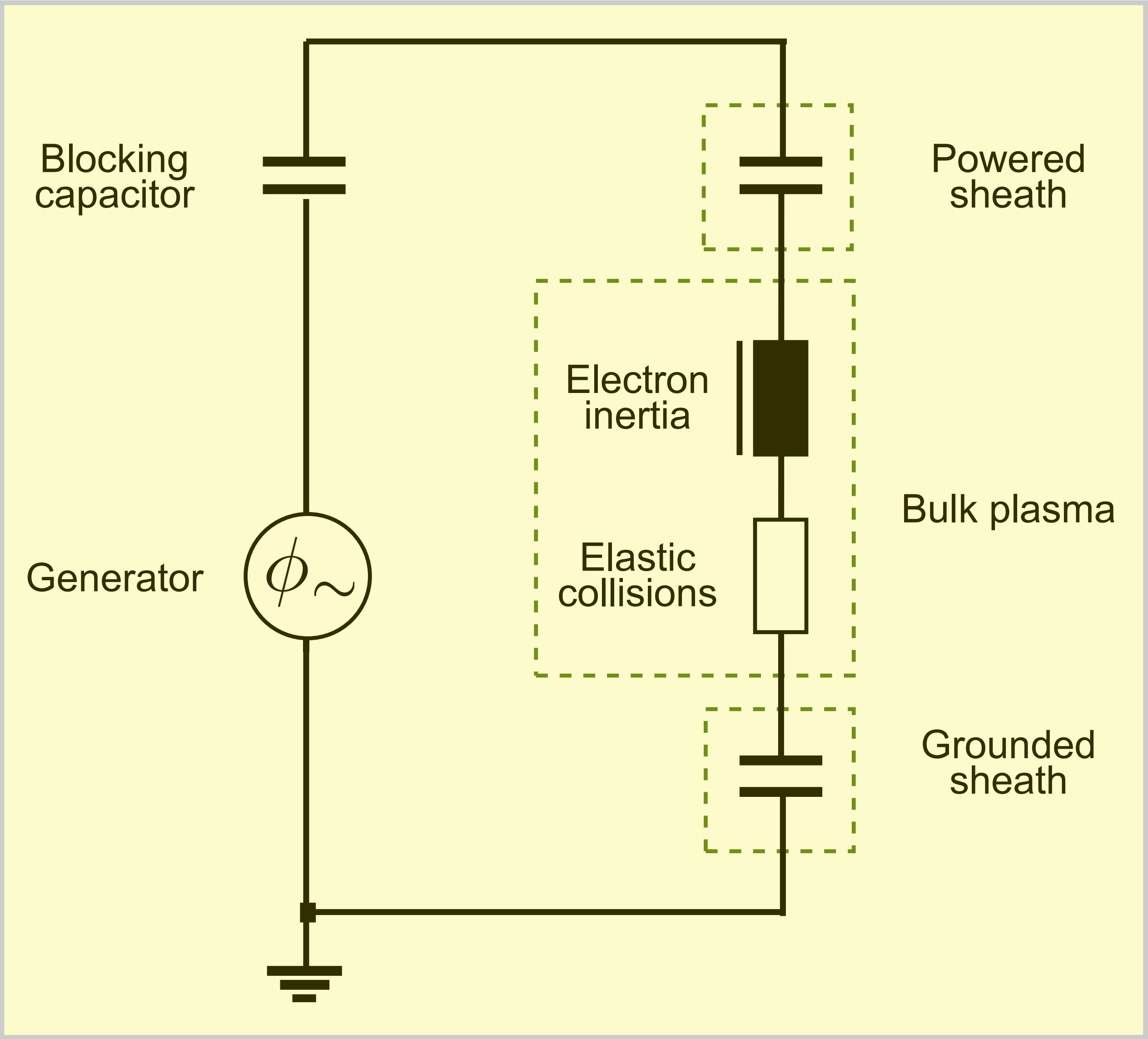
- biomedical technologies
- semiconductor technology
- restoration, ...

e.g.
Park G Y, Park S J, Choil G, Kool G, Byun J H, Hong J W, Sim J Y, Collins G J and Lee J K 2012 Plasma Sourc. Sci. Technol. 21 043001;
Benedikt J, Hofmann S, Knake N, Bottner H, Reuter R, von Keudell A and Schulz-von der Gathen V 2010 Europ. Phys. J. D 60 539;
Knake N, Reuter S, Niemi K, Schulz-von der Gathen V and Winter J 2008 J. Phys. D 41 194006;
Hemke T, Wollny A, Gebhardt M, Brinkmann R P and Mussenbrock T 2011 J. Phys. D 44 285206,
L. Schaper, J. Waskoenig, M. G. Kong, V. Schulz-von der Gathen, T. Gans, IEEE Transactions on Plasma Science 39, (2011), 2370.
J. Waskoenig, THIS CONFERENCE

Methods

Theoretical models
Numerical simulations
Plasma diagnostics

CCRF discharges: Theoretical model



Discharge voltage:

$$\phi(t) = \eta + \phi_\sim(t) = \eta + \sum_{k=1}^{N} \phi_k \cos(2\pi f_k t + \Theta_k)$$

Total driving voltage amplitude: $\phi_{tot} = \sum_{k=1}^{N} \phi_k$

Voltage balance:

$$\phi_\sim(t) + \eta = \phi_{sp}(t) + \phi_b(t) + \phi_{sg}(t)$$

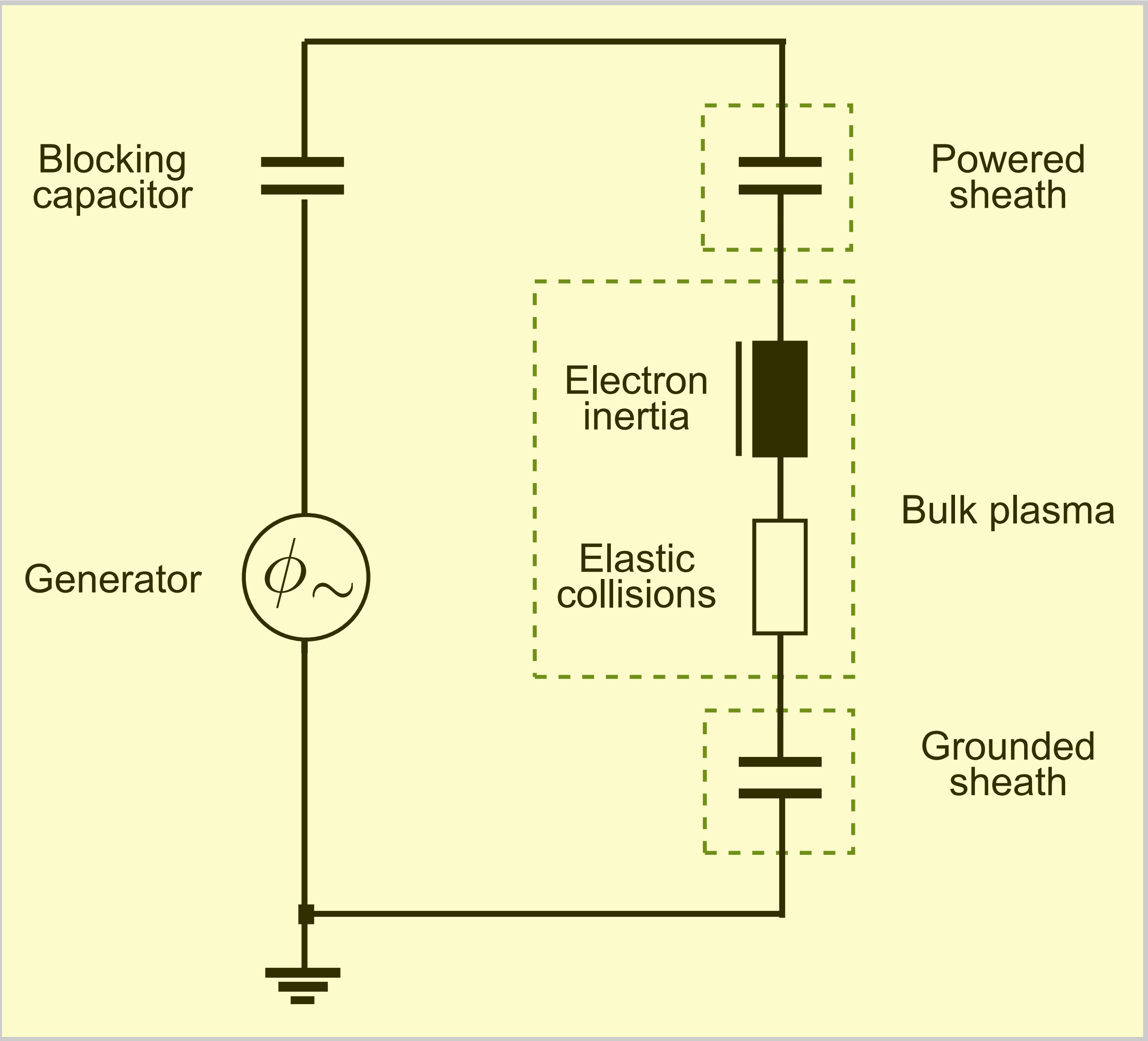
Symmetry parameter: $\varepsilon = \left| \frac{\hat{\phi}_{sg}}{\hat{\phi}_{sp}} \right|$

Derive the dc self-bias from equations for the times for the maximum and minimum applied voltage:

$$\eta = -\frac{\phi_{\sim max} + \varepsilon \phi_{\sim min}}{1+\varepsilon} + \frac{\phi_{sp}^f + \varepsilon \phi_{sg}^f}{1+\varepsilon} + \frac{\phi_{b,max} - \varepsilon \phi_{b,min}}{1+\varepsilon} \cong -\frac{\phi_{\sim max} + \varepsilon \phi_{\sim min}}{1+\varepsilon}$$

Symmetrical generator waveform AND geometrical symmetry → $\eta = 0$
Asymmetric generator waveform AND/OR geometrical asymmetry → $\eta \neq 0$

CCRF discharges: Theoretical model



U. Czarnetzki, T. Mussenbrock and R. P. Brinkmann, Phys. Plasmas 13, 123503 (2006)
U. Czarnetzki, J. Schulze, E. Schüngel and Z Donkó, Plasma Sources Sci. Technol. 20, 024010 (2011)

$$\phi_\sim(t) + \eta = \phi_{sp}(t) + \phi_b(t) + \phi_{sg}(t)$$

Sheaths: $$\phi_s(t) = -\frac{1}{2e\epsilon_0 \overline{n}_s A^2} Q_s^2(t)$$

Bulk: $$\phi_b(t) = -\frac{L}{\overline{A}} \frac{m}{e\overline{n}_b} \left(\frac{\partial}{\partial t} + \nu \right) I(t)$$

After normalization by the total voltage amplitude and the maximum charge in the sheath:

$$\overline{\phi}_{sp}[q(t)] = -q^2(t)$$

q: normalized charge in the powered sheath

$$\overline{\phi}_{sg}[q(t)] = \varepsilon[q_t - q(t)]^2$$

Voltage balance expressed in terms of the normalized charge in the powered sheath describes the dynamics of the discharge

$$\overline{\phi}_\sim(t) + \overline{\eta} = -q^2(t) + \varepsilon[q_t - q(t)]^2 - 2\beta^2 \left[\frac{\partial^2}{\partial t^2} + \nu \frac{\partial}{\partial t} \right] q(t)$$

$$\beta^2 = A_p L \hat{\phi}_{sp} \epsilon_0 m / (\overline{A} \hat{s}_p \phi_{tot} e^2 \overline{n}_b)$$

CCRF discharges: Simulations

Particle-in-Cell simulation with Monte Carlo collisions (PIC/MCC)

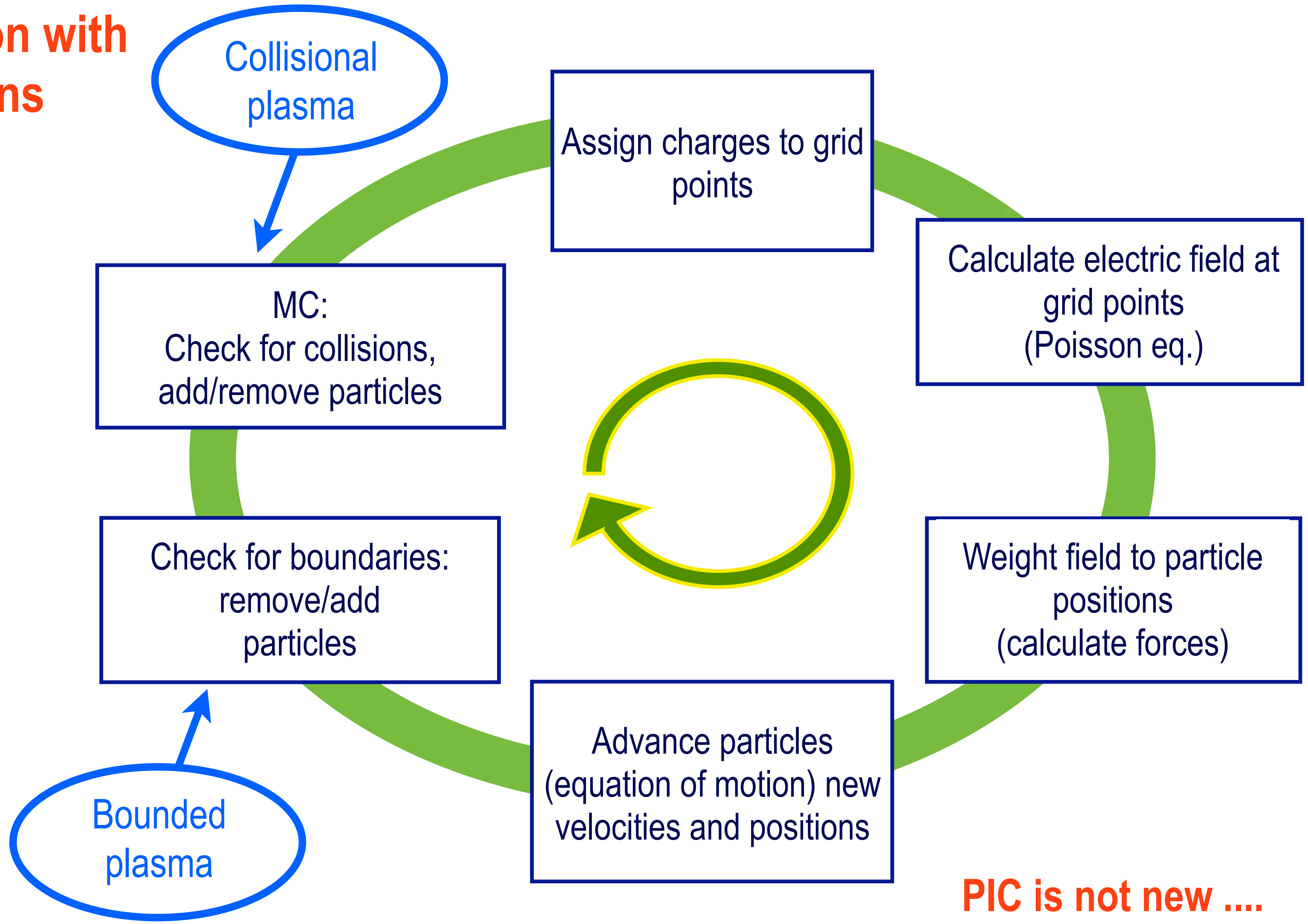
The IDEAS:

Due to the huge number of particles in real plasmas considering all pairwise interactions is hopeless

→ *let particles interact via the electric field*

Still too many particles ...

