FAST ANALYTICAL/NUMERICAL MODEL OF ATMOSPHERIC PRESSURE RADIO-FREQUENCY CAPACITIVE DISCHARGES

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OUTLINE

- Motivation, scope and goal
- Electron energy probability function (EEPF)
  in atmospheric pressure capacitive rf discharges
- Fast analytical/numerical model formulation
- Brief summary of results

MOTIVATION

- Biomedical — example of reactive oxygen species

  (Review article: H.W. Lee et al, J. Phys. D **44**, 053001, 2011)

  — Applications to sterilization, cancer cell treatment, blood coagulation, wound healing

- Unique materials — example of anatase crystalline TiO_2

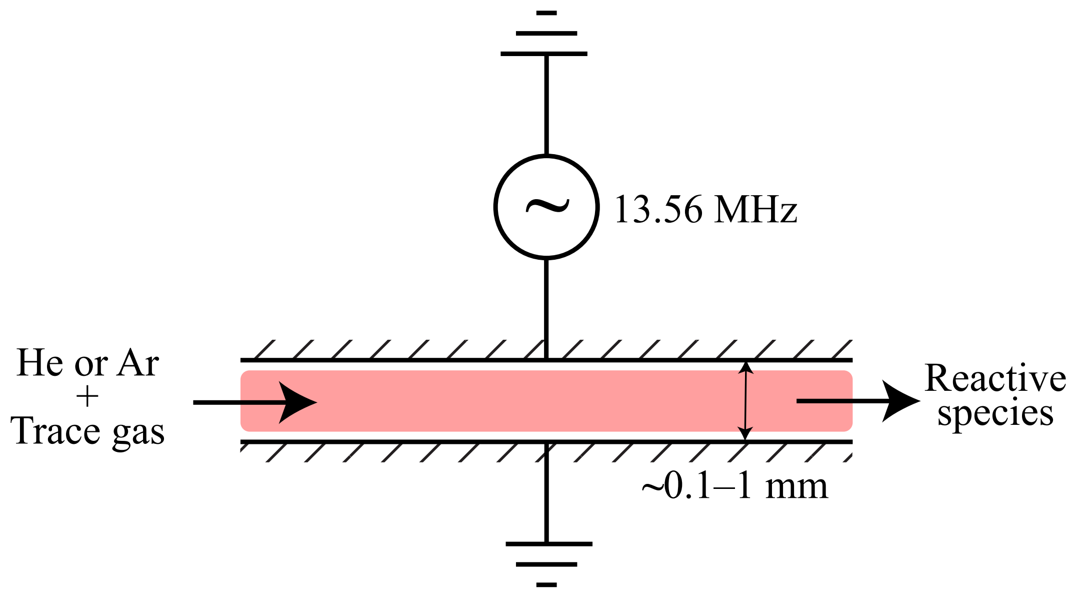
  (Review article: D. Mariotti and R.M. Sankaran, J. Phys. D, 323001, 2010)

  (Anatase TiO_2: H.G. Yang et al, Nature **453**, 638, 2008)

  — Applications to photonics crystals, photo/electrochromic devices, gas sensors, spintronic devices, anticancer or gene therapies, solar cells for electric energy or hydrogen production

SCOPE

- Atmospheric pressure
- He or Ar with trace reactive gases
- 1D plane-parallel geometry (~0.1–1 mm gap)
- RF-driven (~13.56 MHz)



GOAL

- Fluid models
  — Space variations (sheaths and bulk plasma resolved)
  — Time variations (rf timescale resolved)
  — Simulations are slow
- Global models
  — No consideration of space and time variations
  — Simulations are fast
- Incorporate space and time variations into a fast global model
- The main issue is the time-variation of the EEPF

