Introduction to kinetic modeling in low-temperature plasmas

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Outline

1. Motivation: applications of low-temperature plasmas.
2. Plasma processing for semiconductor production equipment makers: $20B/year industry
3. Plasma sources for semiconductor manufacturing
4. Plasma needs to be considered using kinetic theory
5. Particle-in-cell codes
6. Quantum Chemistry Codes
7. Princeton collaborative user facility
8. Conclusions

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1. Motivation: Applications of low-temperature plasmas Why low temperature plasma is important?

Multiple multibillion-dollar industries:

- Plasma processing, \$15-20 B/year wafer fab equipment for 2T/year semiconductor industry;
  - (R. Gottscho, et al, White Paper for Plasma2020)
- Thin films;
- Electric propulsion;
- Nanomaterial synthesis;
- Advanced plasma manufacturing (welding, cutting);
- Plasma switches for electric grids and pulsed power.

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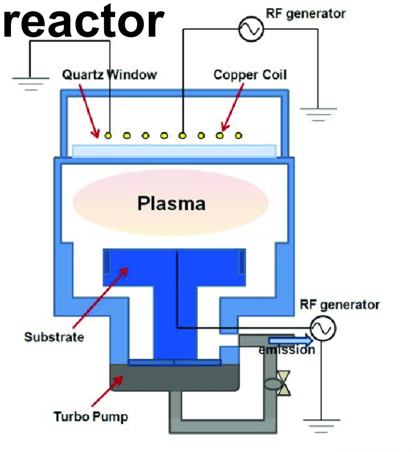
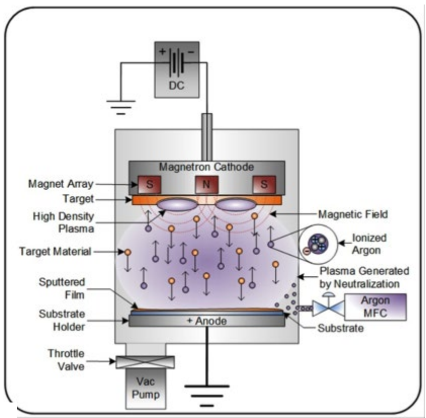
Low Temperature Plasma Applications

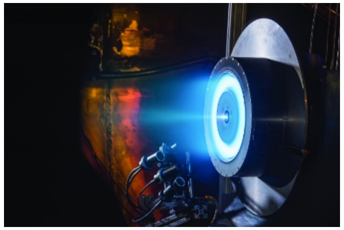
Plasma processing reactor



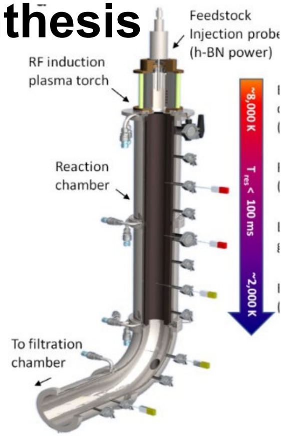
Magnetron for thin films



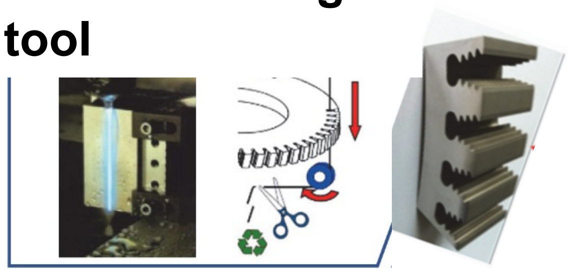
Plasma thruster

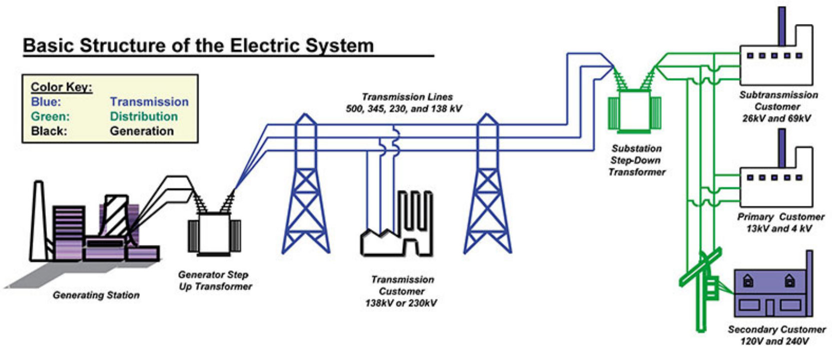


Plasma Reactor for nanomaterial synthesis

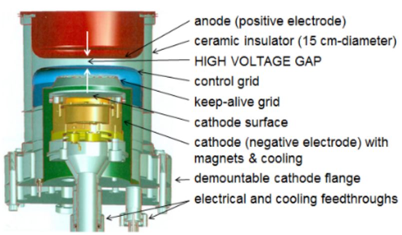


Plasma cutting tool





Gas tube switch



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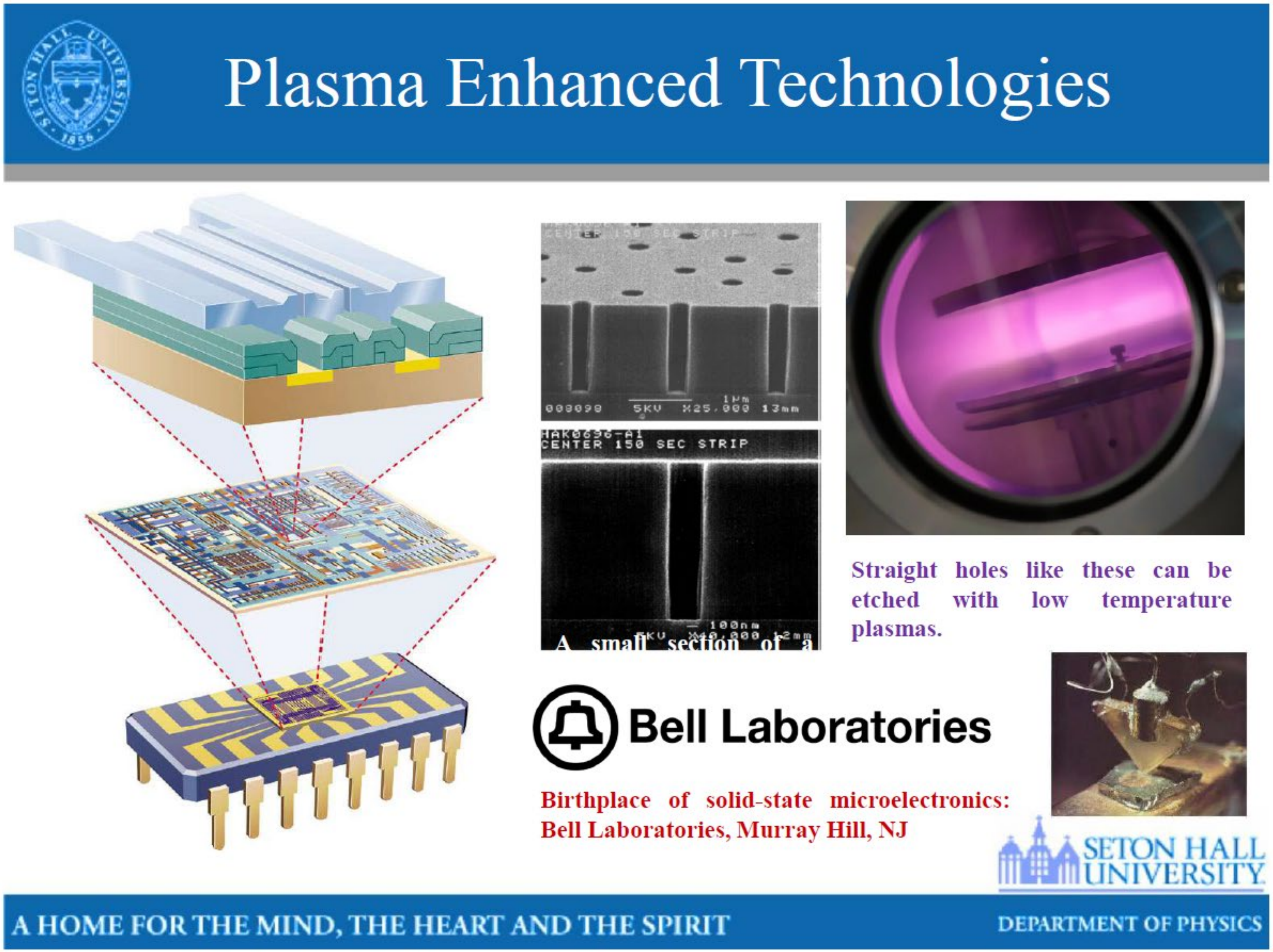
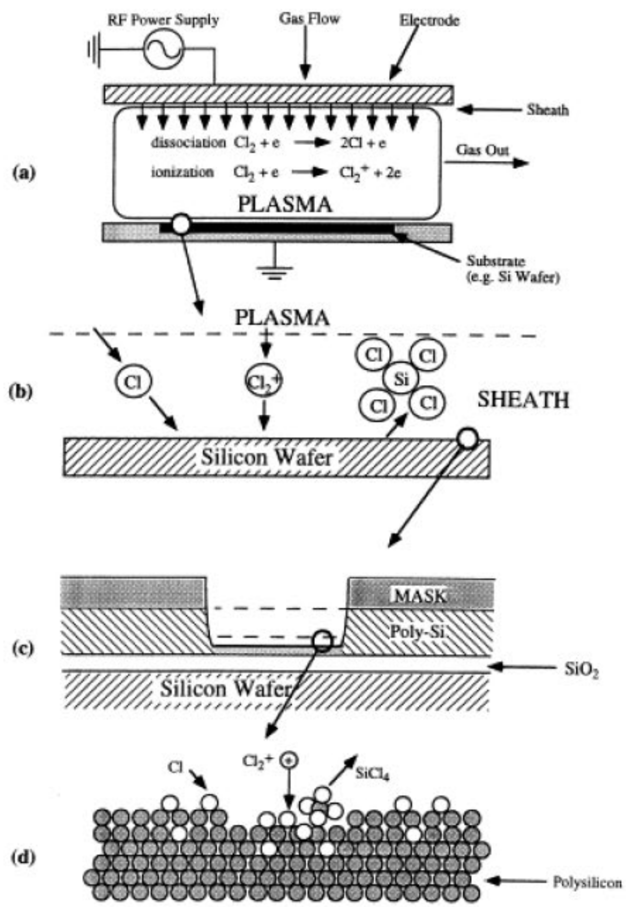
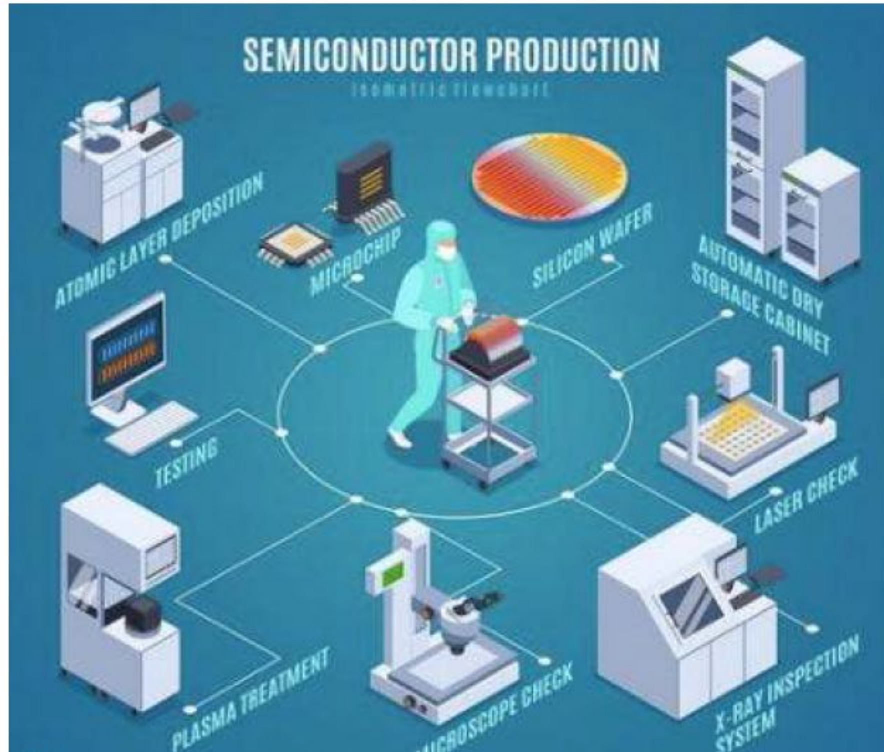
2. Plasma processing for semiconductor production equipment makers: $20B/year industry