→ *use superparticles which represent a large number of real particles*

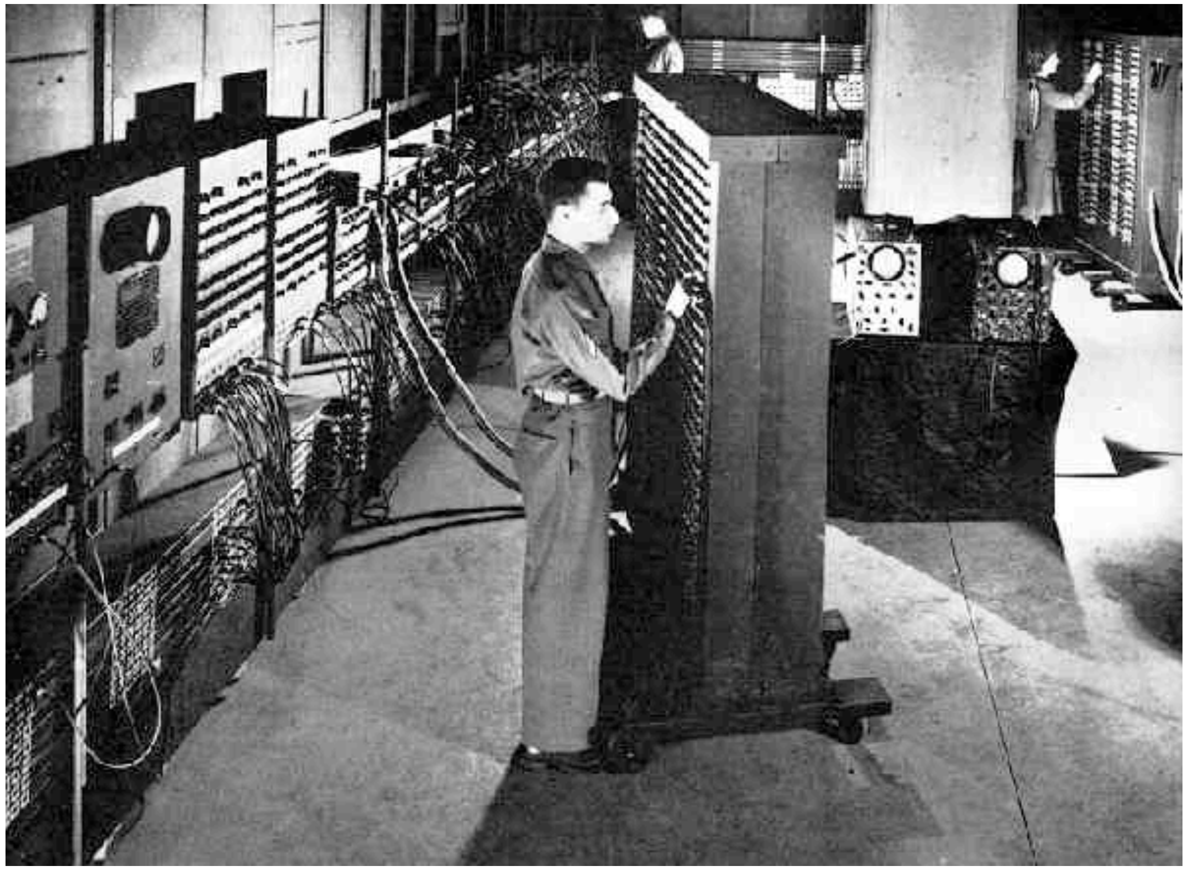


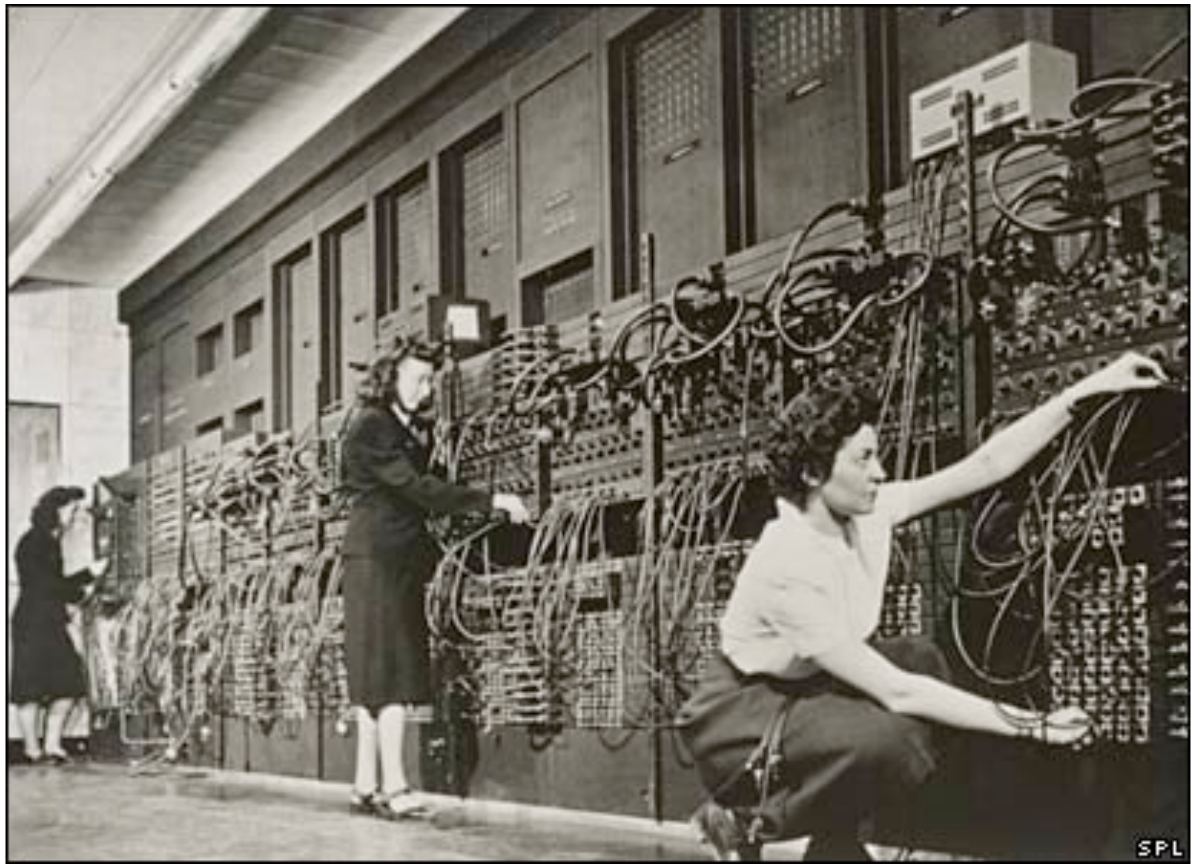
C. K. Birdsall, IEEE Trans. Plasma Sci, **19** (1991).
J. P. Verboncoeur, Plasma Phys. Control. Fusion 47 A231–A260 (2005).
K. Matyash, R. Schneider, F. Taccogna, A. Hatayama, S. Longo, M. Capitelli, D. Tskhakaya, and F. X. Bronold, Contrib. Plasma Phys. 47 595 (2007).

PIC is not new .... what is new:

- **widely available**
- **benchmarking**

Dramatic advance of computational resources ...

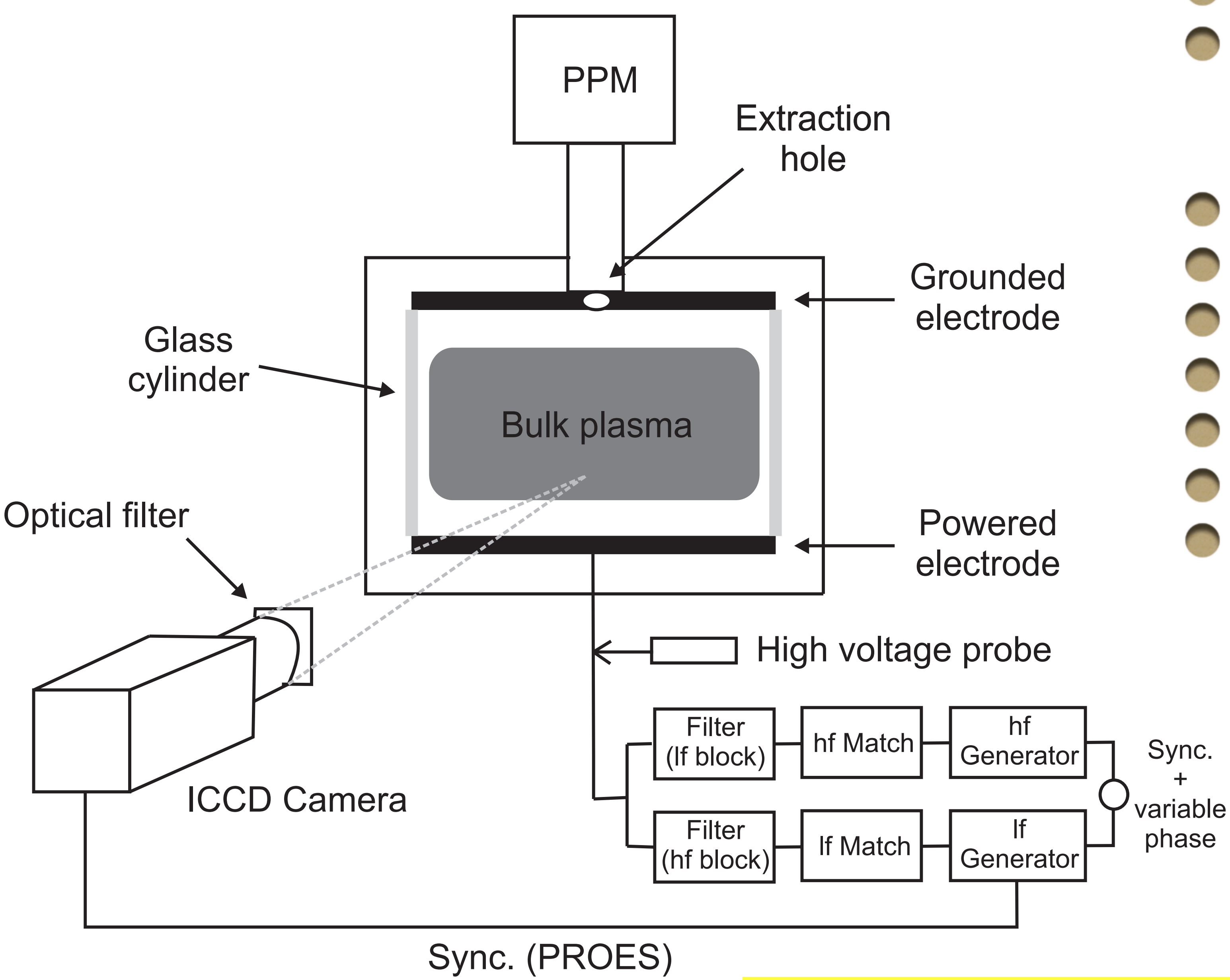




"Where a calculator on the ENIAC is equipped with 18,000 vacuum tubes and weighs 30 tons, computers in the future may have only 1,000 vacuum tubes and weigh only 1.5 tons."
[Popular Mechanics, 1949]

CCRF discharges: Diagnostics

Setup for experimental studies at RUB EP5



- Electrical measurements
- Phase Resolved Optical Emission Spectroscopy
- Plasma Process Monitor

- **Emission Spectroscopy**
- **Atomic Absorption Spectroscopy**
- **Laser Induced Fluorescence**
- **Laser Absorption**
- **Probes**
- **....**
-

J. Schulze, E. Schüngel and U. Czarnetzki, J. Phys. D: Appl. Phys. 42 (2009) 092005

Different modes of electron heating

(electrons are the particles that react first to fields, initiate charged particle production, drive plasma chemistry, ...)

Alpha / gamma modes in capacitive discharges: fluid model

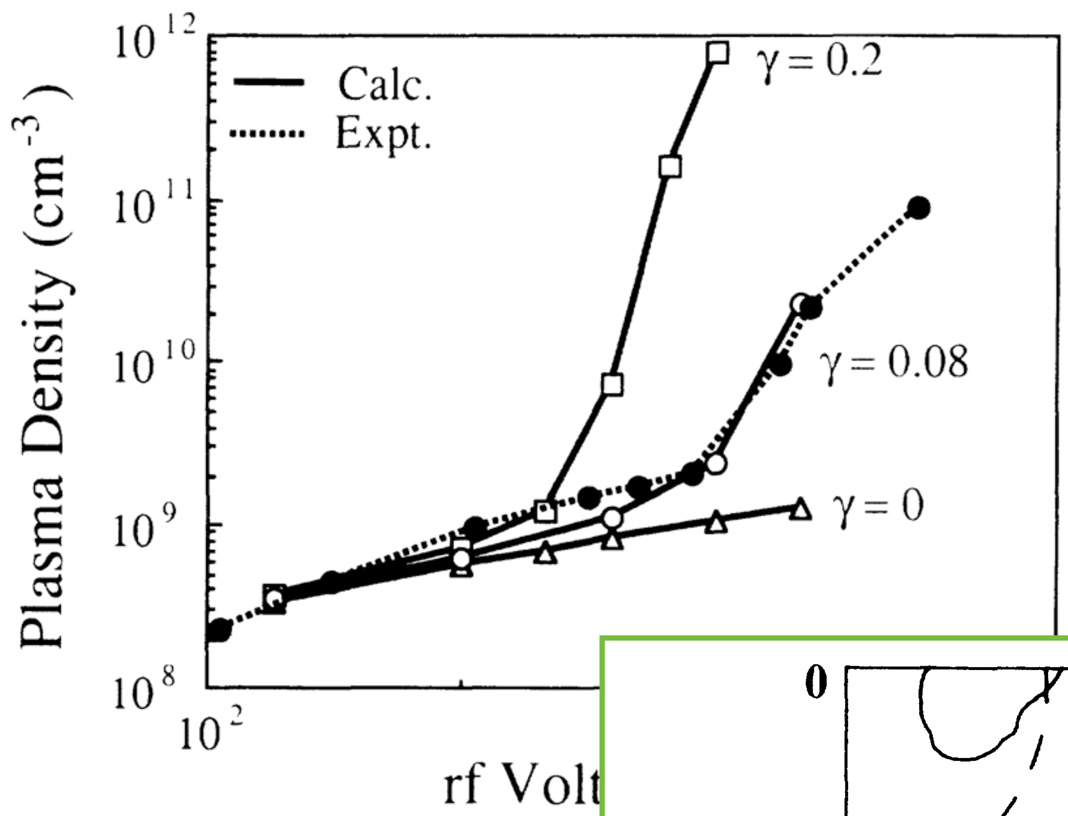
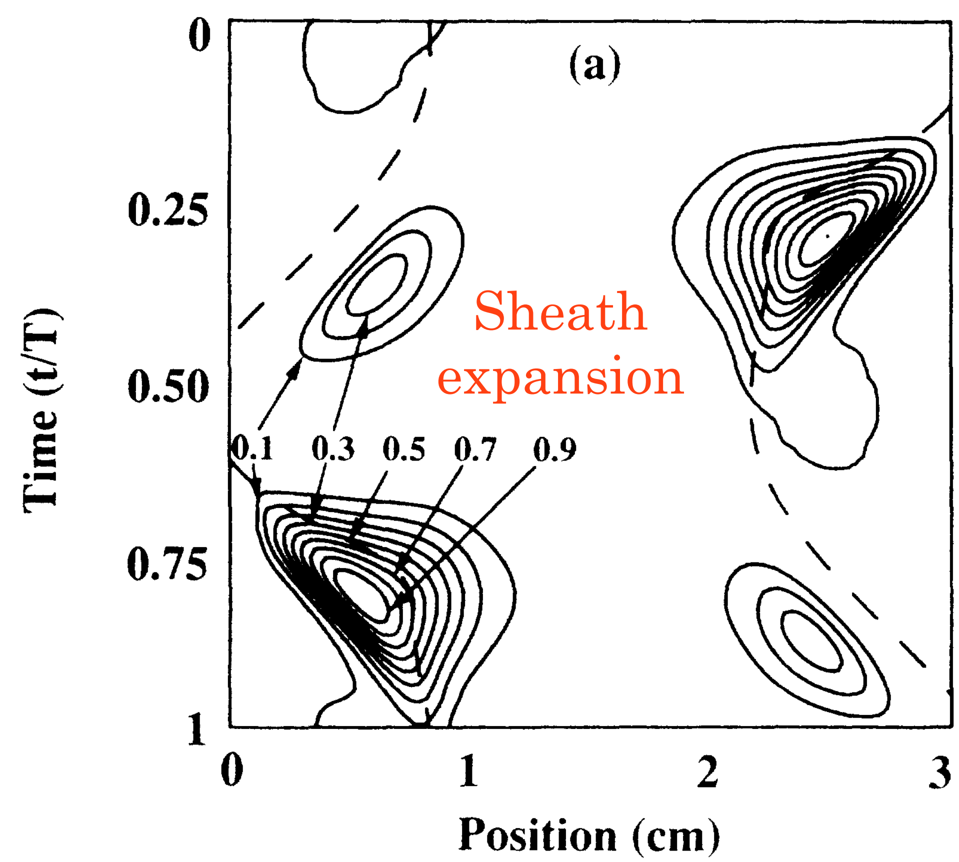
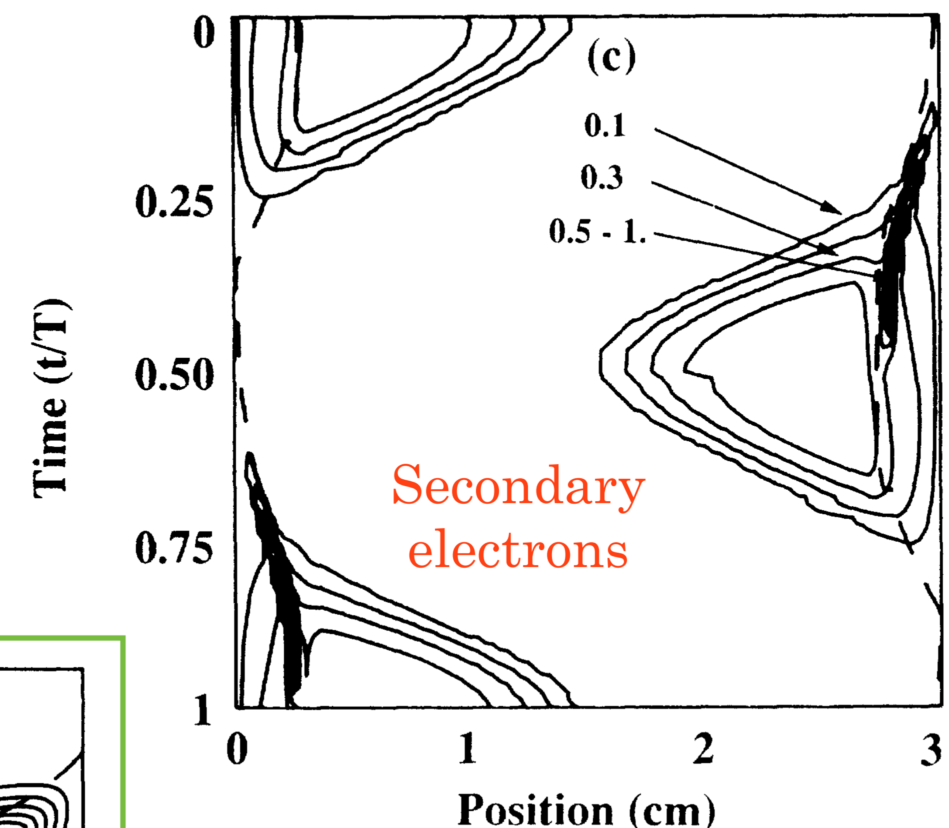