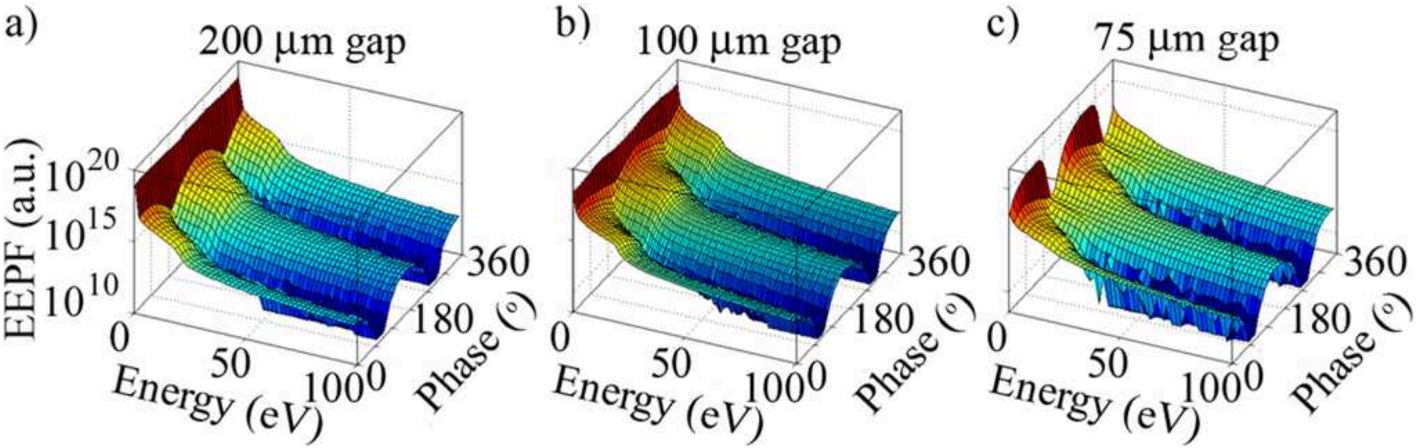
  — At low pressures $\tau_\epsilon \gg \tau_{\rm rf}$ (τ_ϵ = electron energy relaxation time)
  $\Longrightarrow$ EEPF varies only weakly with time

  — At atmospheric pressure $\tau_\epsilon \lesssim \tau_{\rm rf}$
  $\Longrightarrow$ EEPF is strongly modulated in time

EEPF's IN ATMOSPHERIC PRESSURE RF CAPACITIVE DISCHARGES

EEPF TIME VARIATIONS — He 1D PIC SIMULATION

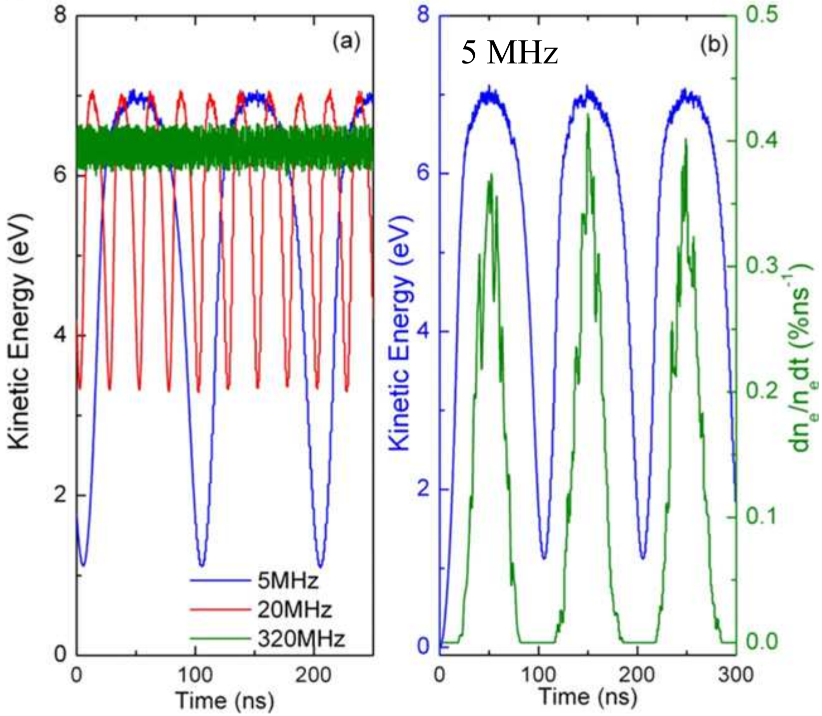
- At high pressures, $\tau_\epsilon \ll \tau_{\rm rf}$; the EEPF varies strongly with time



(F. Iza et al, Phys. Rev. Lett. **99**, 075004, 2007)