Slide curtesy of J. Lopez, Seton Hall University

Low- Temperature Plasma enabled Microchip Fabrication





Reference:
https://eenews.cdnartwhere.eu/sites/default/files/styles/inner_article/public/sites/default/files/images/chipproductio n630_0.jpg?itok=rzZu2ANq

Reference: Demetre J. Economou. Modeling and Simulation of plasma etching reactors for microelectronics. Thin Solid Films. Vol. 365, Issue 2, p. 348-367. April 2000.

SETON HALL UNIVERSITY

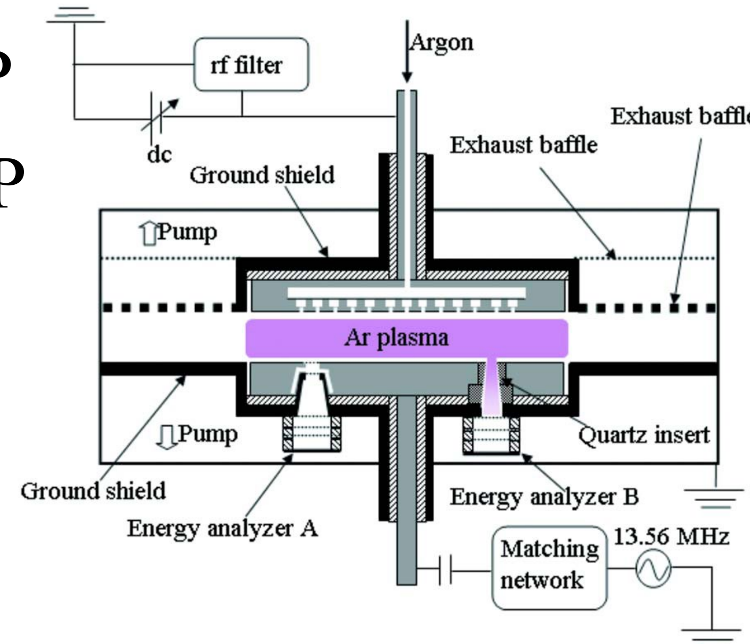
Slide curtesy of J. Lopez, Seton Hall University

Plasma processing, $15-20 B/year wafer fab equipment for 2T/year semiconductor industry.

- The CHIPS for America Act includes a range of federal investments to advance U.S. semiconductor manufacturing. The bill also includes a refundable investment tax credit $10 billion for a new federal grant program that would incentivize new domestic semiconductor manufacturing facilities.
- Research is critical to advancing semiconductor innovation in the U.S. American semiconductor design and manufacturing companies invest approximately one-fifth of revenue in R&D, almost $40 billion in 2019 (https://www.semiconductors.org/chips-for-america-act-would-strengthen-u-s-semiconductor-manufacturing-innovation/)
- PPPL: the Princeton Plasma Innovation Center (**PPIC**). New national facility will explore low-temperature plasma — a dynamic source of innovation for modern technologies. https://www.princeton.edu/news/2019/09/05/new-national-facility-will-explore-low-temperature-plasma-dynamic-source-innovation

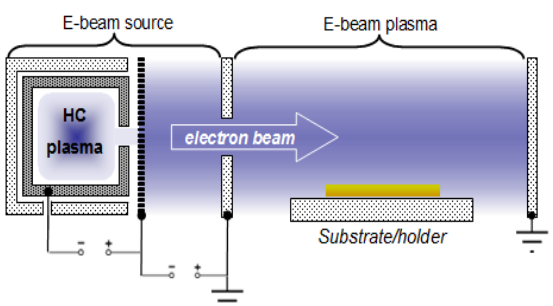
3. Plasma Sources for semiconductor manufacturing

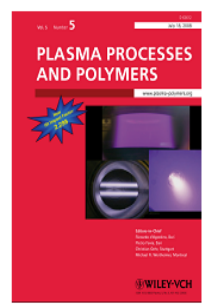
- Capacitively-coupled plasmas CCP
- Inductively-coupled discharges ICP
- Electron and Ion beams



Electron beam generated plasma processing system

Large Area Plasma Processing System (LAPPS)*

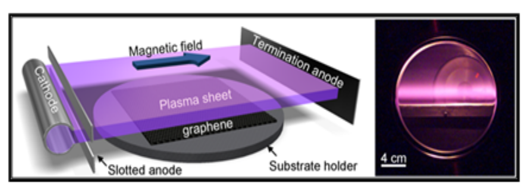




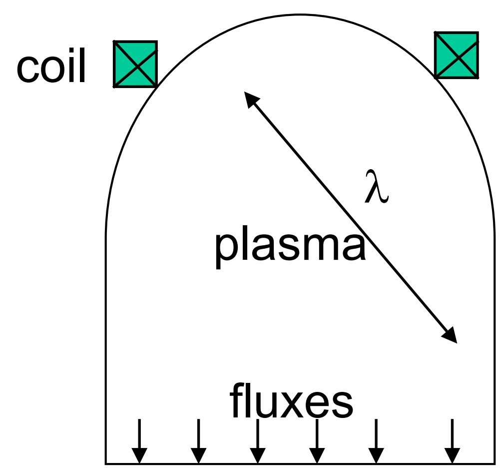
S.G. Walton *et al*, Plasma Proc. and Poly. 5, 453 (2008)

Basic Operation for Processing System

- High-energy (few keV), sheet beam injected into background
- Magnetically collimated to minimize spreading
- Creates plasma sheet parallel to substrate surface
- Large amount of flexibility in system design



M. Baraket *et al*, App. Phys. Lett. 100, 233123 (2012)



* Meger et al., US patent no. 5,874,807 (Feb. 1999)

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Plasma needs to be considered using kinetic theory

- Most remote from thermodynamic equilibrium:
  - T_e differs from T_i

3eV	**$3\ 10^{-2}$ eV**	**glow discharges**
$3\ 10^{-3}$ eV	**$3\ 10^{-2}$ eV**	**afterglow**
10keV	**1eV**	**ECR ion sources**

- Electron, ion, photon energy distribution functions (DF) are all nonMaxwellian:
  - Parts of the DFs are very flexible and almost independent.