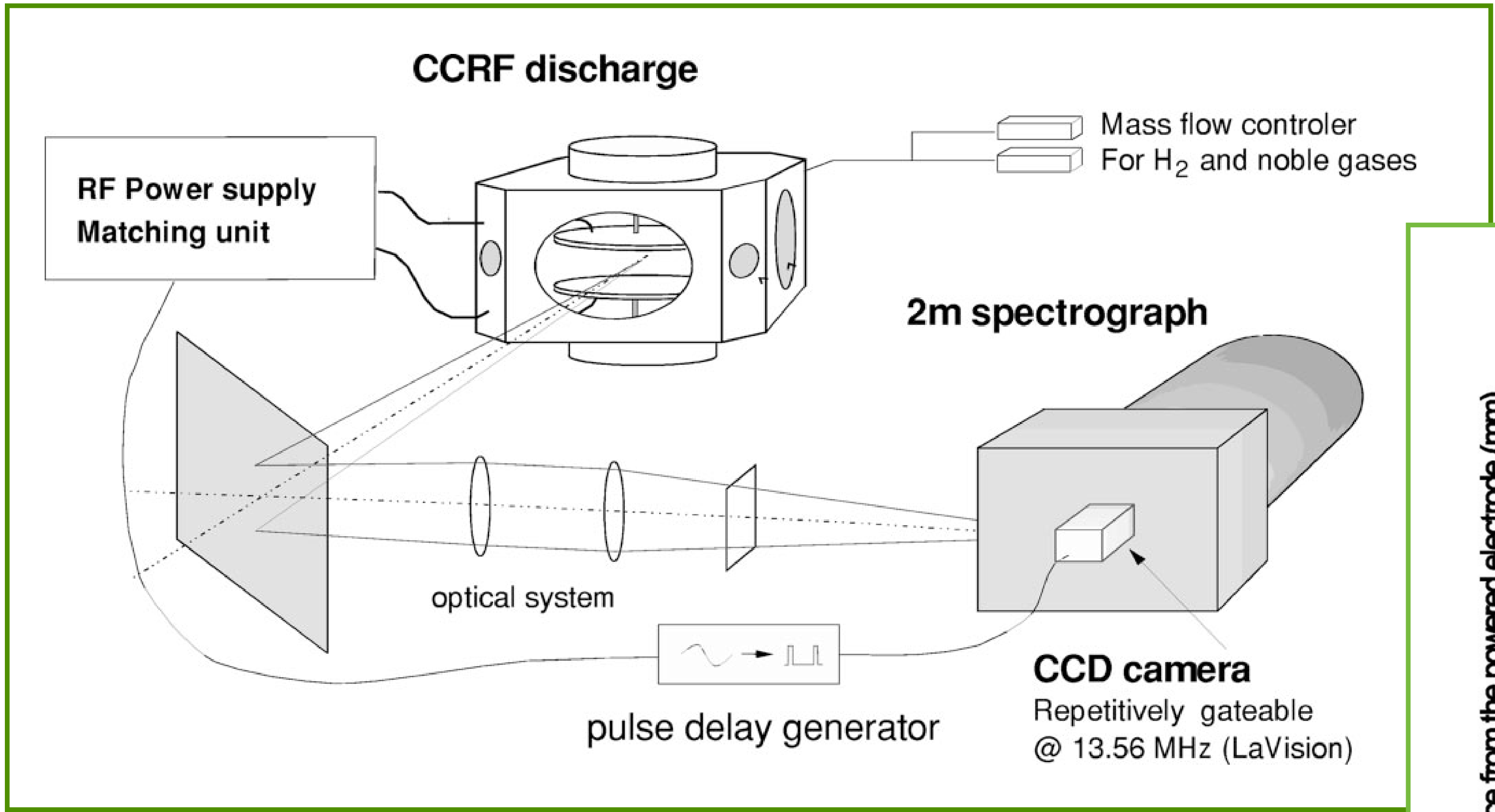
FIG. 2. Variations of the plas
rf voltage amplitude for a dischar
cm, F=3.2 MHz) for three val
emission coefficient (γ=0.0, 0.0
results of Godyak and Kanneh
represented.



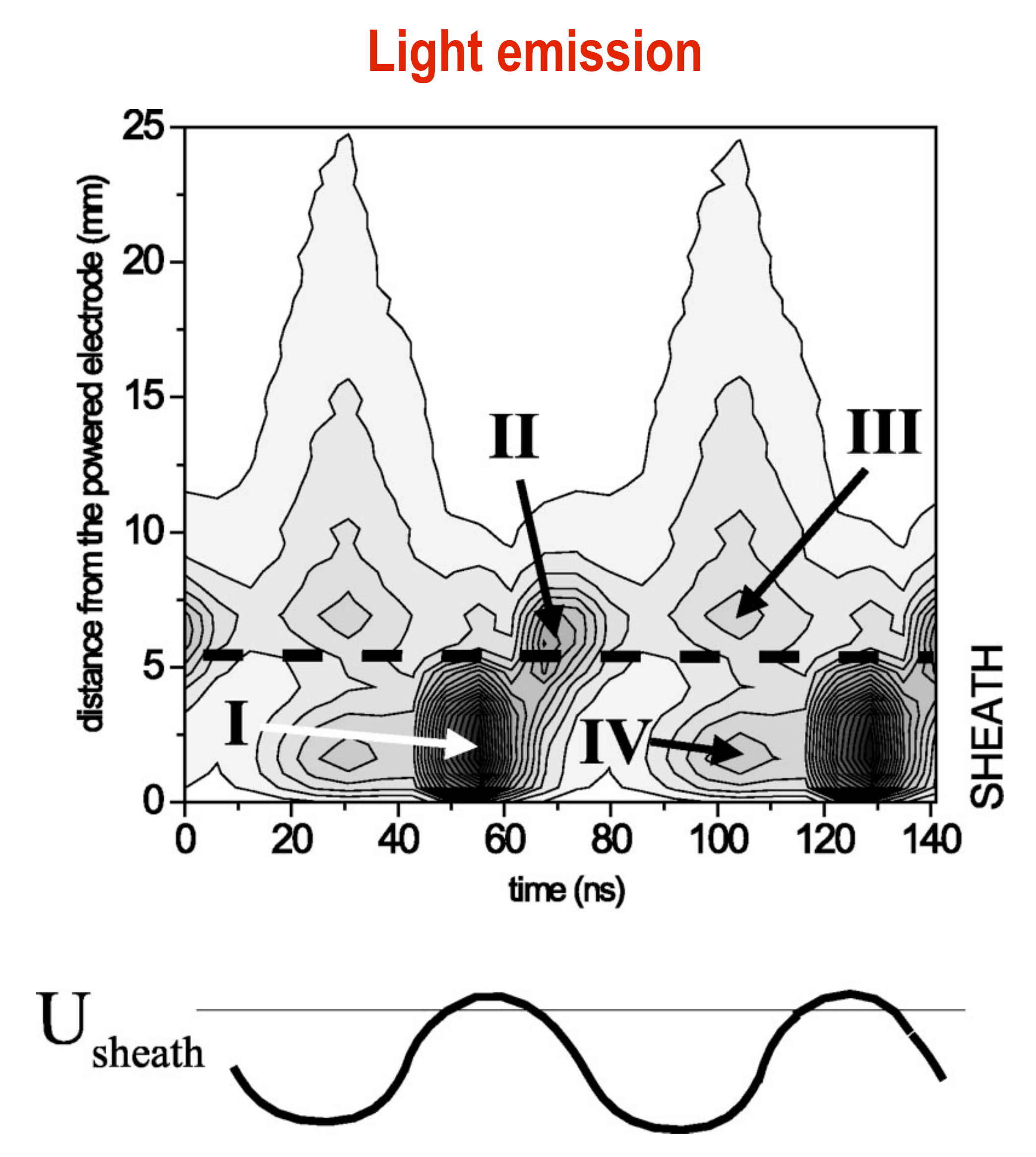


Ph. Belenguer and J.-P. Boeuf,
Phys. Rev. A 41, 4447 (1990)

Alpha / gamma modes in capacitive discharges: experiments



I: field reversal
II: sheath expansion
III: secondary electrons
IV: fast atoms



Phase Resolved Optical Emission Spectroscopy (PROES)

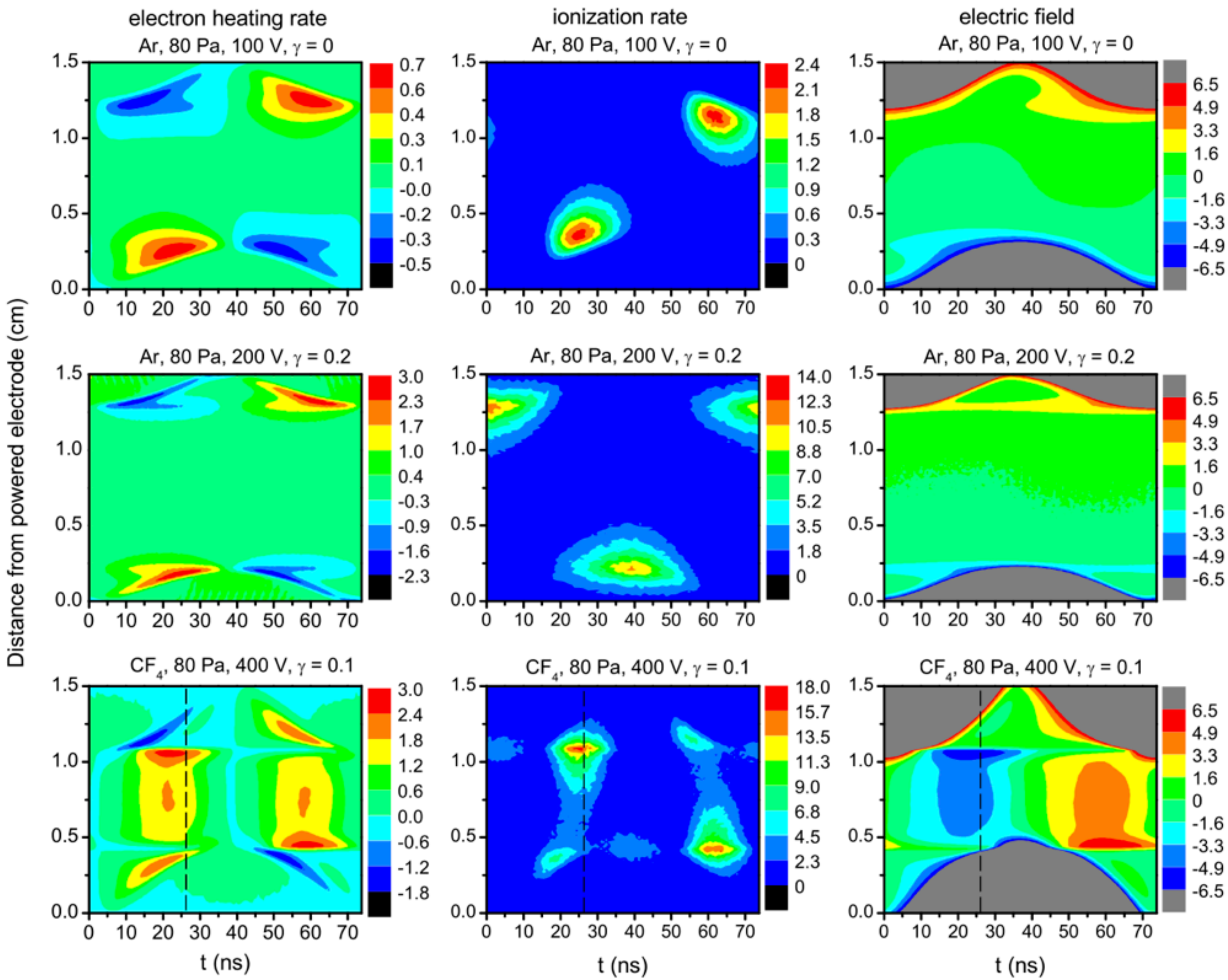
T. Gans, C.C. Lin. V. Schulz-von der Gathen and H. F. Döbele, Phys. Rev. A 67 012707 (2003).

H_2 discharge

Drift-ambipolar electron heating mode in electronegative discharges

EPS ICPP 2012

PIC/MCC results



Argon
alpha mode

Argon
gamma mode

CF_4
drift-ambipolar (DA)
mode

Drift-ambipolar heating mode in electronegative discharges

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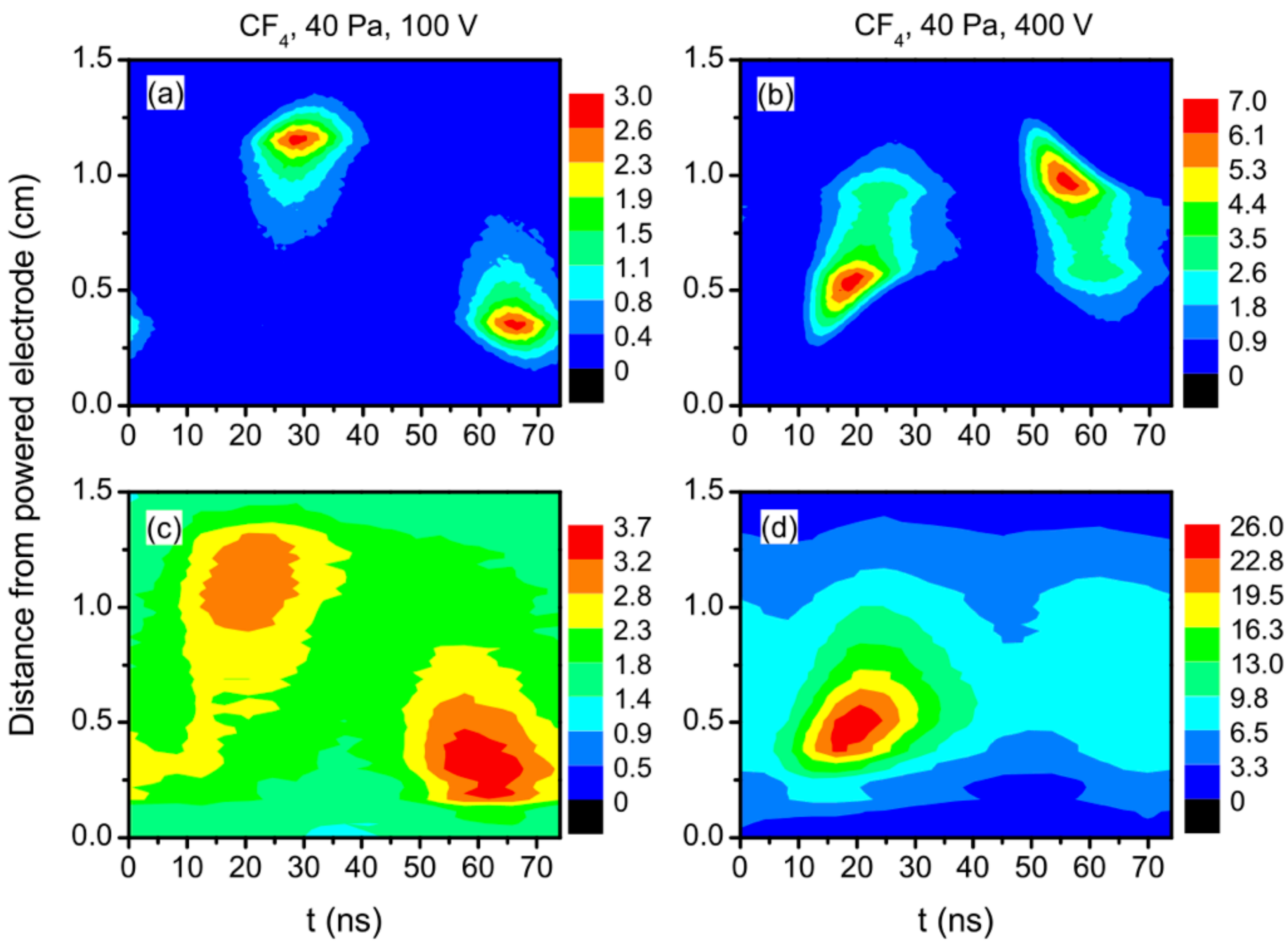
ICPP
2012