ELECTRON KINETIC ENERGY TIME VARIATIONS

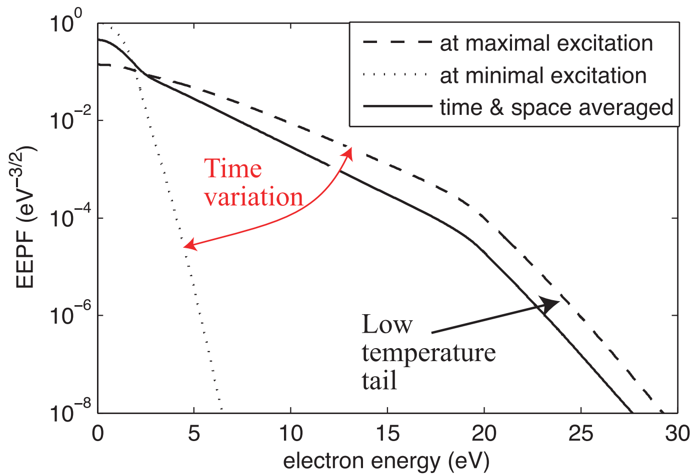
- He 1D PIC simulation at various frequencies



(J.L. Walsh et al, Appl. Phys. Lett. **93**, 221505, 2010)

EEPF TIME VARIATIONS

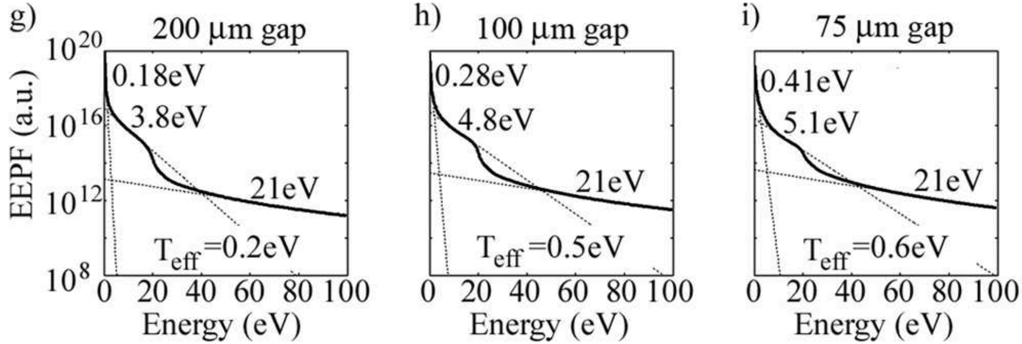
- He/N2 fluid simulation with kinetic (Bolsig+) EEPF calculation



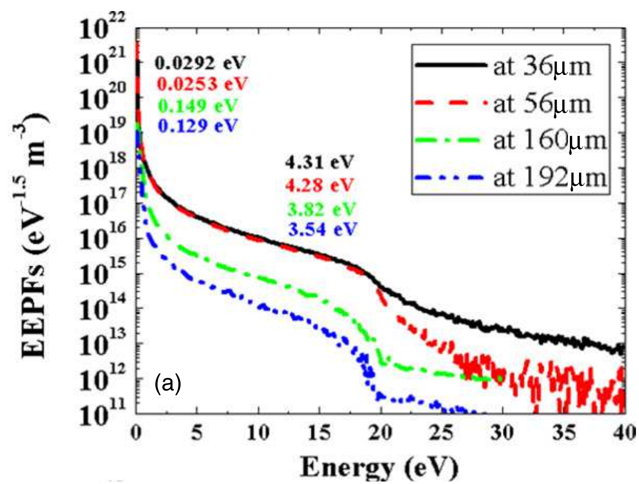
(J. Waskoenig, PhD Thesis, Queens U Belfast, 2010)

- Conclusions used in modeling
  — The EEPF oscillates in time with the rf electron power absorbed
  — The EEPF is Maxwellian below a break energy $\mathcal{E}_b \approx 20$ V (metastable He excitation energy)
  — The EEPF has a low temperature tail above the break energy

TIME-AVERAGE EEPF — PIC SIMULATIONS



(F. Iza et al, Phys. Rev. Lett. **99**, 075004, 2007)

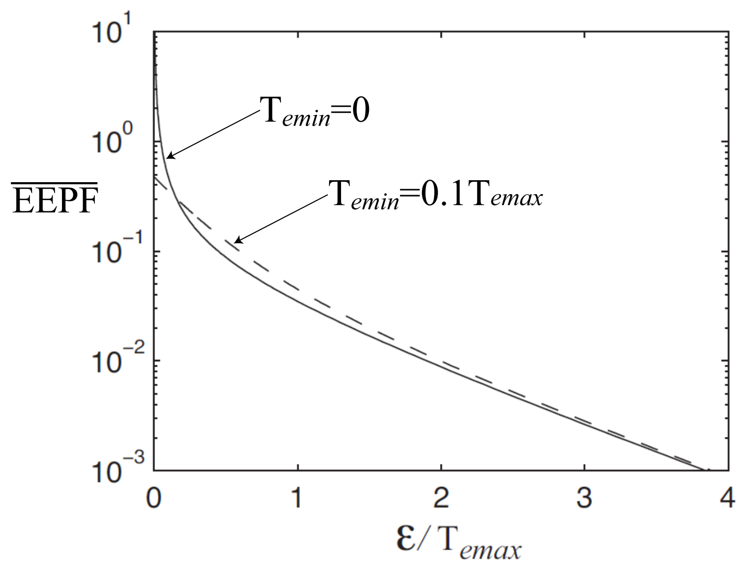


(Y.J. Hong et al, J. Phys. D **41**, 245208, 2008)