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Plasma parameters in CCP and ICP

- Plasma density $n = 10^9$ - 10^{13} cm^{-3}
- Gas pressure = few mTorr
- Small degree of ionization < 10^{-4}
- Electron temperature T_e = few eV
- Ion temperature T_i = 0.03 eV
- Spatial scale = mm- m

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Global model: particle balance determines T_e

Assuming steady-state and a Maxwellian Electron Energy Distribution Function =>

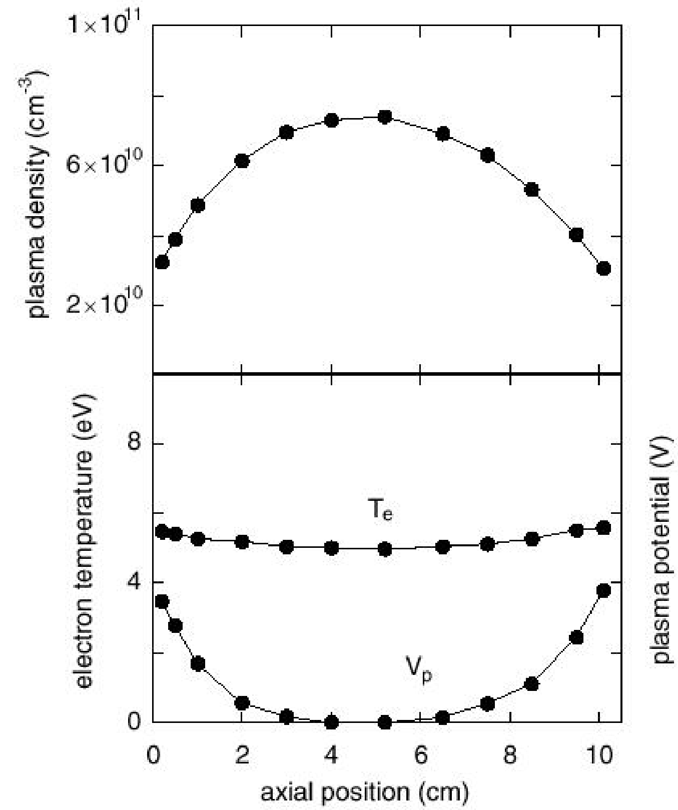
- Rate of plasma production = rate of plasma loss,
- Ionization frequency = loss frequency to the wall

$$\nu_{iz}(T_e) = \nu_{loss}(T_e)$$

$$\nu_{iz} = n_g V_T \sigma_{iz} e^{-I/T_e},$$

$$\nu_{loss} = \gamma C_s / L \quad C_s = \sqrt{T_e / M}$$

$$n_g V_T \sigma_{iz} e^{-I/T_e} = \gamma \frac{\sqrt{T_e / M}}{L}$$



V. Godyak's experiment is benchmark.

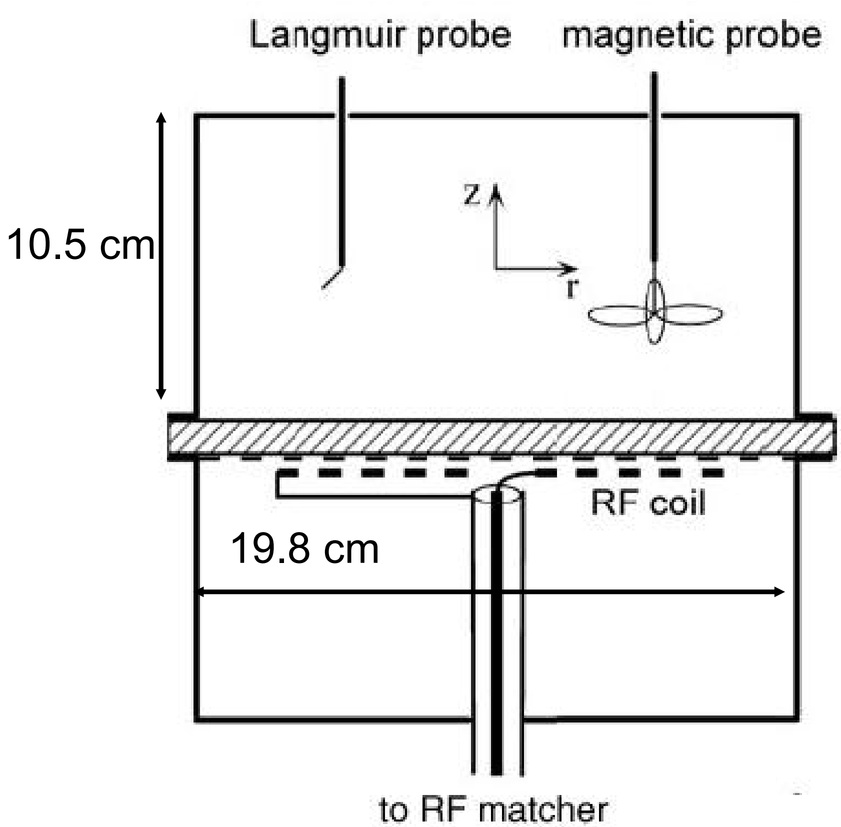


Figure 1. Experimental discharge chamber.

The Inductively Coupled Plasmas (ICP)

f=0.45-13.56 MHz

argon gas pressures 0.3-300 mTorr

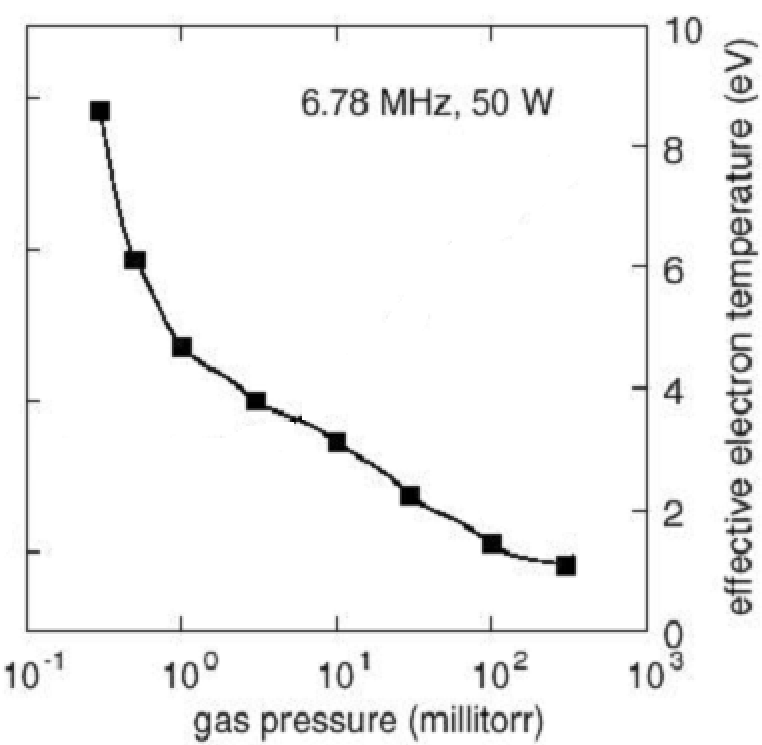
rf power 6-400 W.

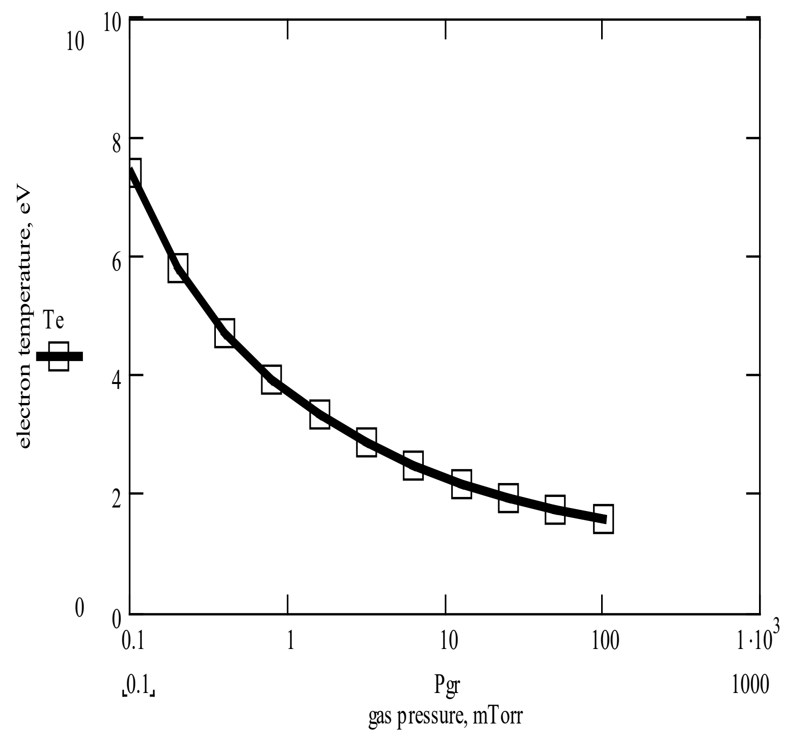
V. A. Godyak, R. B. Piejak, and B. M. Alexandrovich, J. Appl. Phys. **85**, 703 (1999).