DA mode alpha mode



PIC/MCC:
total electronic excitation

J. Schulze, A. Derzsi, K. Dittmann, T. Hemke, J. Meichsner, Z. Donkó
Phys. Rev. Lett. 107, 275001 (2011)

PROES:
emission at 250 nm
(not completely symmetrical due to experimental configuration)

In collaboration with

J. Meichsner, K. Dittmann
Institute of Physics, University of Greifswald

Mode transitions in CF_4 discharges have been seen earlier:
K. Denpoh and K. Nanbu J. Vac. Sci. Technol. A 16 1201 (1998)
K. Denpoh and K. Nanbu, Japan. J. Appl. Phys. 39 2804 (2000)
O. V. Proshina, T. V. Rakhimova, A. T. Rakhimov and D. G. Voloshin, Plasma Sources Sci. Technol. 19 065013 (2010)

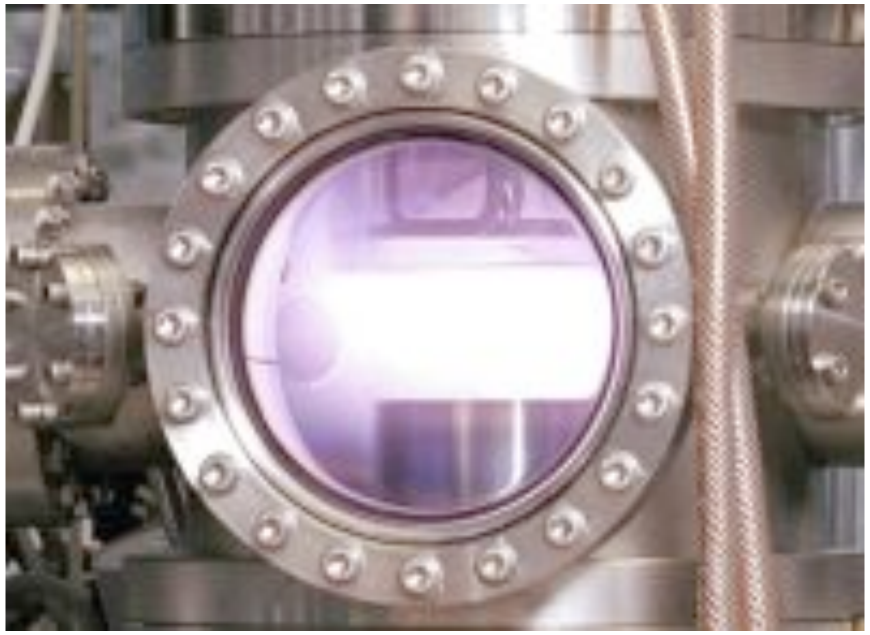
Control of ion properties

Why ion properties?
Limitations of single-frequency discharges
“Classical” Dual-Frequency discharges
The Electrical Asymmetry Effect

Control of ion properties

E Kawamura, V Vahedi, M A Lieberman and C K Birdsall:
"Ion energy distributions in rf sheaths; review, analysis and simulation"
(REVIEW ARTICLE) 1999 Plasma Sources Sci. Technol. 8 R45

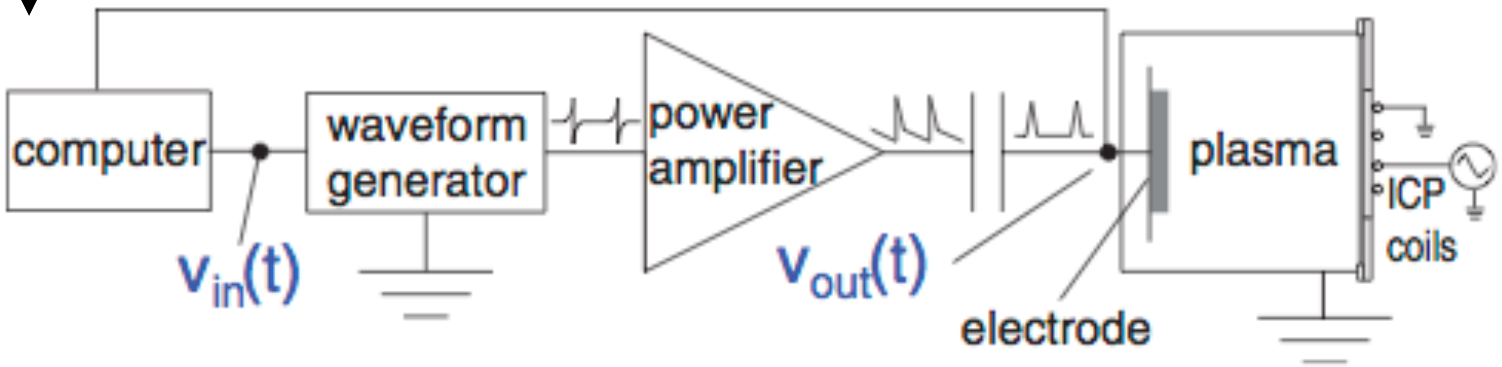
Kazuhide Ino, Toshikuni Shinohara, Takeo Ushiki, and Tadahiro Ohmi:
"Ion energy, ion flux, and ion species effects on crystallographic and electrical properties of sputter-deposited Ta thin films"
J. Vac. Sci. Technol. A 15, 2627 (1997)



- **Single-frequency discharges**
- **Dual-(Multiple-) frequency discharges**

- Hybrid discharges
  - inductive + capacitive
  - helicon + capacitive
  - DC + RF
- Customized waveforms

Wang S-B and Wendt A E 2000 J. Appl. Phys. 88 643;
Johnson E V, Verbeke T, Vanel J C, and Booth J P 2010 J. Phys. D: Appl. Phys. 43 412001
Rauf S and Kushner M J 1999 IEEE Trans. Plasma Sci. 27 1329;
Dudin S V, Zykov A V, Polozhii K I, and Farenik V I 1998 Tech. Phys. Lett. 24 881
Kawamura E, Lieberman M A, Lichtenberg A J, Hudson E A 2007 J. Vac. Sci. Technol. A 25 1456
Kawamura E, Lichtenberg A J, Lieberman M A 2008 Plasma Sources Sci. Technol. 17 045002
Jiang W, Xu X, Dai Z L, and Wang Y N 2008 Phys. Plasmas 15 033502
Patterson M M, Chu H-Y, and Wendt A E 2007 Plasma Sources Sci. Technol. 16 257
Novikova T. et al. THIS CONFERENCE



Tailoring the flux-energy distributions of ions reaching the electrodes (single freq.)

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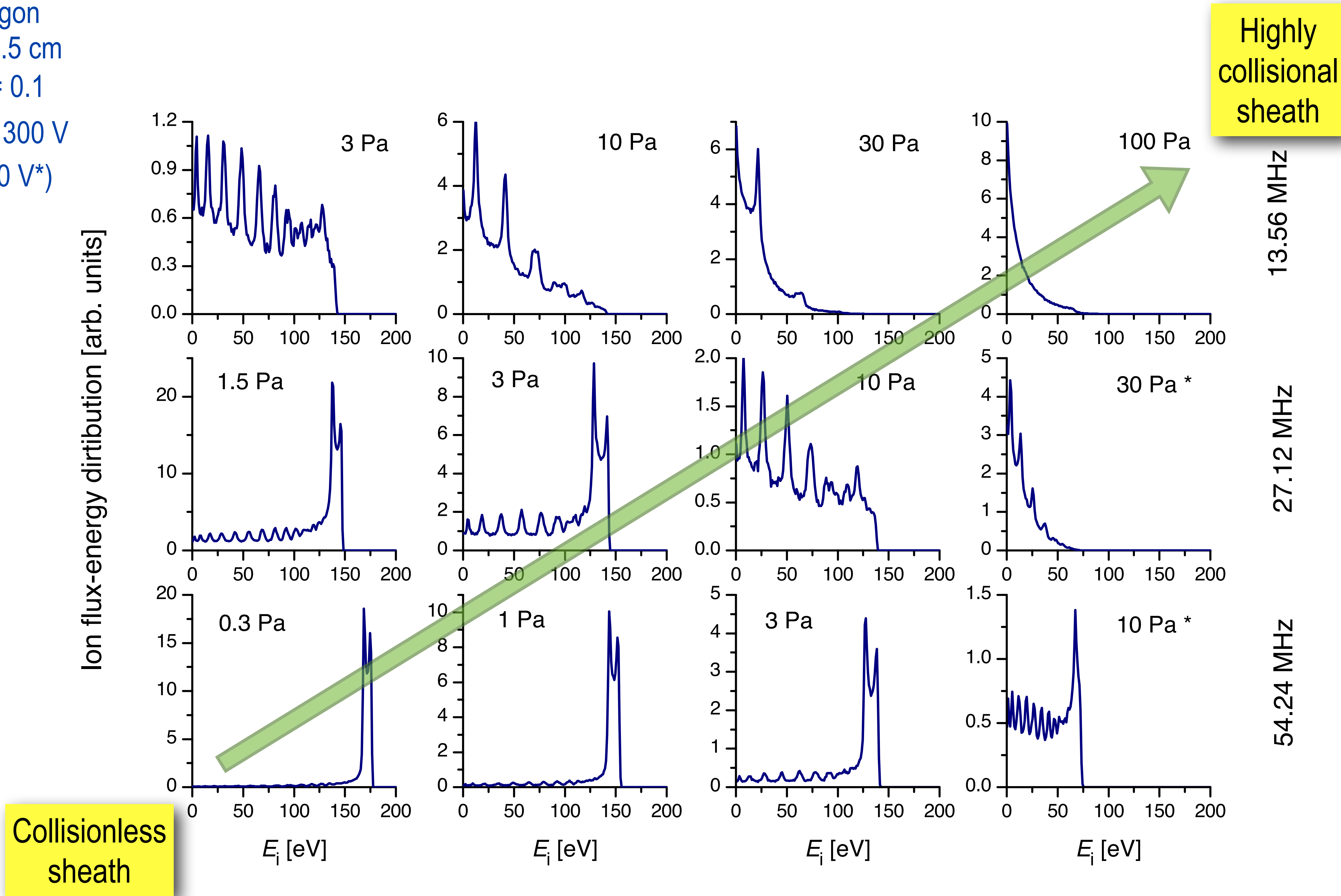
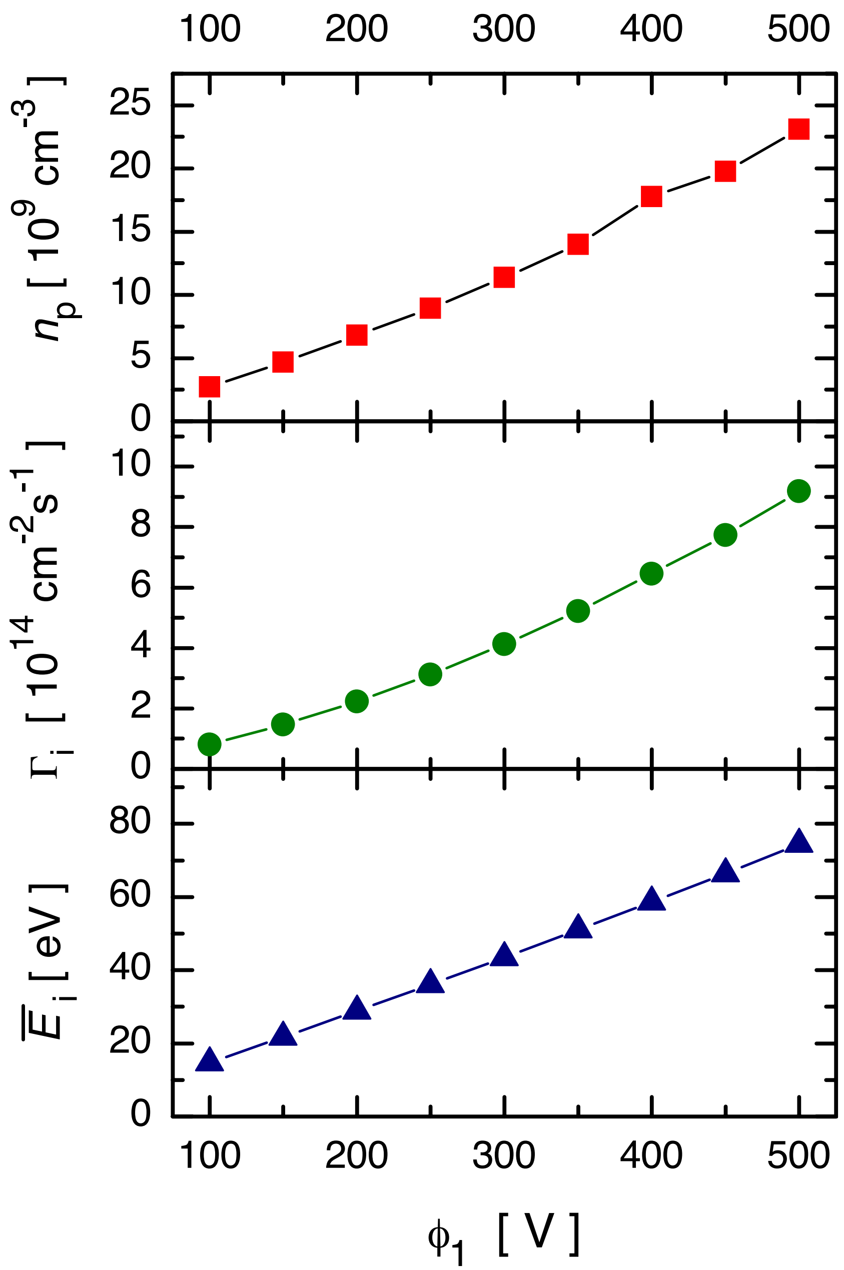
ICPP
2012

Argon
L= 2.5 cm
γ = 0.1
ϕ_1 = 300 V
(150 V*)



Limitations of single-frequency CCRF discharges

Single frequency discharge:
Argon @ 10 Pa
f = 13.56 MHz
L = 2.5 cm



The RF voltage influences both the ion flux and mean ion energy

Independent control ??