TIME-AVERAGE EEPF — ANALYTICS

- The time average of a Maxwellian distribution with time-varying T_e is non-Maxwellian, with an enhanced low-temperature tail
- For $\mathrm{T}_{emin} \to 0$

$$\overline{\mathrm{EEPF}} \approx \frac{4}{3\,\mathrm{T}_{emax}^{3/2}}\left(1 + \frac{\mathrm{T}_{emax}}{3\,\mathcal{E}}\right)\mathrm{erfc}\left(\sqrt{\mathcal{E}/\mathrm{T}_{emax}}\right)$$

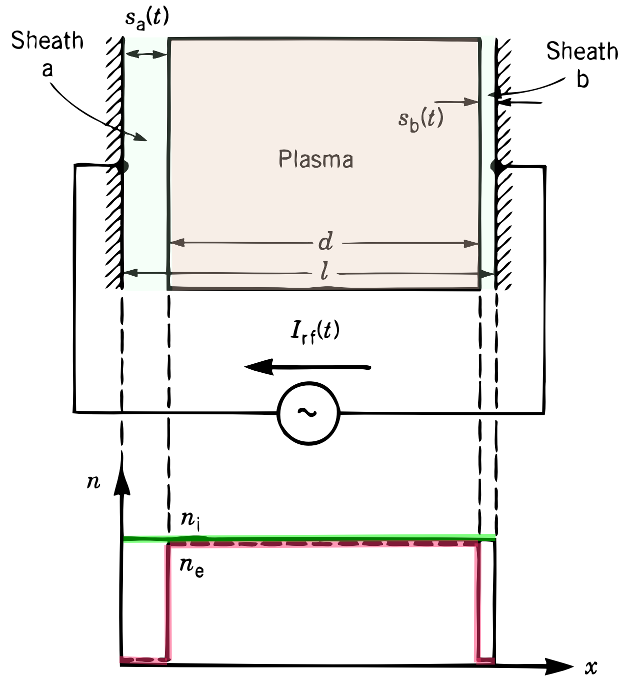


FAST ANALYTICAL/NUMERICAL MODEL FORMULATION

HOMOGENEOUS DISCHARGE MODEL

- Uniform density ions with rf oscillation of electron cloud

  $\Longrightarrow$ analytical expression for electron power absorbed $P_e(t)$



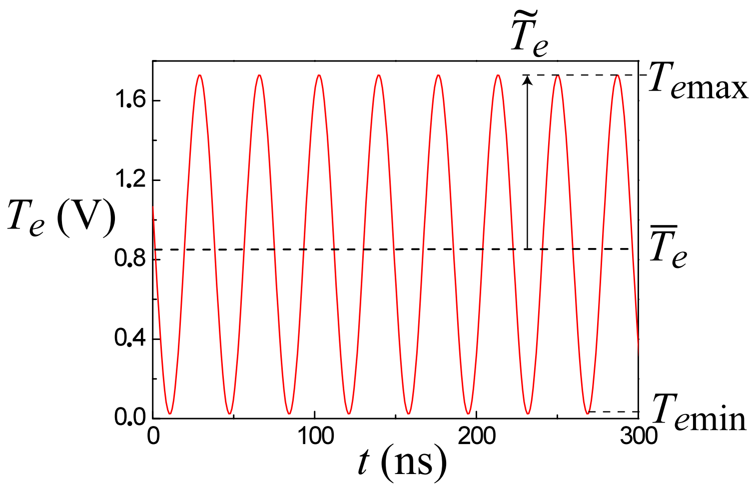
(V.A. Godyak, Sov. Phys. Tech. Phys. **16**, 1073, 1972)

ELECTRON ENERGY BALANCE

$$\frac{3}{2} e n_e \frac{\mathrm{dT}_e}{\mathrm{d}t} = P_e(t) - \underbrace{e n_e \frac{3m}{M} \nu \mathrm{T}_e}_{\text{Elastic loss}} - \underbrace{\sum_j e n_e \nu_j \mathcal{E}_j}_{\text{Inelastic loss (small)}}$$