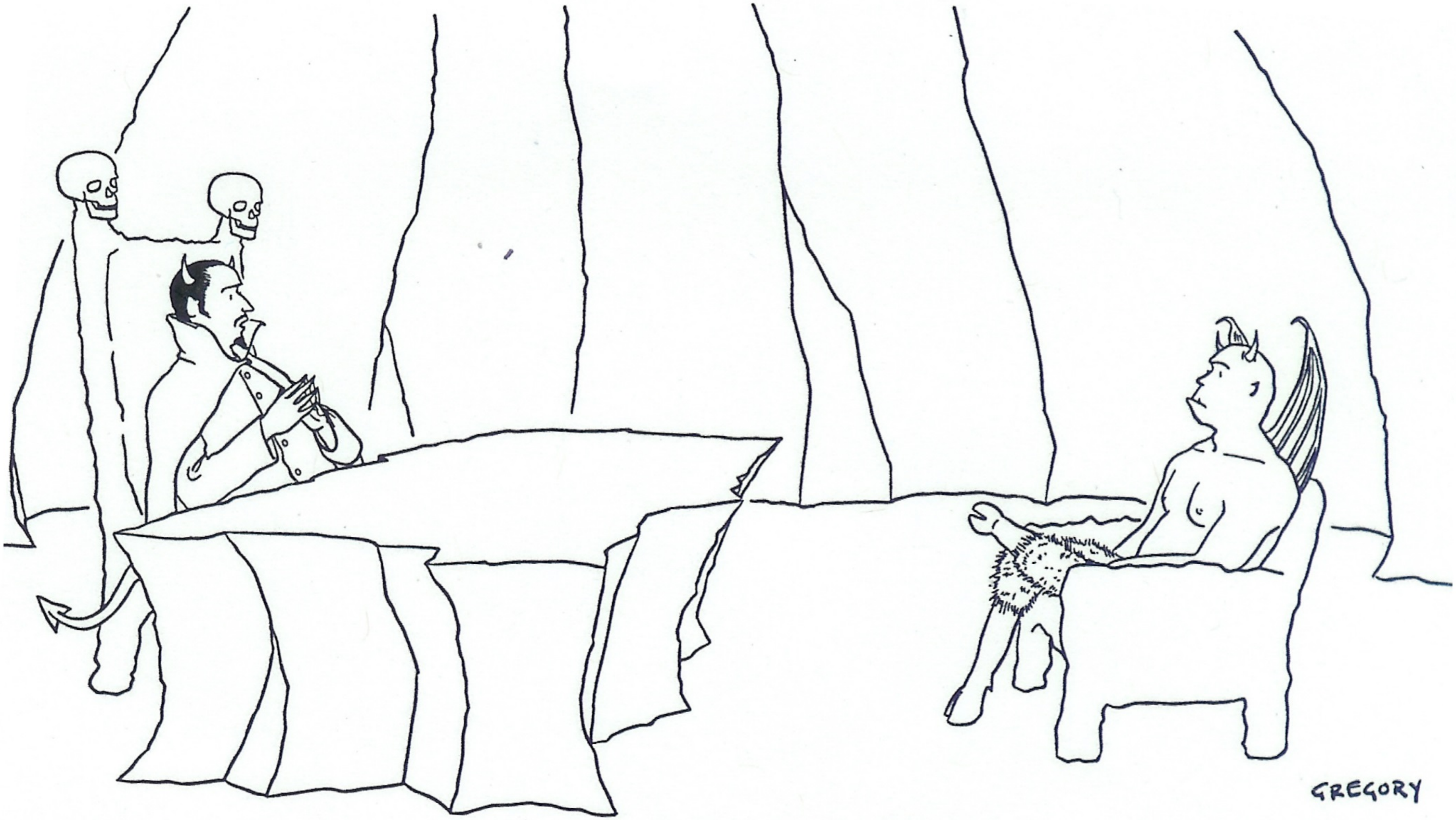
Experiment (left) vs global model (right)

$$n_g V_T \sigma_{iz} e^{-I/T_e} = \gamma \frac{\sqrt{T_e / M}}{L} \quad \uparrow n_g L \ \downarrow T_e$$

T_e as a function of gas pressure at f=6.78 MHz







"I need someone well versed in the art of torture—do you know PowerPoint?"

TABLE 1.1. Range of Parameters for rf Diode and High-Density Discharges

Parameter	rf Diode	High-Density Source
Pressure p (mTorr)	10–1000	0.5–50
Power P (W)	50–2000	100–5000
Frequency f (MHz)	0.05–13.56	0–2450
Volume $\mathcal{V}$ (L)	1–10	2–50
Cross-sectional area A (cm^2)	300–2000	300–500
Magnetic field B (kG)	0	0–1
Plasma density n (cm^{-3})	10^9–10^{11}	10^{10}–10^{12}
Electron temperature T_e (V)	1–5	2–7
Ion acceleration energy $\mathcal{E}_i$ (V)	200–1000	20–500
Fractional ionization x_{iz}	10^{-6}–10^{-3}	10^{-4}–10^{-1}

$$j = \frac{\partial E}{4\pi \partial t} + \sigma E$$

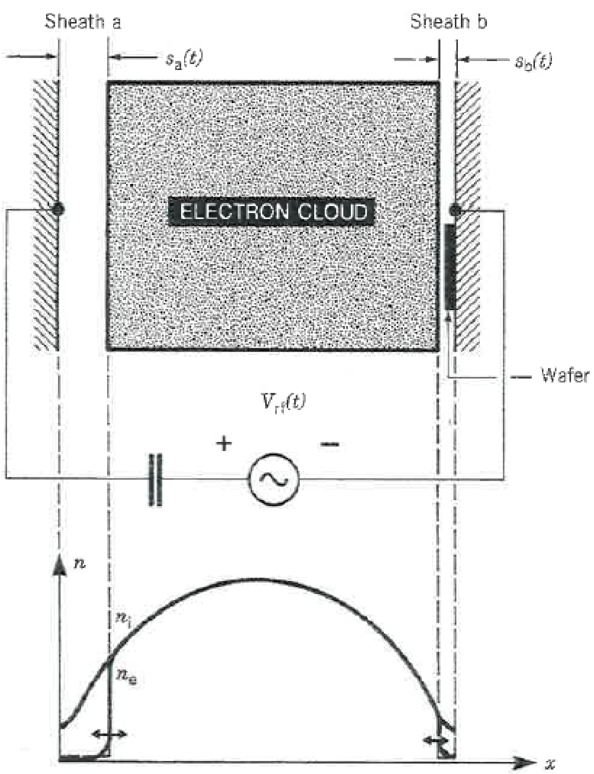
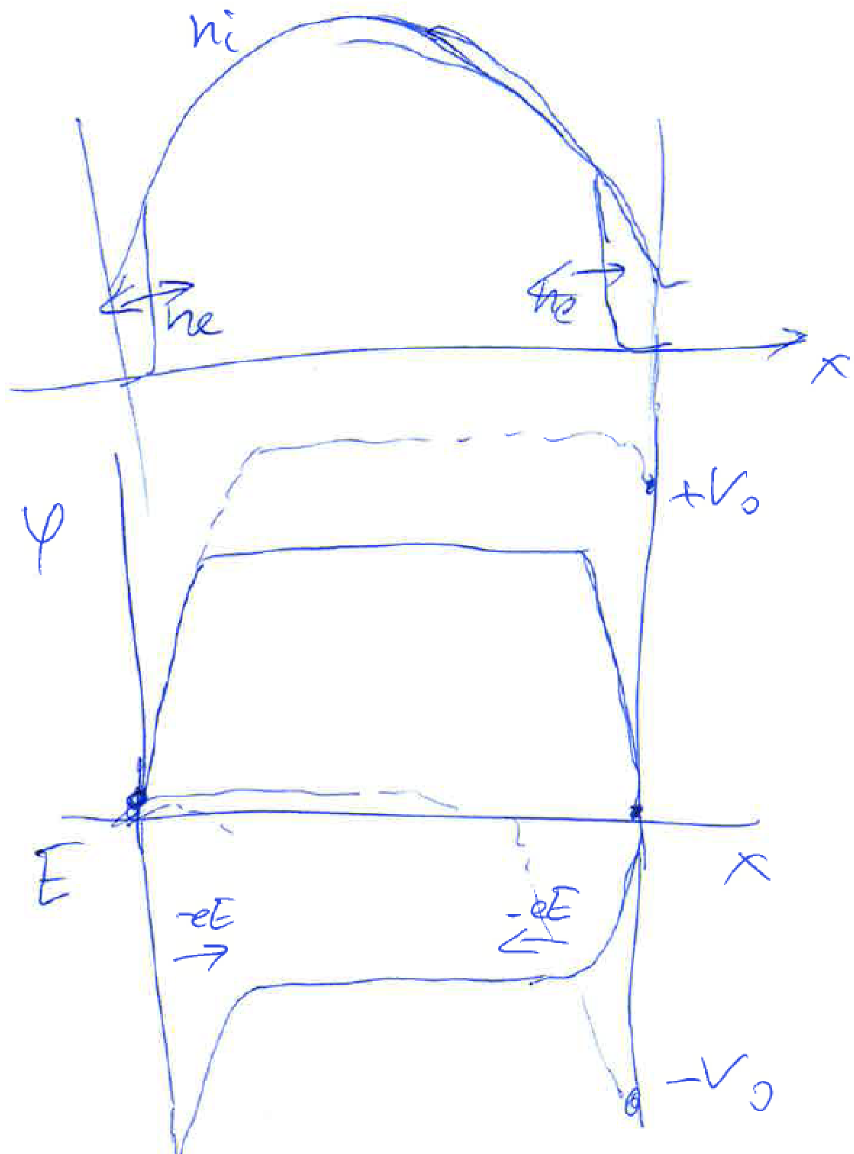
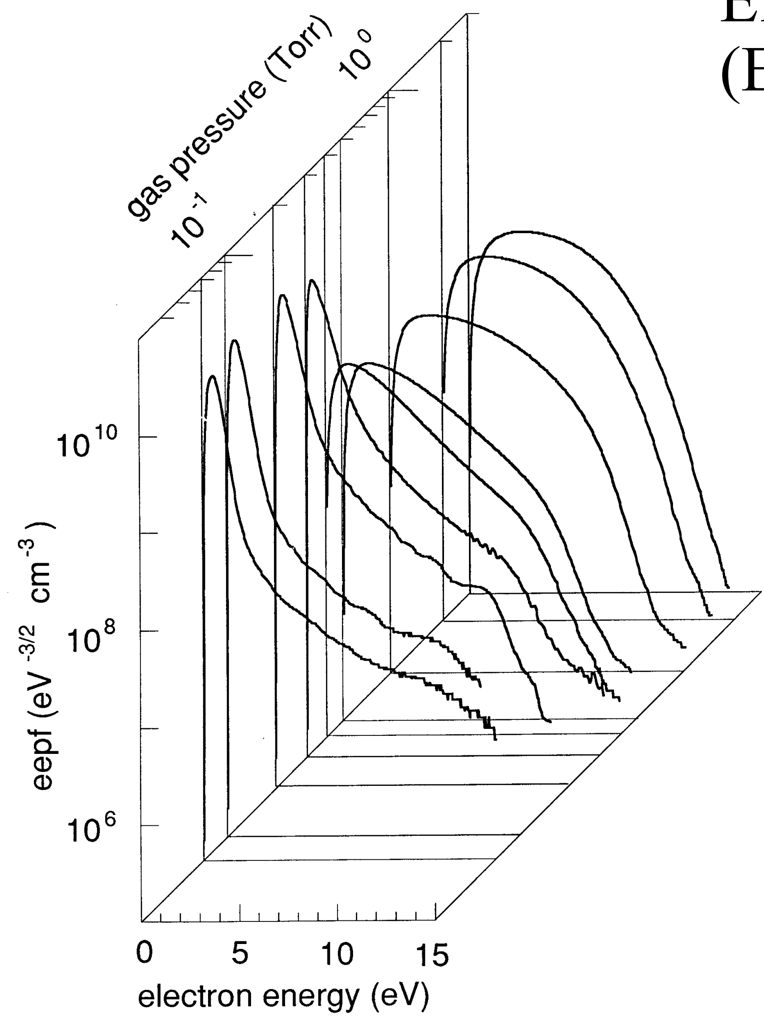