Dual frequency excitation

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

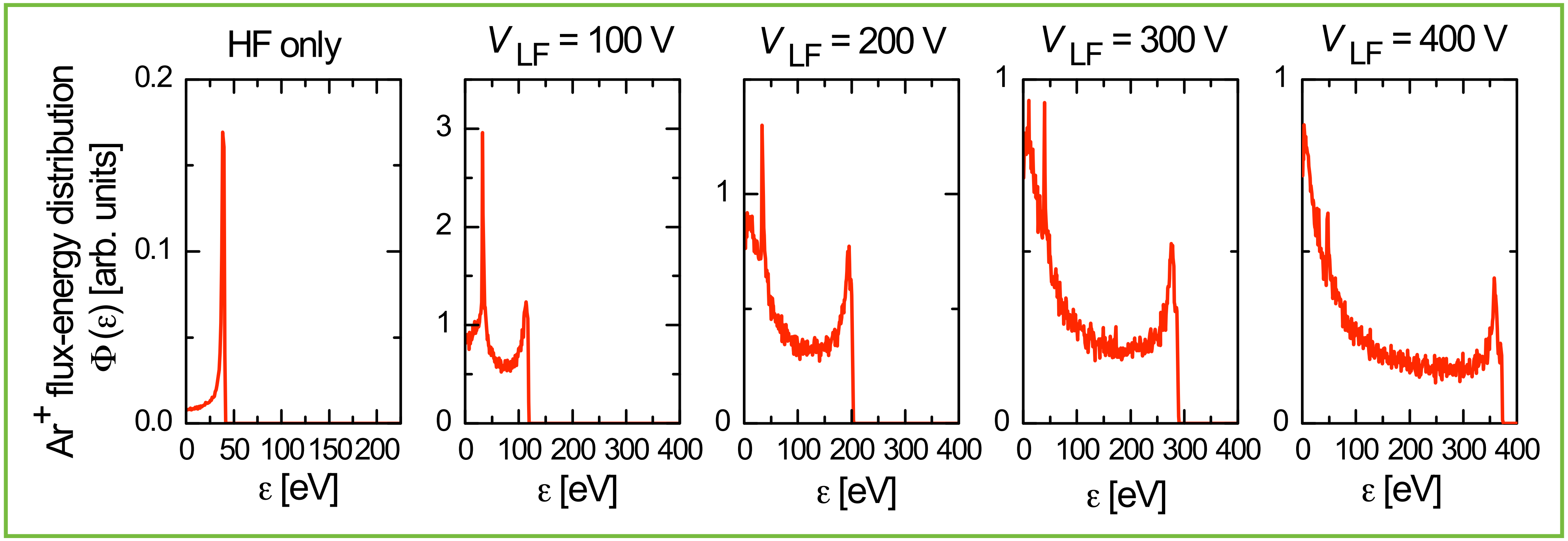
Heating & ion flux | **Ion energy**

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
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
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

Control of ion properties in classical dual-frequency discharges

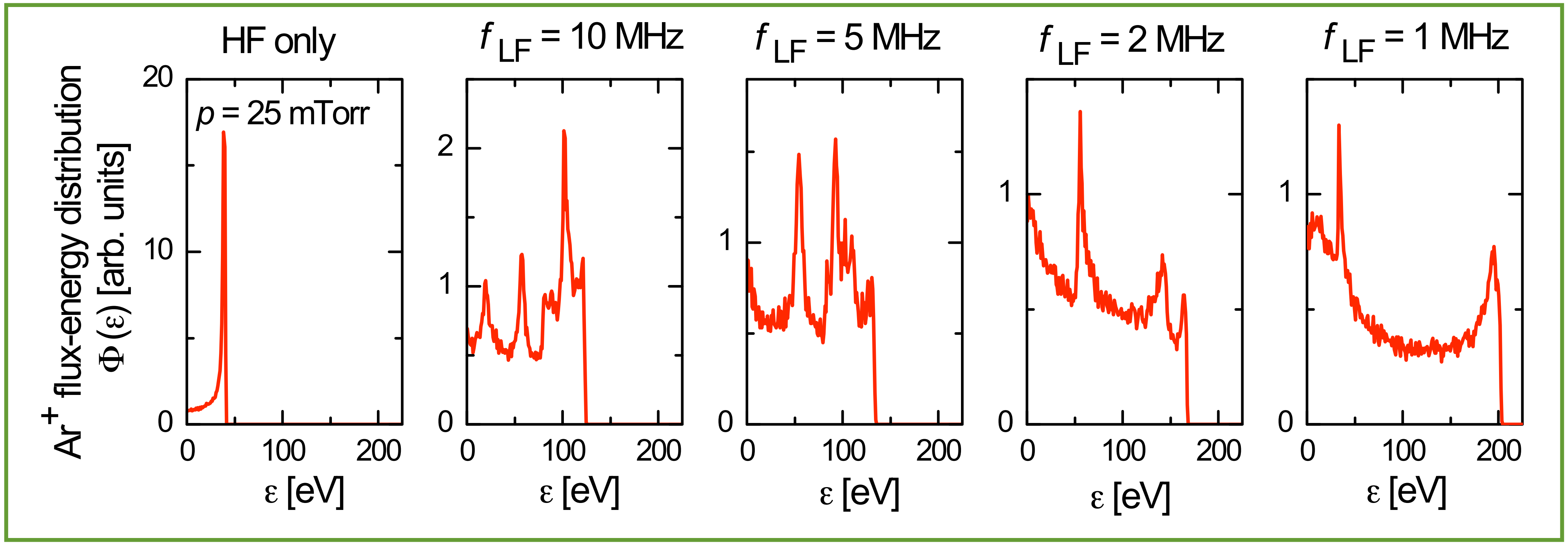
- Ar discharge
- p = 25 mTorr
- L = 2 cm
- V_{HF} = 60 V
- f_{HF} =100 MHz

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



$\gamma = 0$

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

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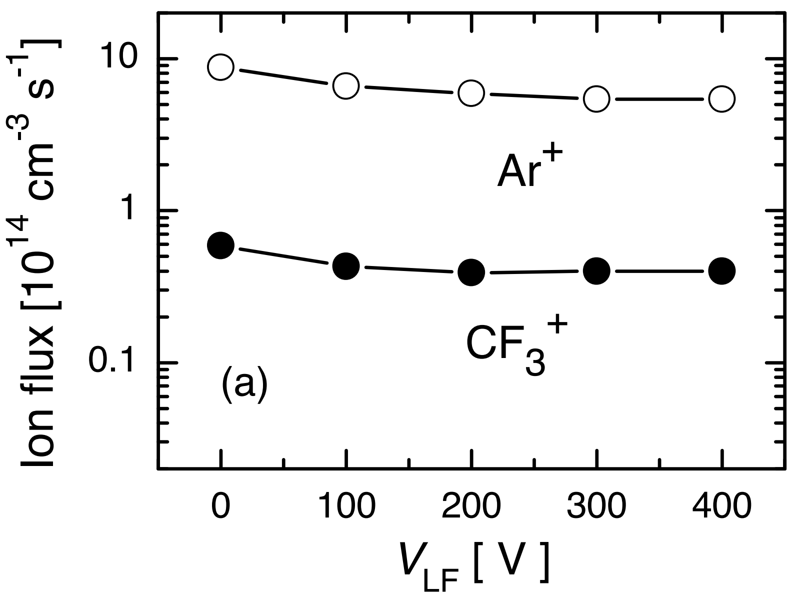
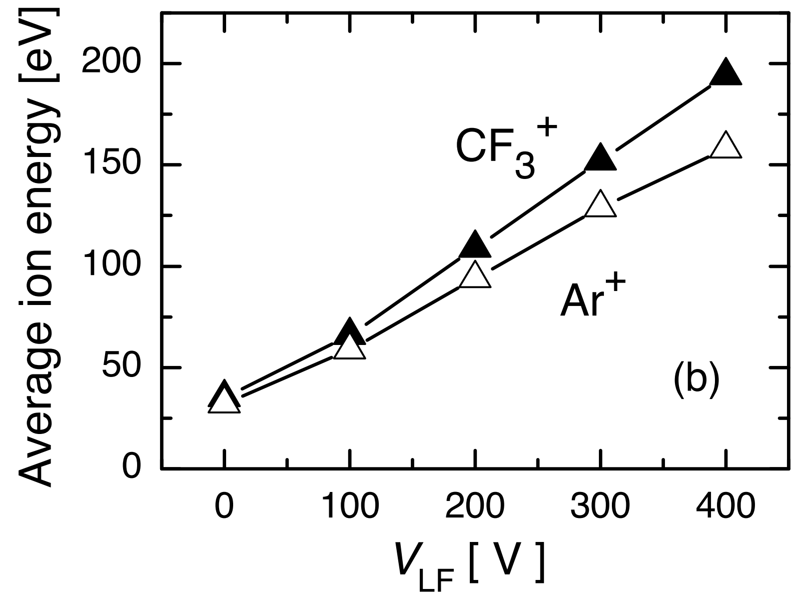
$\gamma = 0$

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

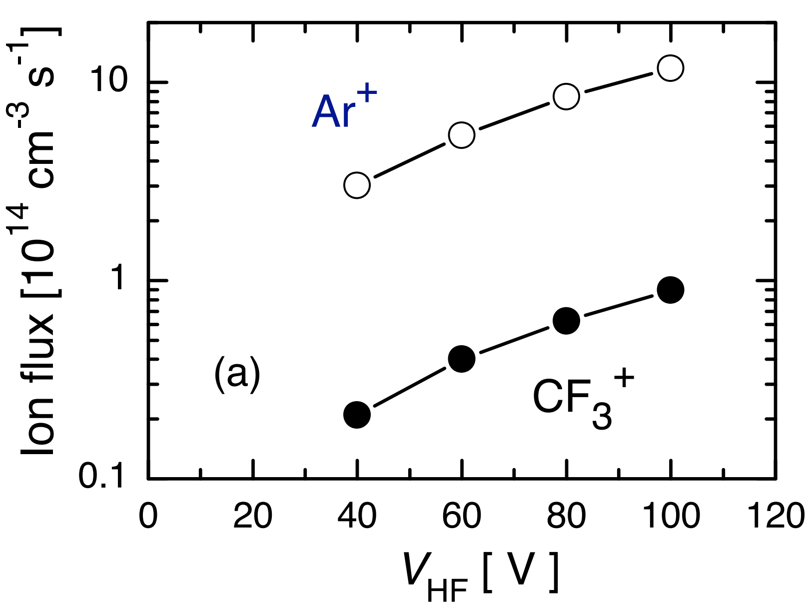
Effect of V_{LF} @ V_{HF}=60 V

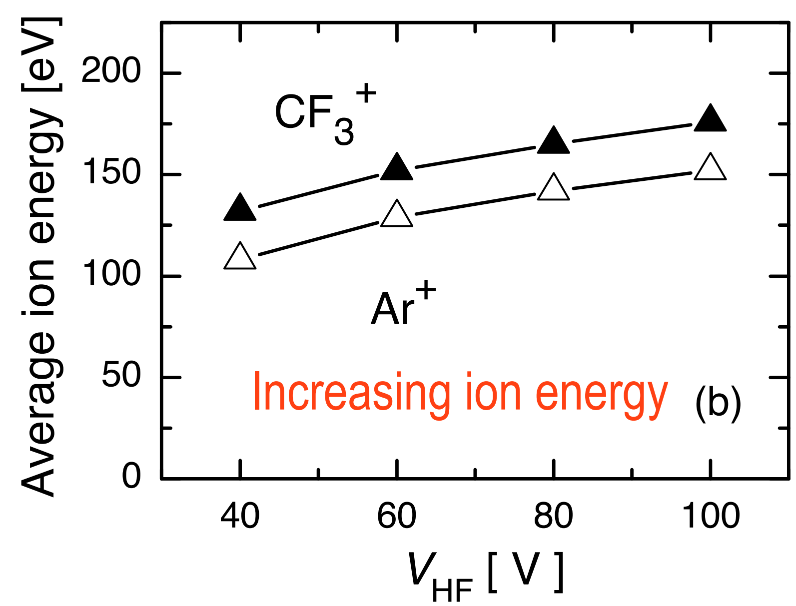
Decreasing ion fluxes





Effect of V_{HF} @ V_{LF}=300 V





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}

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

Control of ion properties in classical dual-frequency discharges

Other studies:

P. C. Boyle, A. R. Ellingboe and M. M. Turner, J. Phys. D 37, 697 (2004)
Ar, $\gamma = 0$
Flux remains nearly constant

V. Georgieva and A. Bogaerts, J. Appl. Phys. 98, 023308 (2005)
$Ar / CF_4 / N_2$
Complicated behavior depending on driving frequencies

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 <u>increase</u> with increasing low-frequency power

The somewhat contradicting results motivated a systematic study, covering a wide range of discharge conditions and **secondary yield** values

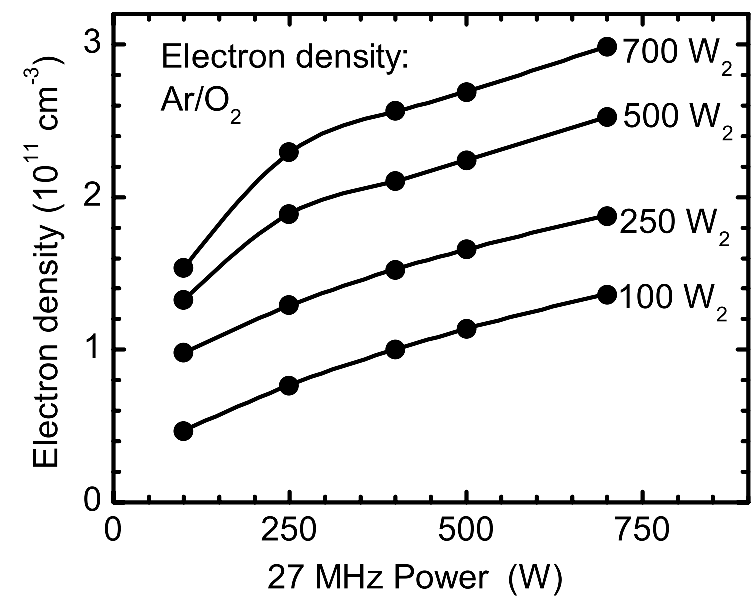


Figure 1. Electron density in an Ar/O_2 plasma as a function of 2 and 27 MHz power.

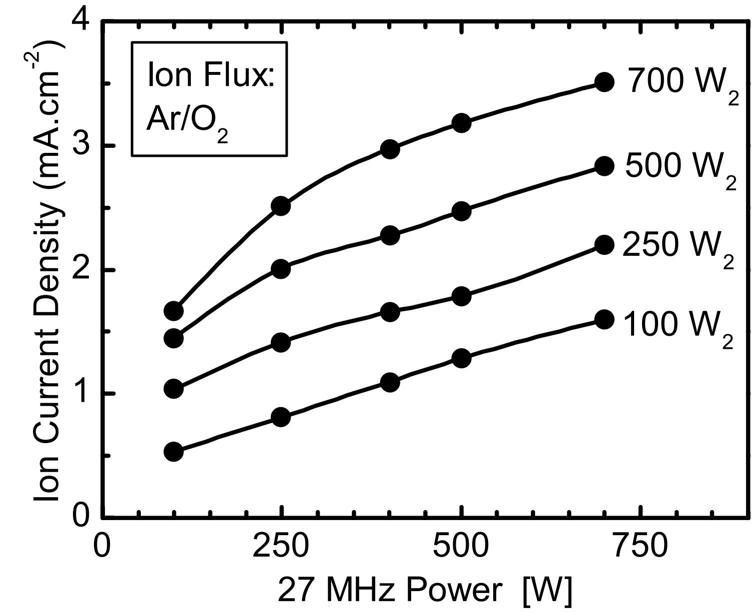
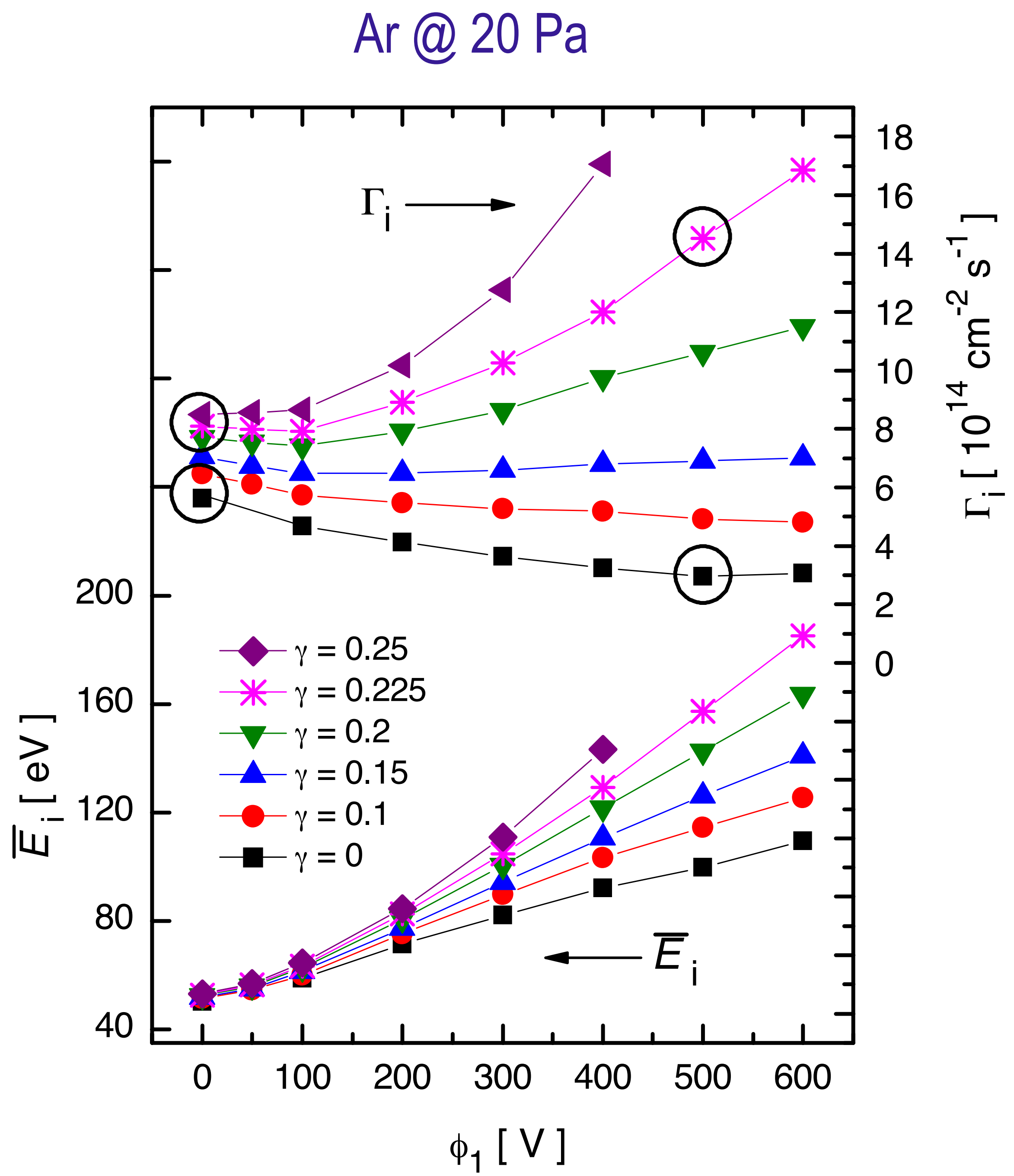


Figure 2. Ion flux in an Ar/O_2 plasma as a function of 2 and 27 MHz power.