$\Longrightarrow$ analytical expression for $T_e(t)$

$$\mathrm{T}_e(t) = \overline{T}_e + \widetilde{T}_e \cos(2\omega t + \phi_0)$$



EFFECTIVE RATE COEFFICIENTS

- Electron-activated processes strongly affected by $T_e(t)$
- Maxwellian rate coefficient

$$K = K_0 \exp(-\mathcal{E}_a/\mathrm{T}_e)$$

- Average over oscillating temperature

$\Longrightarrow$ Effective rate coefficient

$$\overline{K} = K_0(2\widetilde{\mathrm{T}}_e)\,\mathrm{erfc}\left(\sqrt{\mathcal{E}_a/2\widetilde{\mathrm{T}}_e}\right)$$

for energies below the break (He metastable) energy $\mathcal{E}_b$

- Effective rate coefficient $\overline{K} = \overline{K\big(\mathrm{EEPF}(t)\big)} \neq K\big(\,\overline{\mathrm{EEPF}(t)}\,\big)$

RATE COEFFICIENTS ABOVE THE BREAK ENERGY

- Above the He metastable energy $\mathcal{E}_b$, the tail temperature is $\mathrm{T}_c < \mathrm{T}_e$
- T_c is found analytically from kinetic theory

$$\mathrm{T}_c = \frac{E_\mathrm{b}}{n_g}\left(\frac{1}{3\sigma_{\mathrm{m0}}\sigma_{\mathrm{exc0}}}\right)^{1/2}$$

where E_b is the bulk plasma electric field, n_g is the gas density, σ_{m0} and σ_{exc0} are the elastic and excitation cross sections

(B.M. Smirnov, 1981; M.A. Lieberman and A.J. Lichtenberg, 2005)

- Effective rate coefficient for the He metastable excitation

$$\overline{K} = K_0(2\widetilde{\mathrm{T}}_e)\underbrace{\left(\frac{\widetilde{\mathrm{T}}_c}{\widetilde{\mathrm{T}}_e}\right)^2}_{\text{Due to break energy}}\mathrm{erfc}\left(\sqrt{\mathcal{E}_a/2\widetilde{\mathrm{T}}_e}\right)$$

DISCHARGE CHEMISTRY

- Particle balance for each species

$$\frac{\mathrm{d}n_j}{\mathrm{d}t} = G_j - L_j$$

G_j = volume creation rate (2-body, 3-body and surfaces)
L_j = volume loss rate (2-body, 3-body, and surfaces)

- Numerically integrated using the effective rate coefficients $\overline{K}$

- The analytical solution of
  — the discharge dynamics
  — the time-varying $\mathrm{T}_e(t)$
  — the effective rate coefficients $\overline{K}$
  AND
  the numerical solution of the particle balances

$$\boxed{\Longrightarrow \text{fast solution of the discharge equilibrium}}$$

SUMMARY OF SYSTEMS STUDIED

- He/0.1%N_2, 8 species, 15 reactions
  — Comparison to 1D fluid plus Bolsig+ kinetic simulations:
  (J. Waskoenig, Thesis, Queens U, 2010; PSST **19**, 045018, 2011)
  — Mostly within a factor of 2 for all species densities, $\overline{\mathrm{T}}_e$, V_{rf}, etc
  — Simulation time ≈ 7 s on a MacBook Pro

- He/0.5%O_2, 16 species, 127 reactions
  — As above; simulation time ≈ 40 s

  (C. Lazzaroni et al, MS submitted to Plasma Sources Sci. Technol. 2011)

- Ar/13%O_2/0.073%$TiCl_4$, 28 species, 195 reactions
  Some crude comparisons to anastase crystalline TiO_2 deposition
  (Dexin Wang et al, MS submitted to Nature, 2011)
  — Precursor identified as TiO_2Cl_3, $DR \approx 0.5$ nm/s, etc
  — Simulation time ≈ 95 s

  (A. Leblanc et al, MS submitted to Plasma Chem. Plasma Process. 2011)

OVERALL SUMMARY

- PIC/kinetic simulations show an EEPF strongly modulated at the rf frequency
- A one-dimensional hybrid analytical-numerical global model of atmospheric pressure, rf-driven capacitive discharges was developed.
- Coupling analytical solutions of the time-varying discharge and EEPF dynamics, and numerical solutions of the discharge chemistry, allows for a fast solution of the discharge equilibrium.

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