FIGURE 1.13. The physical model of an rf diode (after Lieberman and Gottscho, 1994).



Discharge modeling needs to be kinetic!



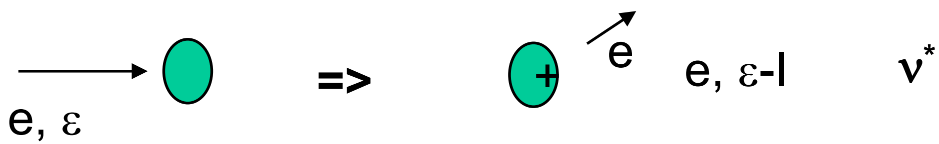
Electron energy distribution functions (EEDF) are non-Maxwellian:

- Parts of the EEDF are very flexible and almost independent.
- An example of a EEDF in capacitive discharge.

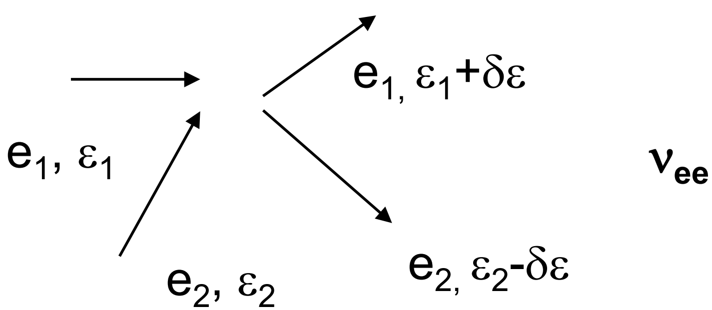
For more info: V. Godyak, IEEE TPS 34, 755 (2006).

Mechanisms of EEDF formation

Cooling is due to energy losses in elastic and inelastic collisions.



Mixing is due to electron-electron collisions.



Electron-electron collisions make EEDF a Maxwellian! =>

If $\nu_{ee} >> \nu^*$ or the degree of ionization, $n_e/n_g > 10^{-4}$, EEDF is a Maxwellian;

If $\nu_{ee} << \nu^*$ or $n_e/n_g < 10^{-4}$, EEDF can have any shape,

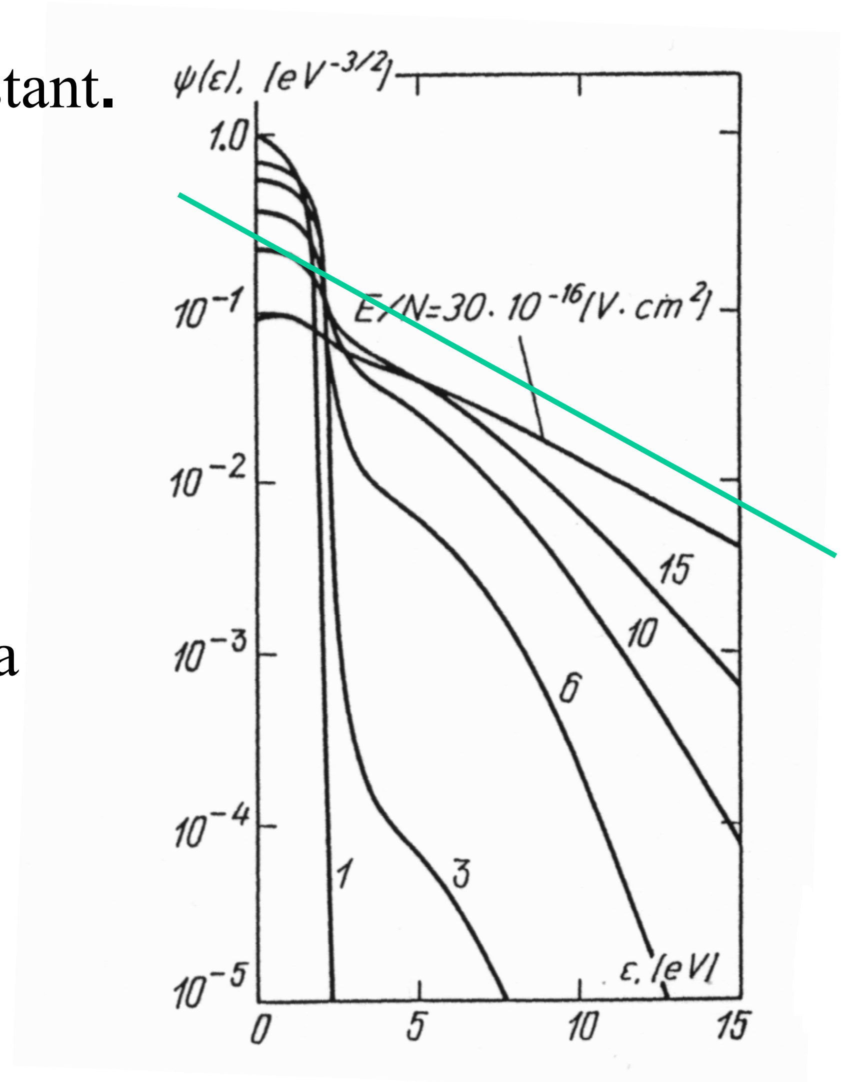
EEDF in nitrogen, constant electric field

1930 Druyvesteyn's EEDF for λ constant.

$$f \sim \exp(-\varepsilon^2/\varepsilon_0^2)$$

Real cross-sections in N_2

N.L. Alexandrov, *et al*. Sov. J. Plasma Phys. 1978



Nonlocal, nonlinear electron kinetics

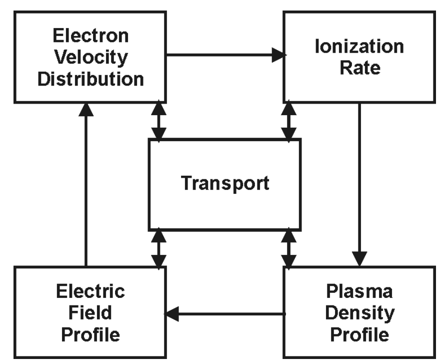
- Formation of non-Maxwellian, non-uniform, *f(v,r,t)* in self-consistent electric field strongly coupled to plasma density profile through ionization and transport for realistic discharge plasmas.