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

EPS ICPP 2012



- 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

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

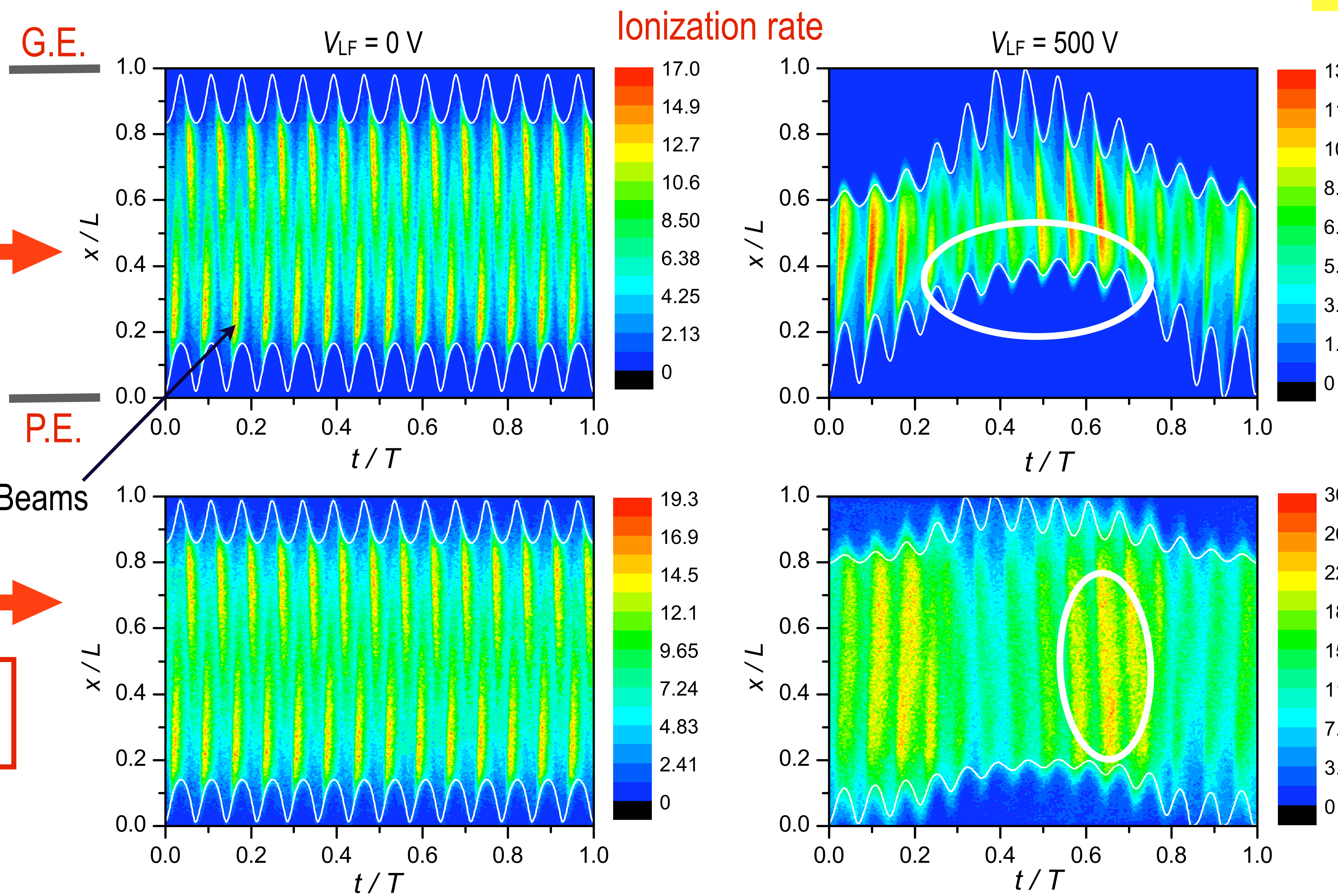
Z. Donkó, et al., Appl. Phys. Lett. 97, 081501 (2010).

$\gamma = 0$

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

$\gamma = 0.225$

FREQUENCY COUPLING



See also:

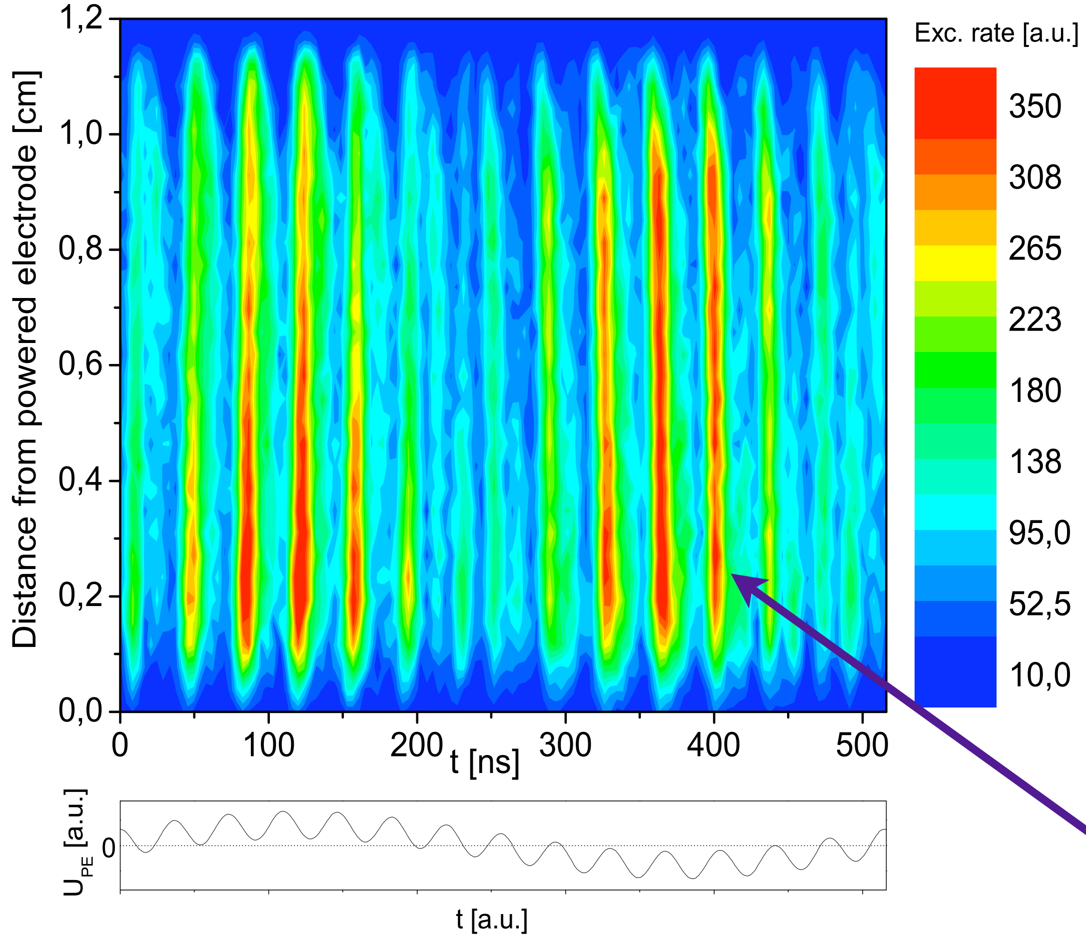
M. M. Turner and P. Chabert, Phys. Rev. Lett. **96** 205001 (2006)
T. Gans, J. Schulze, D. O'Connell, U. Czarnetzki, R. Faulkner, A. R. Ellingboe and M. M. Turner, Appl. Phys. Lett. **89** 261502 (2006)
J. Schulze, Z. Donko, D. Luggenhölscher and U. Czarnetzki, PSST **18**, 034011 (2009)

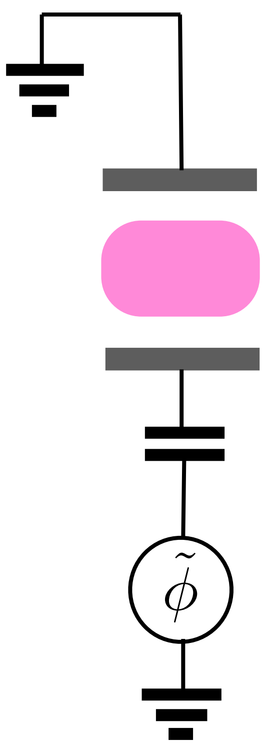
Electron beams launched at sheath expansion

EPS
ICPP
2012

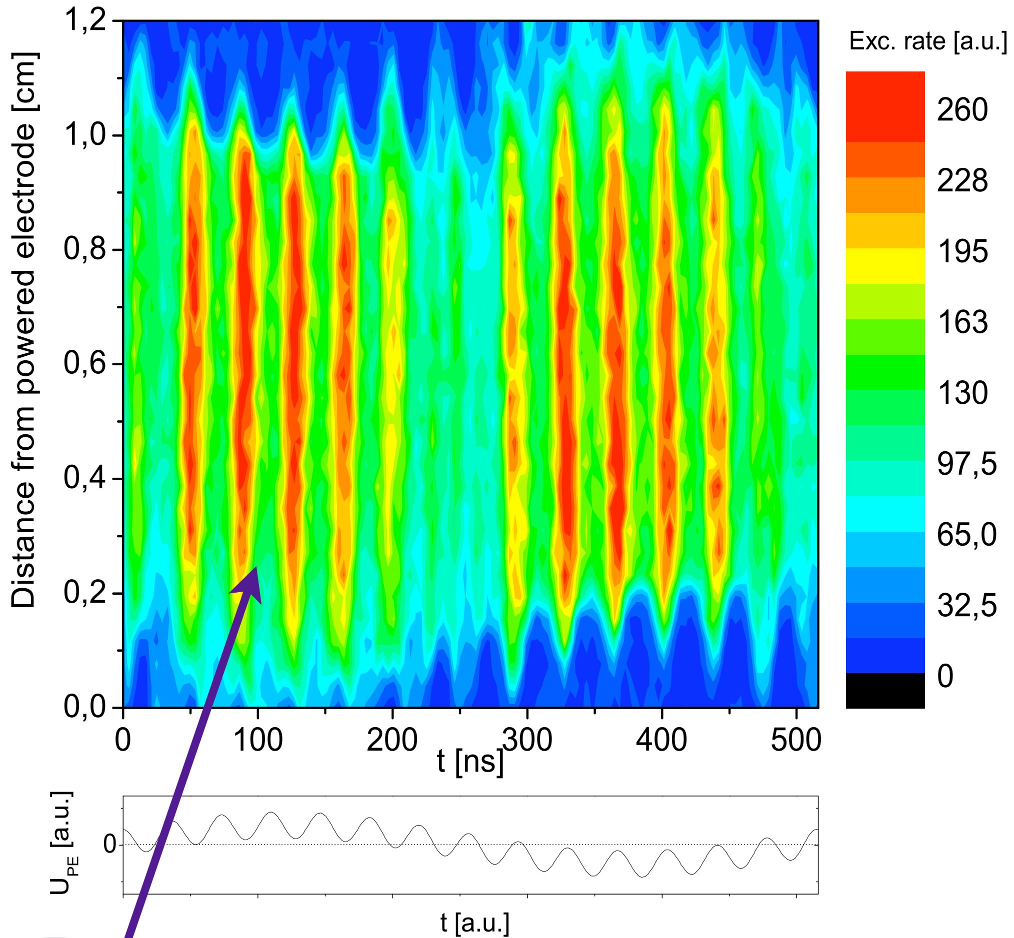
Spatio-temporal distribution of electron impact excitation

Experiment (Bochum/Dublin)





Simulation (Budapest)



Electron beams

He @ 65 Pa
f_{HF} = 27.118 MHz
f_{LF} = 1.937 MHz (= f_{HF}/14)

J. Schulze, T. Gans, D. O'Connell, U. Czarnetzki, A. R. Ellingboe, and M. M. Turner, J. Phys. D **40** 7008 (2007).

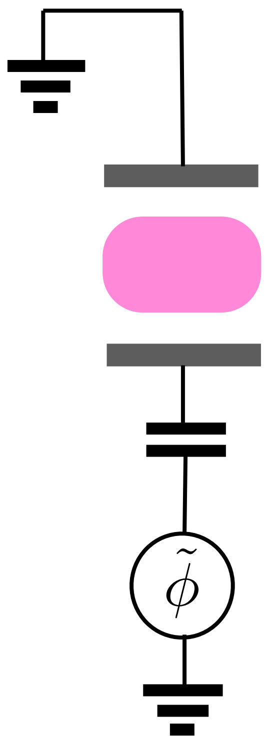
J. Schulze, Z. Donkó, D. Luggenhölscher and U. Czarnetzki, Plasma Sources Sci. Technol. **18**, 034011 (2009).

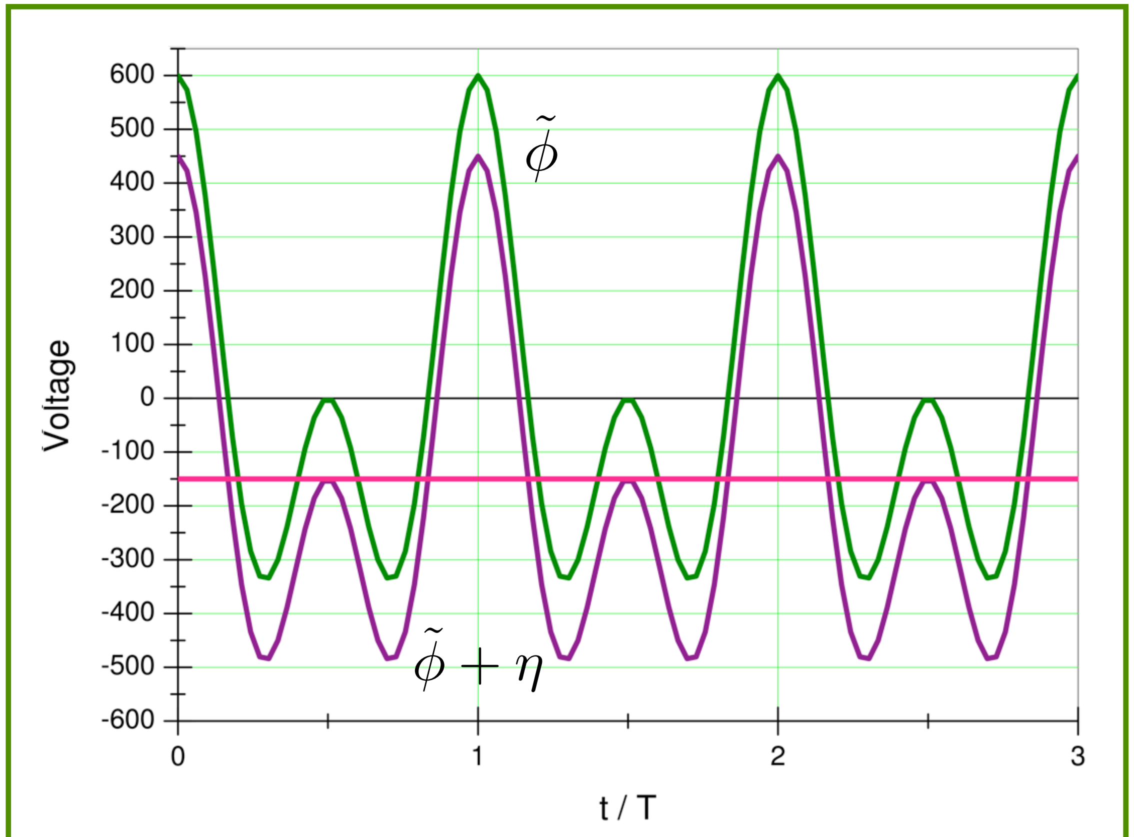
The Electrical Asymmetry Effect

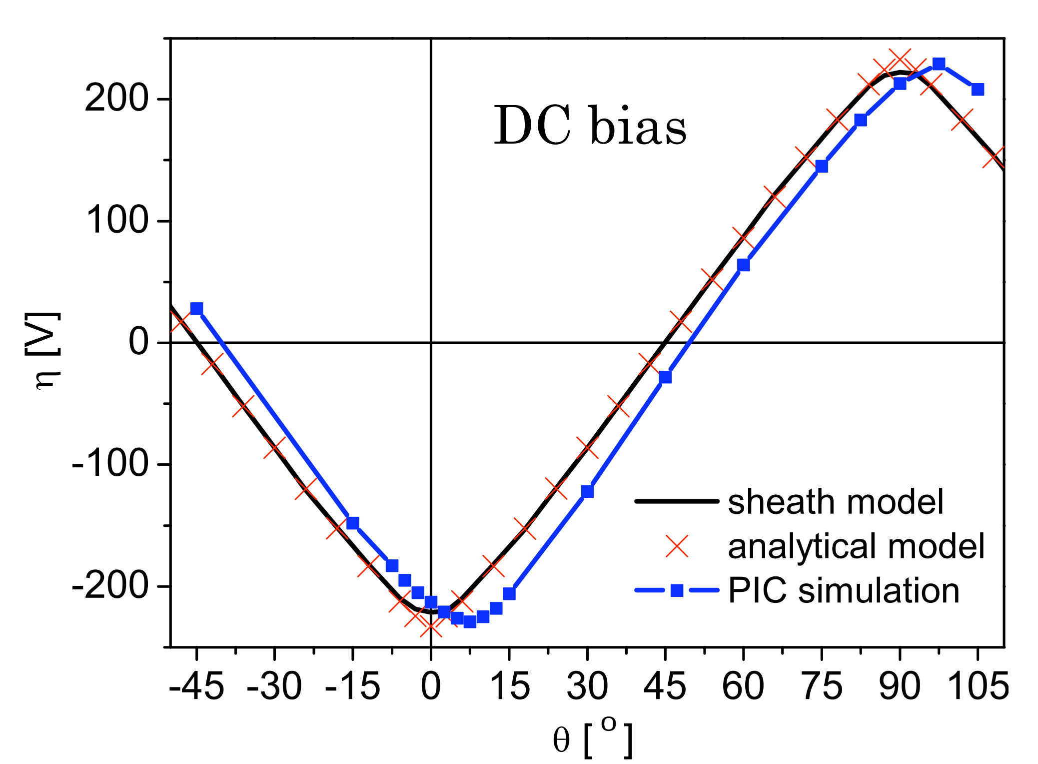
(a novel approach to control ion properties)

The Electrical Asymmetry Effect

B. G. Heil, J. Schulze, T. Mussenbrock, R. P. Brinkmann and U. Czarnetzki, IEEE Trans. on Plasma Sci. 36 1404 (2008)
B. G. Heil, U. Czarnetzki, R. P. Brinkmann and T. Mussenbrock, J. Phys. D 41 165202 (2008)







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

↑ control parameter

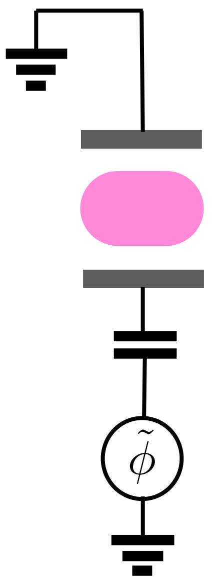
f = 13.56 MHz & 27.12 MHz,
p = 20 mTorr, d = 6.7 cm
V_0 = 315 V (both freq. !!)

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)

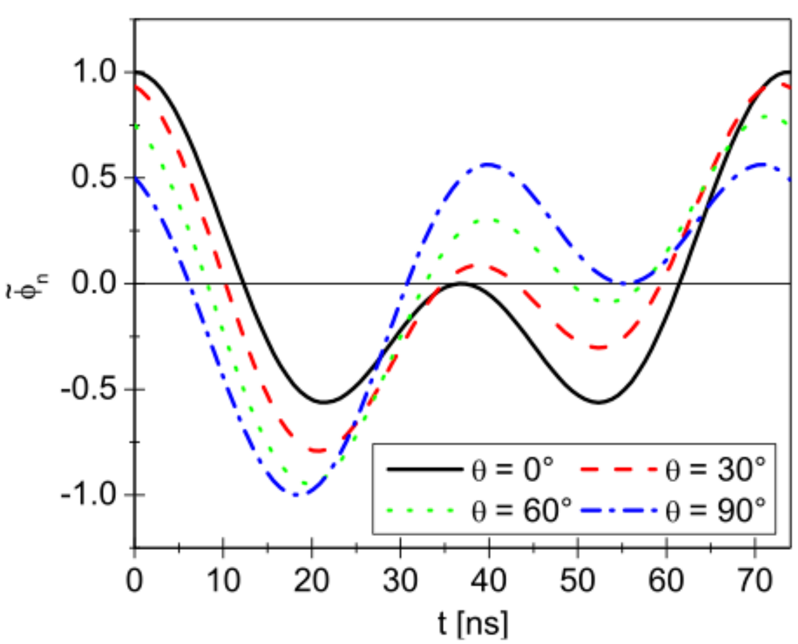
EAE: Control of electron dynamics / excitation / ionization

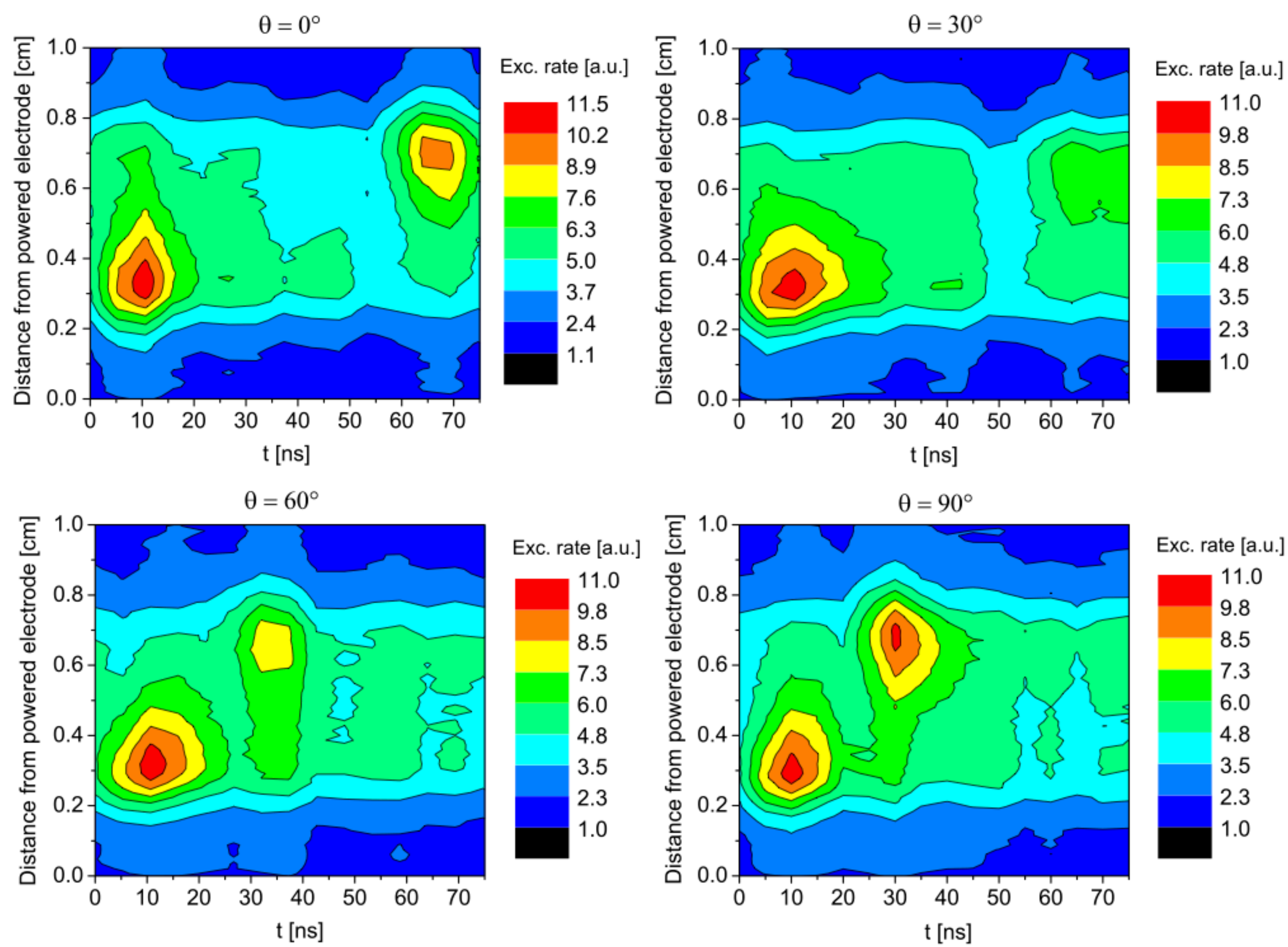
PROES results: Spatio-temporal distribution of the excitation rate at different phase angles



Ar (10% Ne) @ 60 Pa
L = 1 cm
V_0 = 76 V

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





J. Schulze, E. Schüngel, Z. Donkó and U. Czarnetzki, Plasma Sources Sci. Technol. 19 045028 (2010).

The sheath dynamics and, consequently, the excitation dynamics change as a function of θ

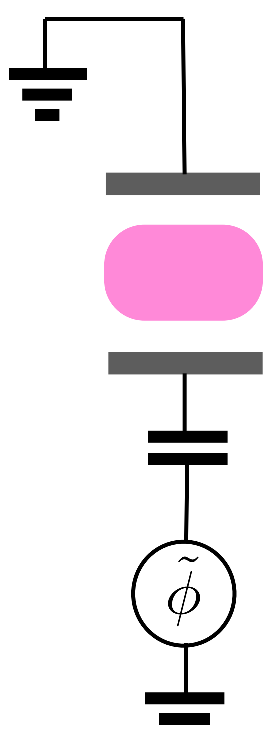
EAE: Control of ion properties

Flux-energy distribution of ions at the powered and grounded electrodes

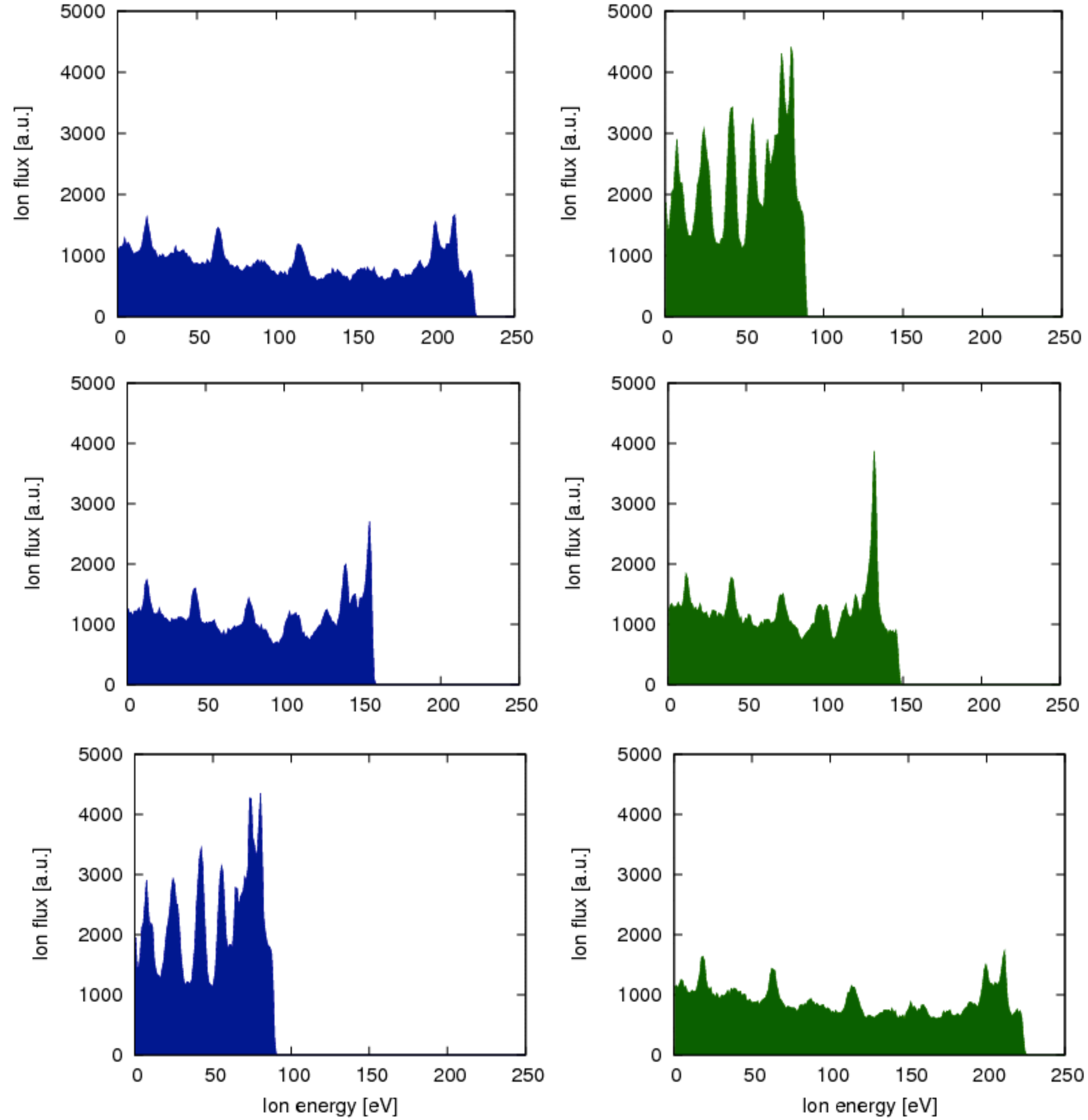
PIC / MCC results

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

Ion flux nearly constant



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



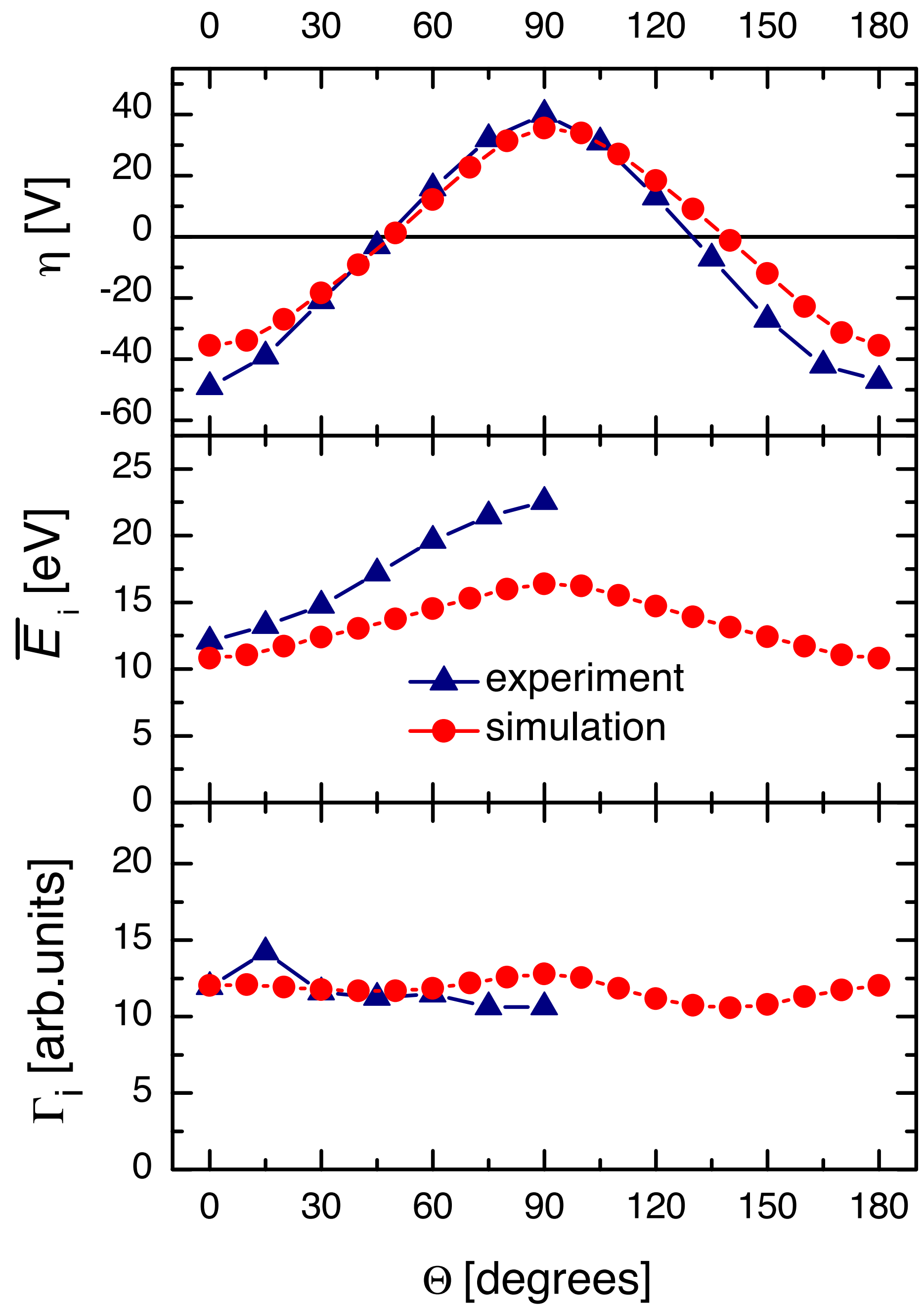
phase= 0 deg.