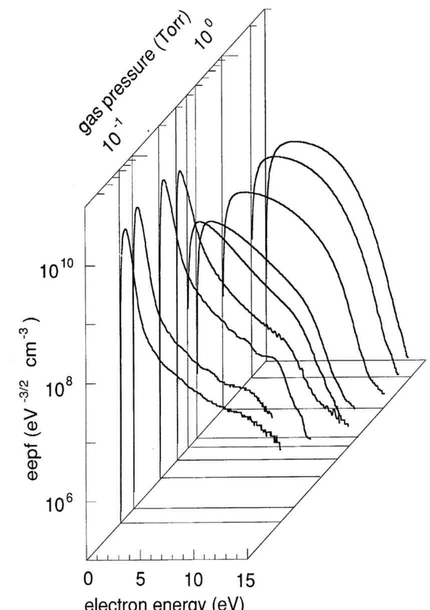
$$\left[\frac{\partial}{\partial t}+(\mathbf{v}\nabla)-e\mathbf{E}\frac{\partial}{m\partial \mathbf{v}}\right]f=\sum_k\left[\nu_k^{*}{}'\frac{\sqrt{u'}}{\sqrt{u}}f\,(w+w_k^*)-\nu_k^* f\right]+St_{ee},$$

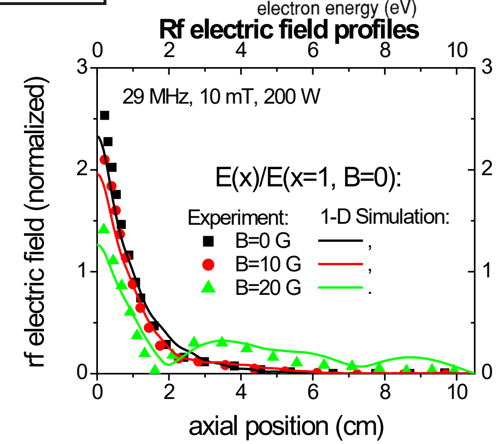
$$\frac{\partial n_i}{\partial t}+\nabla(\mathbf{v}n_i)=\nu_{iz}n_i,\ \nu_{iz}n_i=\int_I^\infty f(u)v\sigma_{iz}d\mathbf{v}$$

$$\nabla\mathbf{E}=4\pi e(n_i-n_e)$$

$$M_i\left(\frac{\partial\mathbf{v}}{\partial t}+(\mathbf{v}\nabla)\mathbf{v}\right)=e\mathbf{E}-\frac{\nabla p_i}{n_i}-(\nu_{ia}+\nu_{iz})\mathbf{v}$$

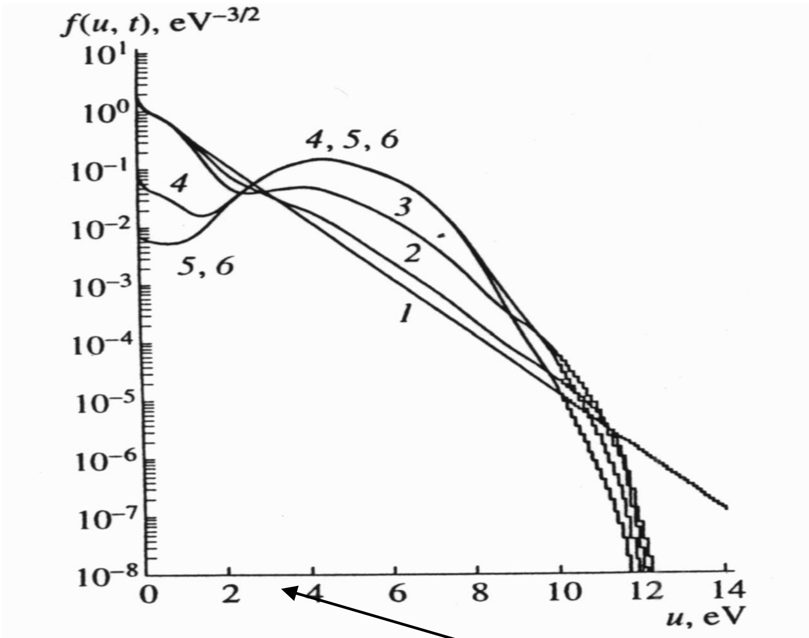






- Igor D. Kaganovich et al, Phys. Rev. Lett. 1999, 2000, 2002, 2009, 2012, 2013;
- Valery Godyak et al, Phys. Rev. Lett. 1990,1992, 1996, 1998, 1999.

EEDF in decaying plasma



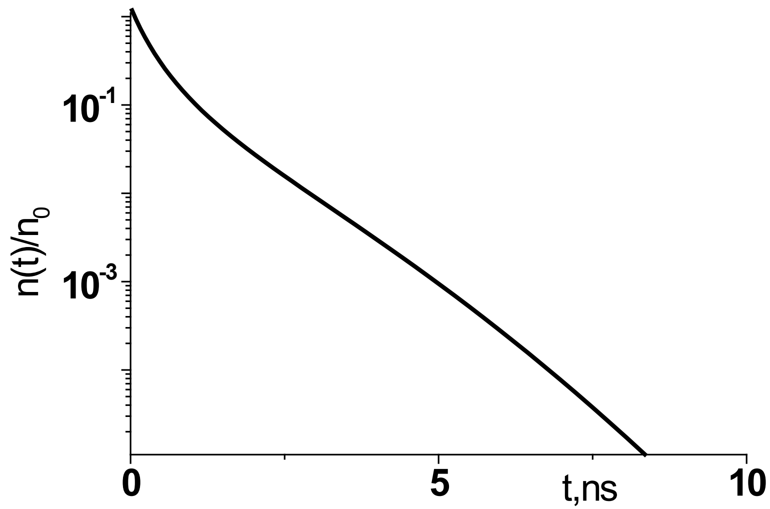


Fig. density as function of time in decaying plasma

Fig. EEDF afterglow Ar:NF_3. E/N=2 10^{-17}Vcm^2

- 1, 2, 3, 4, 5, 6.
- 0, 0.25, 1, 3, 5, 10 ns.

N.A. Dyatko, *et al.* Plasma Phys. Rep. 1998

Strong energy losses due to vibrational excitation

5. Particle-in-cell codes (PIC) Preferred Computational Tool - PIC

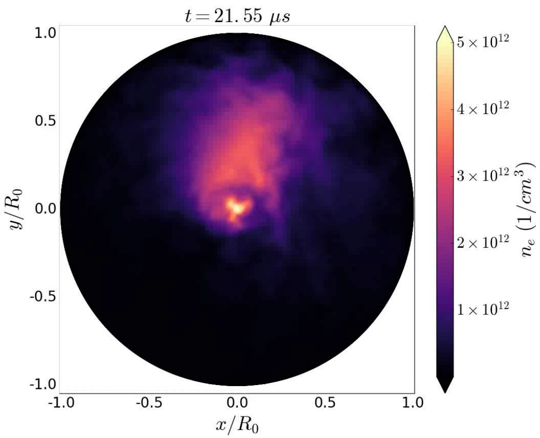
Parallel, implicit particle-in-cell code with energy corrections and noise control is a powerful and versatile tool.

- Ease of coding
- Modular, “easy” for parallelization
- Modern clusters and multi cores PCs are cheap

Selected PPPL computational and scientific accomplishments using PIC

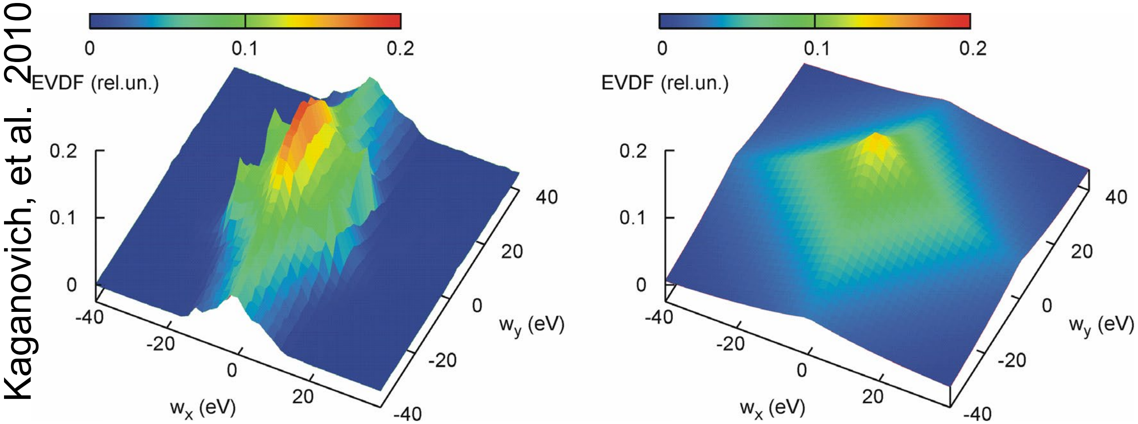
Self-organized structures in magnetized plasmas

A. Powis, et al., 2018



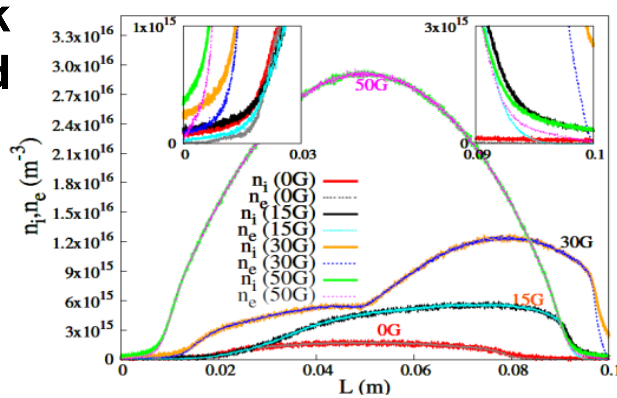
Non-Maxwellian Electron Velocity Distribution in thrusters

D. Sydorenko, I. Kaganovich, et al. 2010



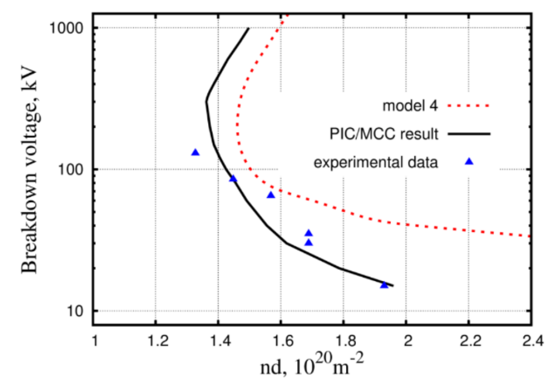
Effect of weak magnetic field on radio-frequency discharge

S. Sharma, et al. 2019



Breakdown in high voltage devices

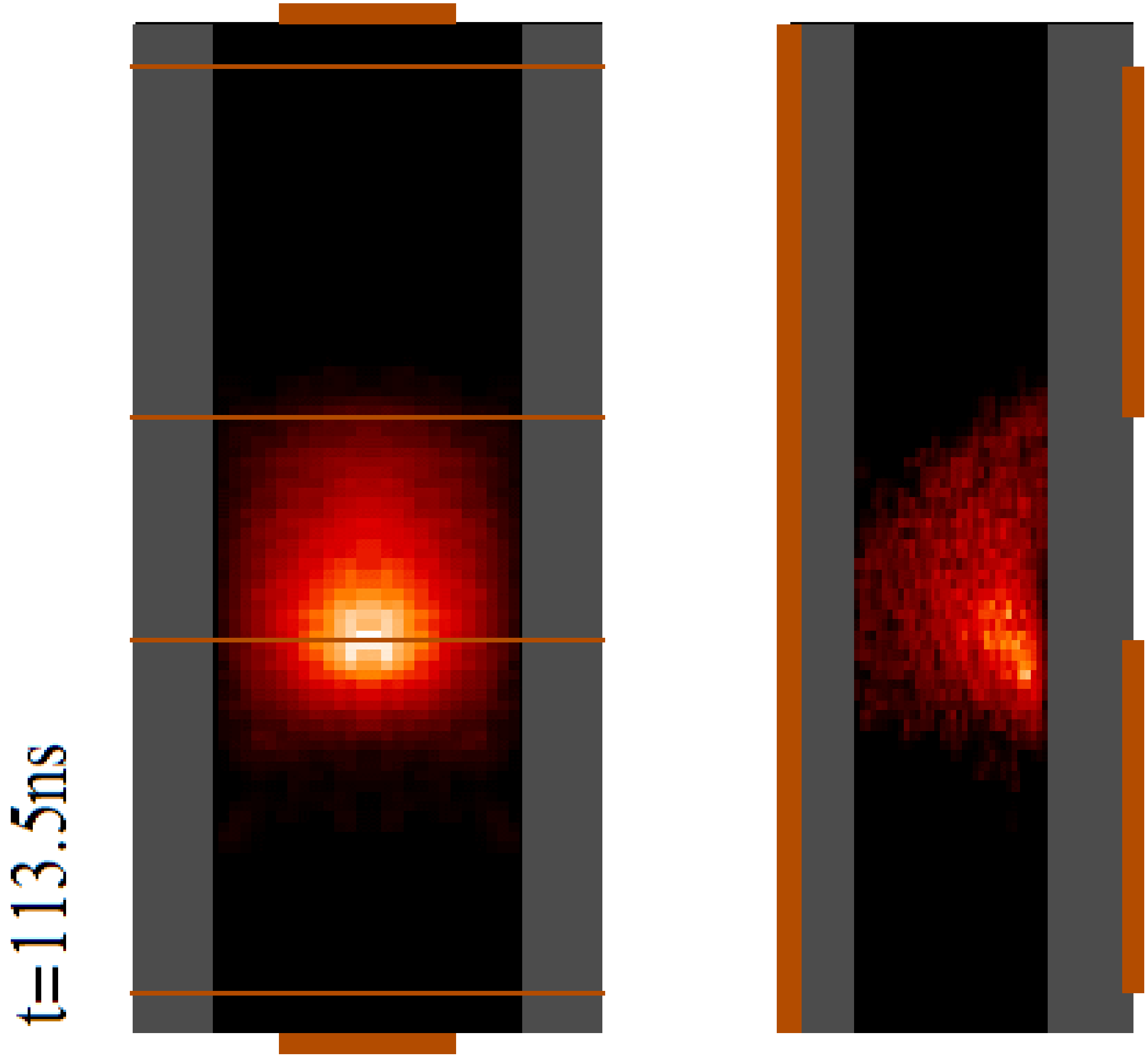
L. Xu, et al. 2019



3D code for Plasma Panel Modeling

Up to 250 million of ions and electrons on up to 64 processors, up to 150x150x150 meshes