45 deg.

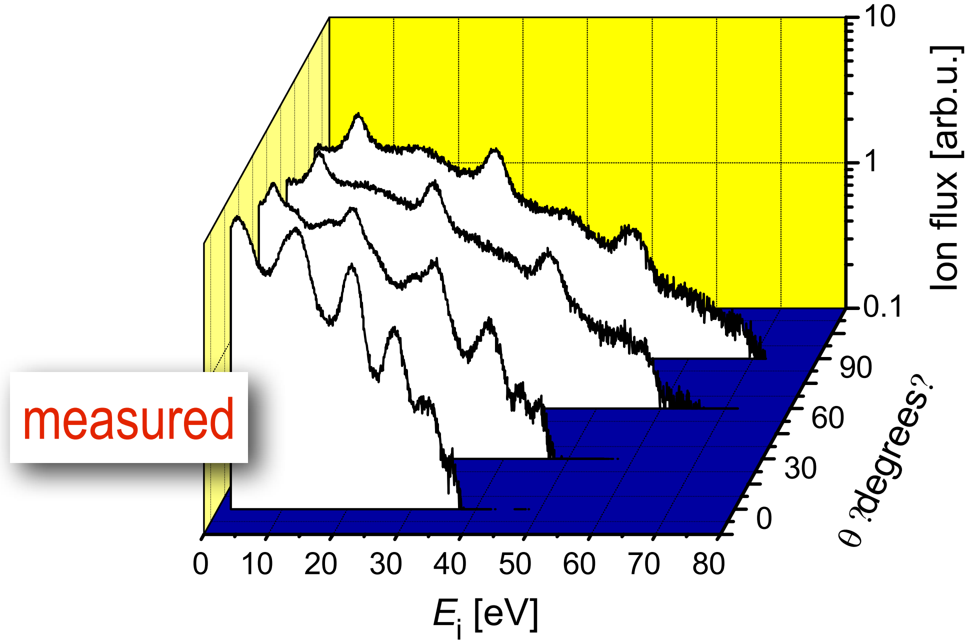
90 deg.

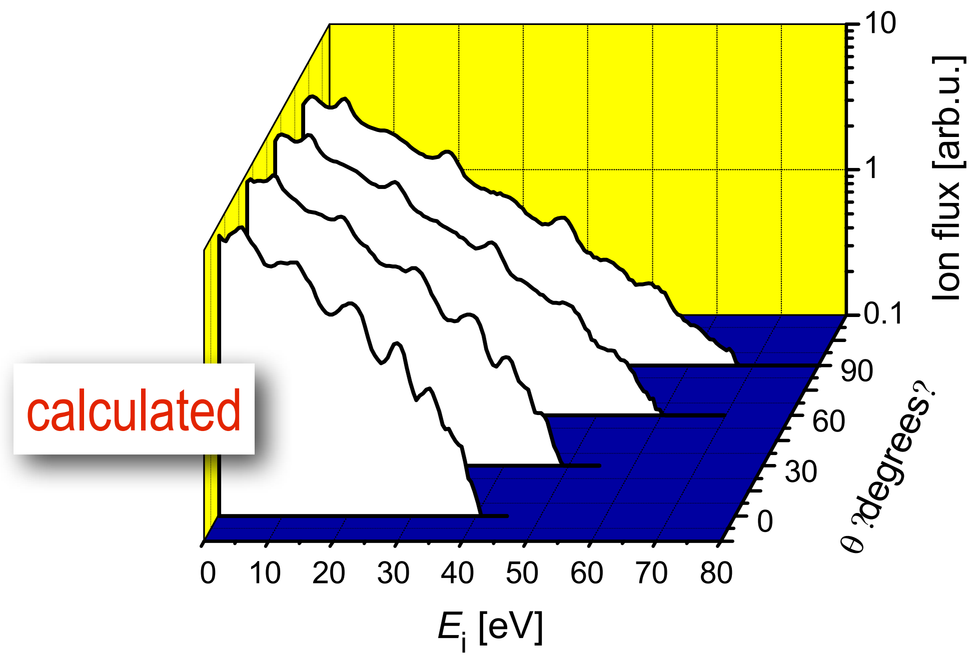
EAE operation: Experiments vs. simulations

Performance of the EAE



Ion flux-energy distributions





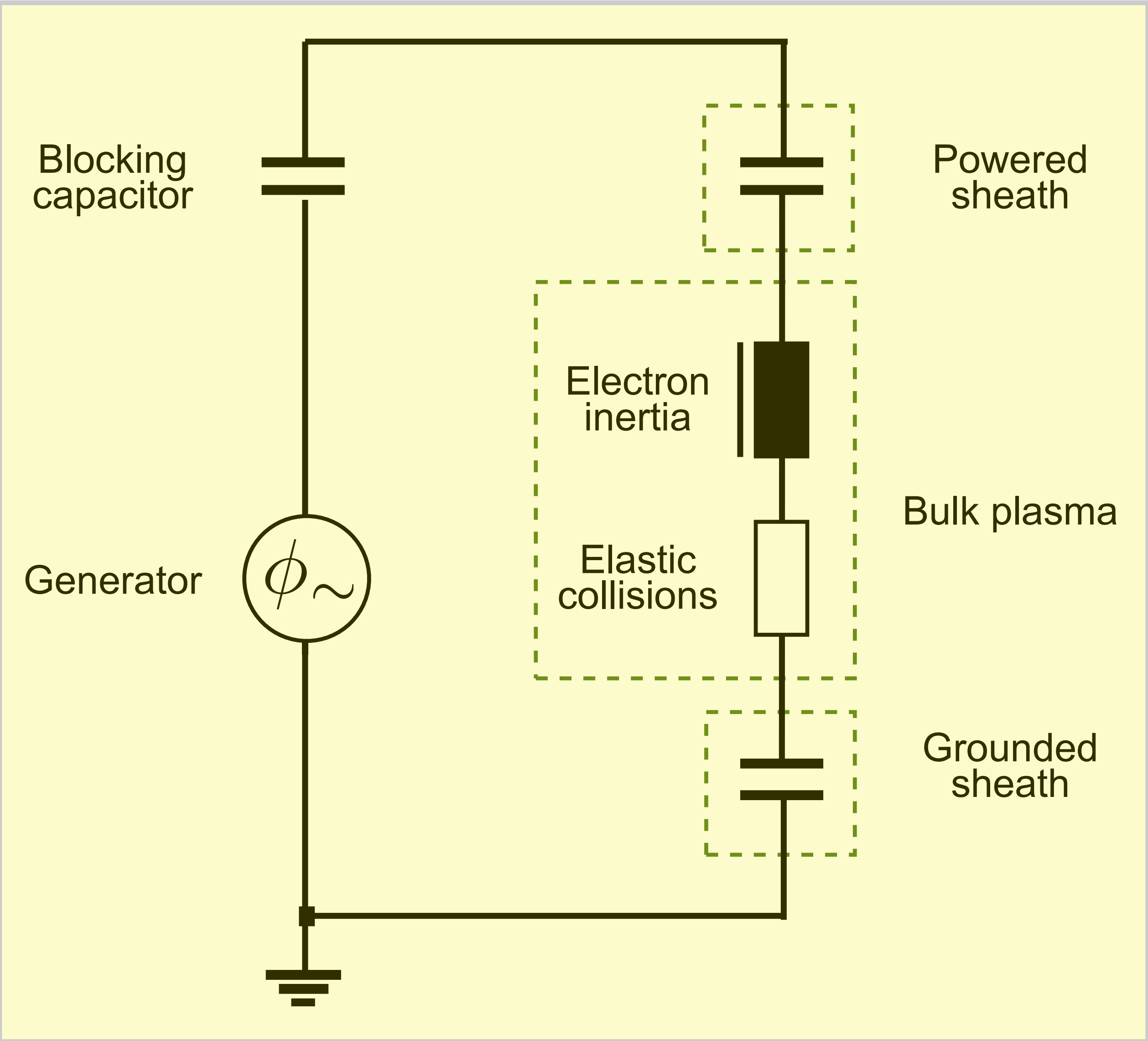
Ar discharge
p = 20 Pa
L = 1 cm
f = 13.56 MHz
V_0 = 76 V

J. Schulze, Z. Donkó, E. Schüngel and U Czarnetzki: Plasma Sources Sci. Technol. 20, 045007 (2011).

The Plasma Series Resonance

(a nonlinear effect in asymmetric RF discharges)

The Plasma Series Resonance



U. Czarnetzki, T. Mussenbrock and R. P. Brinkmann, Phys. Plasmas 13, 123503 (2006).
T. Mussenbrock, R. P. Brinkmann, M. A. Lieberman, A. J. Lichtenberg, and E. Kawamura, Phys. Rev. Lett. 101 085004 (2008).

Voltage balance expressed in terms of the normalized charge in the powered sheath describes the dynamics of the discharge

q: normalized charge in the powered sheath

- If $\varepsilon = 1$ and the bulk voltage is small, the nonlinearity disappears - PSR oscillations show up in asymmetric discharges only
- However, EAE can create asymmetry and PSR oscillations

PSR equation:

$$\bar{\phi}_{\sim}(t) + \bar{\eta} = -q^2(t) + \varepsilon[q_{\mathrm{t}} - q(t)]^2 - 2\beta^2\left[\frac{\partial^2}{\partial t^2} + \nu\frac{\partial}{\partial t}\right] q(t)$$

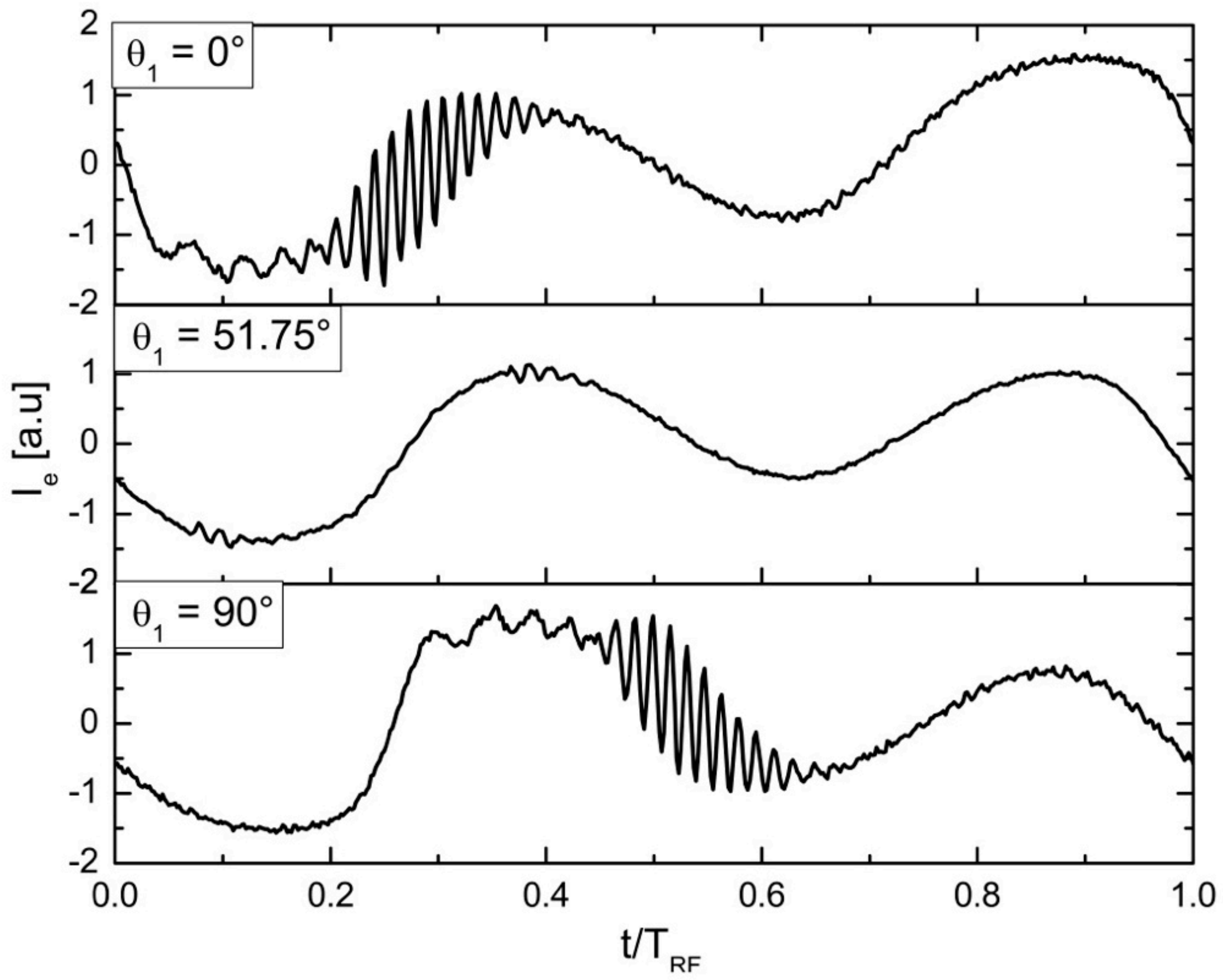
Plasma Series Resonance

Observation of PSR oscillations in **electrically asymmetric** discharges

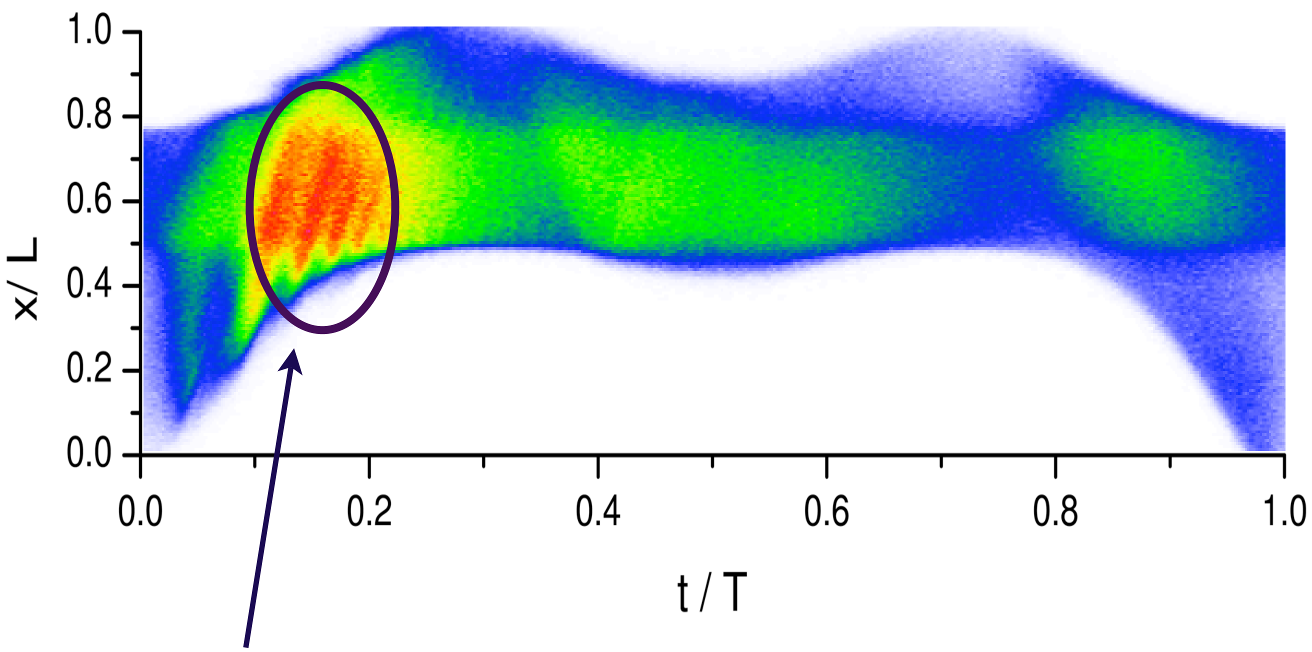
$$\tilde{\phi} = V_0[\cos(\omega t + \theta) + \cos(2\omega t)]$$

V_0 = 800 V, p = 3 Pa

Time dependence of the electron current:



Spatio-temporal distribution of electron impact excitation rate:



Multiple electron beams with a speed of ≈ 3×10^6 m/s.

U. Czarnetzki, J. Schulze , E. Schüngel and Z. Donkó, Plasma Sources Sci. Technol. 20, 024010 (2011)

Summary

- Why capacitive radiofrequency discharges?
- Methods (models, simulation technique, diagnostics)
- Different modes of electron heating
- Control of ion properties:
  - “Classical” dual-frequency discharges
  - Effects of *secondary electrons* in dual-frequency discharges
  - The Electrical Asymmetry Effect
- The Plasma Series Resonance effect

Thanks

- Hungarian Fund for Scientific Research - OTKA
- Collaborators
- All of you for your attention