V. N. Khudik, A. Shvydky, V. P. Nagorny, and C.E. Theodosiou, *IEEE Trans Plasma Sci.* **33**, 510 (2005).

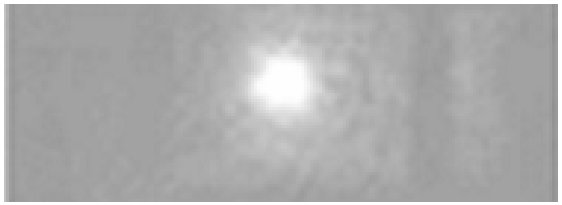


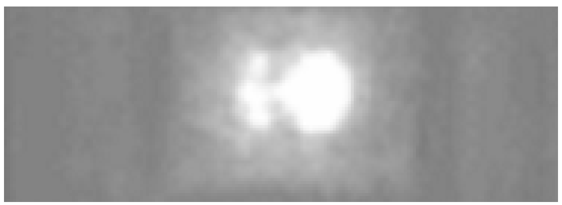
Discharge evolution experiment

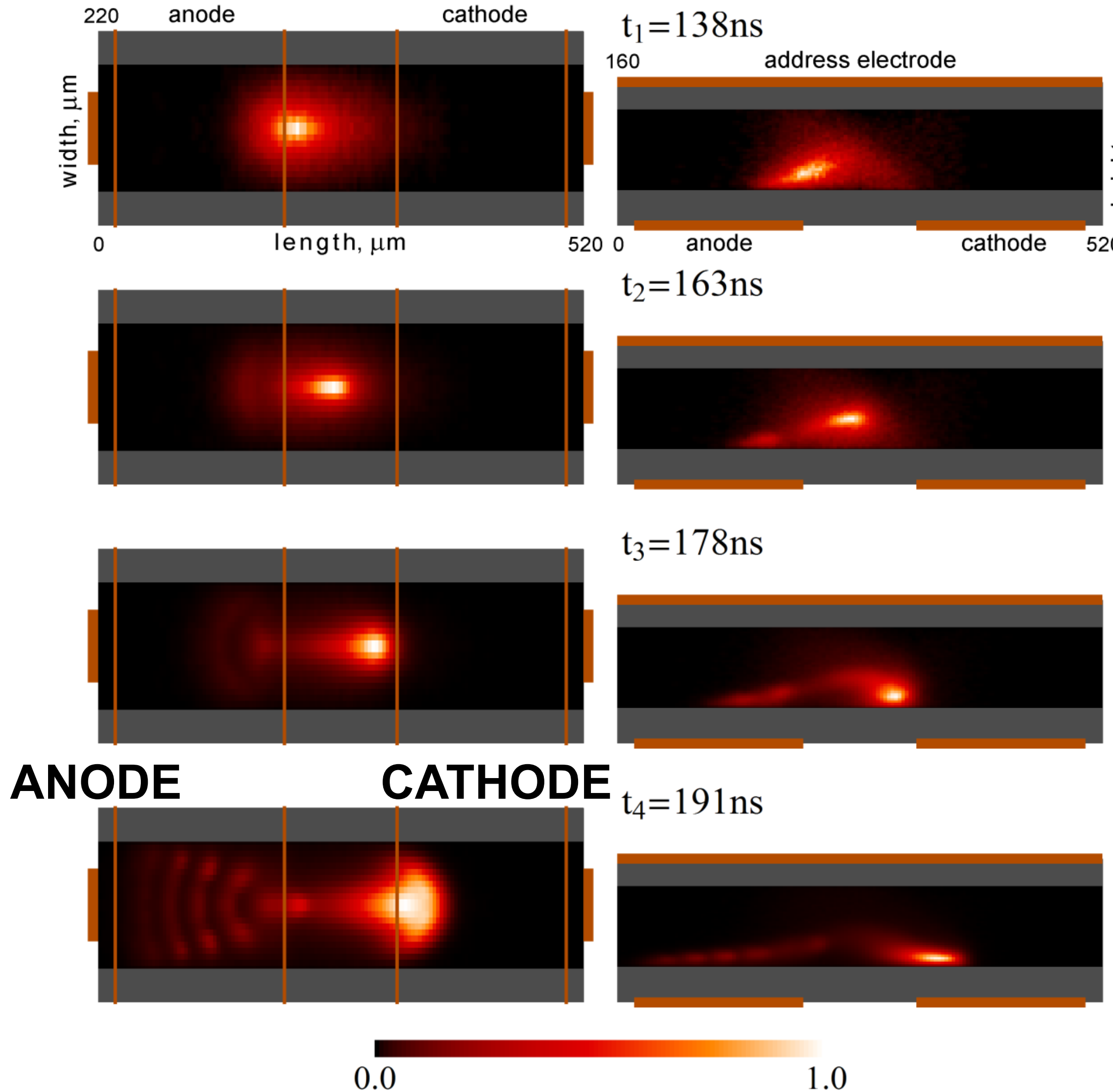
PIC simulations

Top view

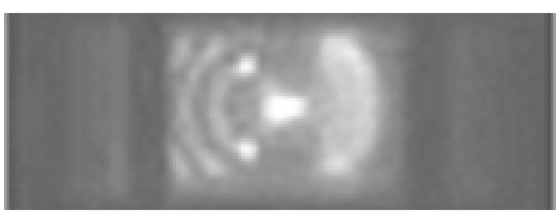
Side view







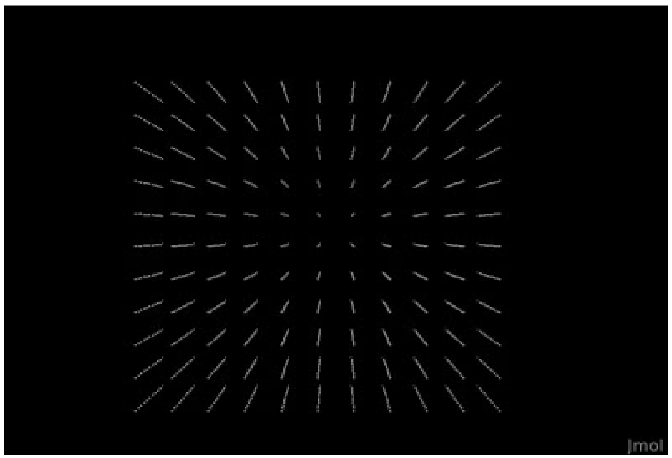
V. N. Khudik, A. Shvydky, V. P. Nagorny, and C.E. Theodosiou, *IEEE Trans Plasma Sci.* **33**, 510 (2005).



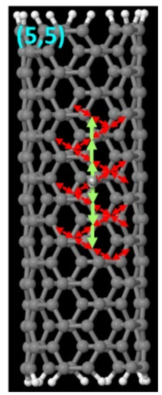
6. Quantum Chemistry Codes

Quantum chemistry calculations for predictions of carbon and boron nitride nanotube synthesis mechanisms, see nano.pppl.gov

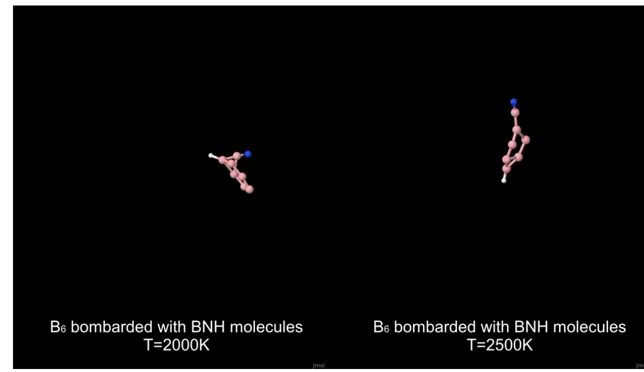
Carbon chains and fullerenes



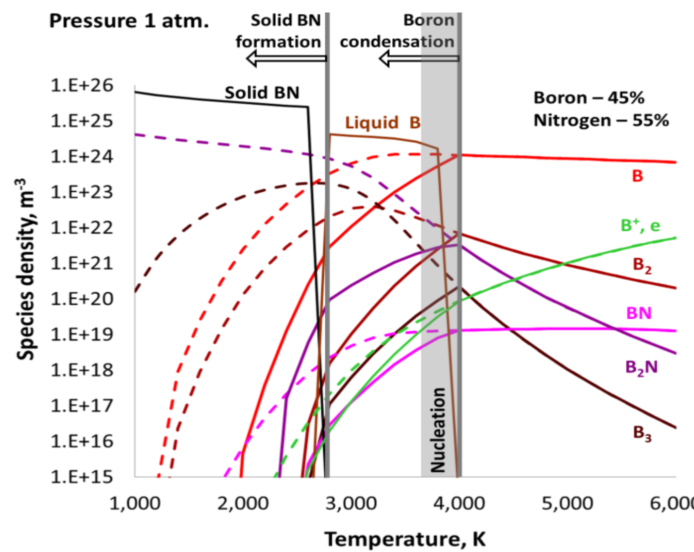
Carbon diffusion



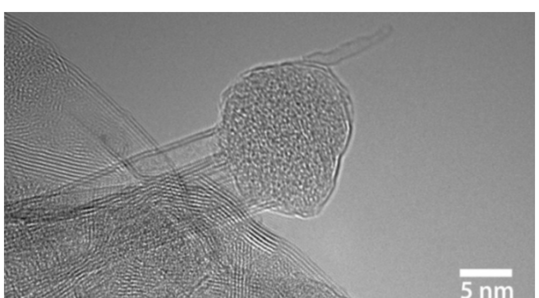
boron nitride fullerenes

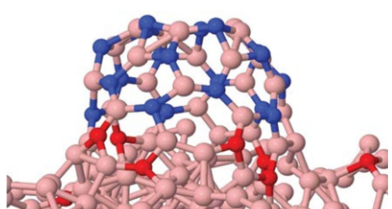


boron nitride feedstock evolution



boron nitride tubes formation





Conclusions

•Plasma processing is a multibillion dollar industry in revenues with inadequately funded fundamental research support.

•Semiconductor equipment requires precise control of plasma surface interaction. Partially-ionized plasmas allows for such control because of great availability in choices of electron, ion and photon energy distribution functions that can be designed and controlled according to required specifications.

•Industry will greatly benefit from innovative methods of designing electron, ion and photon energy distribution functions delivered to required surfaces.

•Examples of such innovative methods for crafting electron, ion and photon energy distribution functions have been recently developed.

PPPL Princeton Collaborative Research Facility

- **Established by DOE FES**
- **PPPL is the lead**
- **Emphasis: Integrated experimental and modeling efforts on LTP**
- **Research staff: 6**
- **Has external research collaboration program (User Program)**

- **Research topics:**
  - **Interactions between plasma and solid state**
    - Plasma-induced surface modifications
    - Surface-induced effects on plasma
  - **Interfacial plasma**
    - Plasma-liquid interactions
  - **Coherent structures**
    - Nanoparticles-plasma interactions
    - Turbulence in plasma

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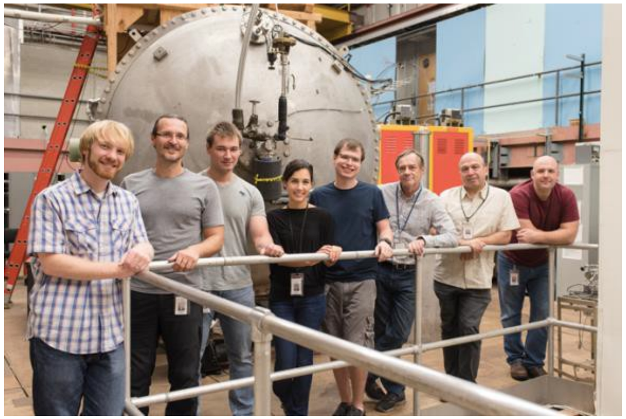
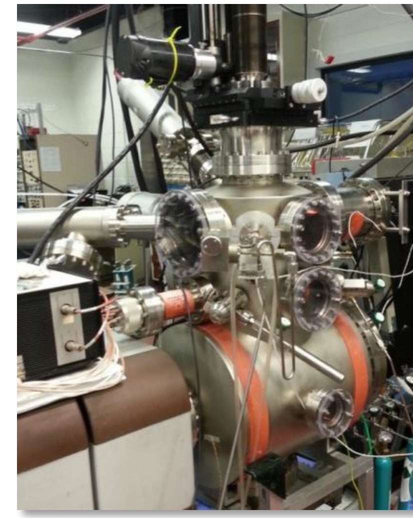
Hall Thruster Experiment (HTX)

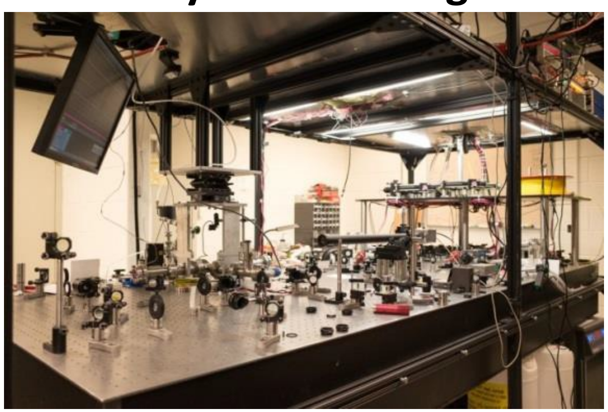


Laboratory for Plasma Nanosynthesis

Surface Science Lab



Laboratory of Laser Diagnostics



- **Total lab space exceeds 5000 sq. ft**
- **Host unique plasma sources and advanced diagnostics for LTP plasmas and applications**
- **Supported by DOE (FES, BES), DOD**

http://htx.pppl.gov & http://nano.pppl.gov 19

+ Princeton University Facility, see talk by A. Dogariu

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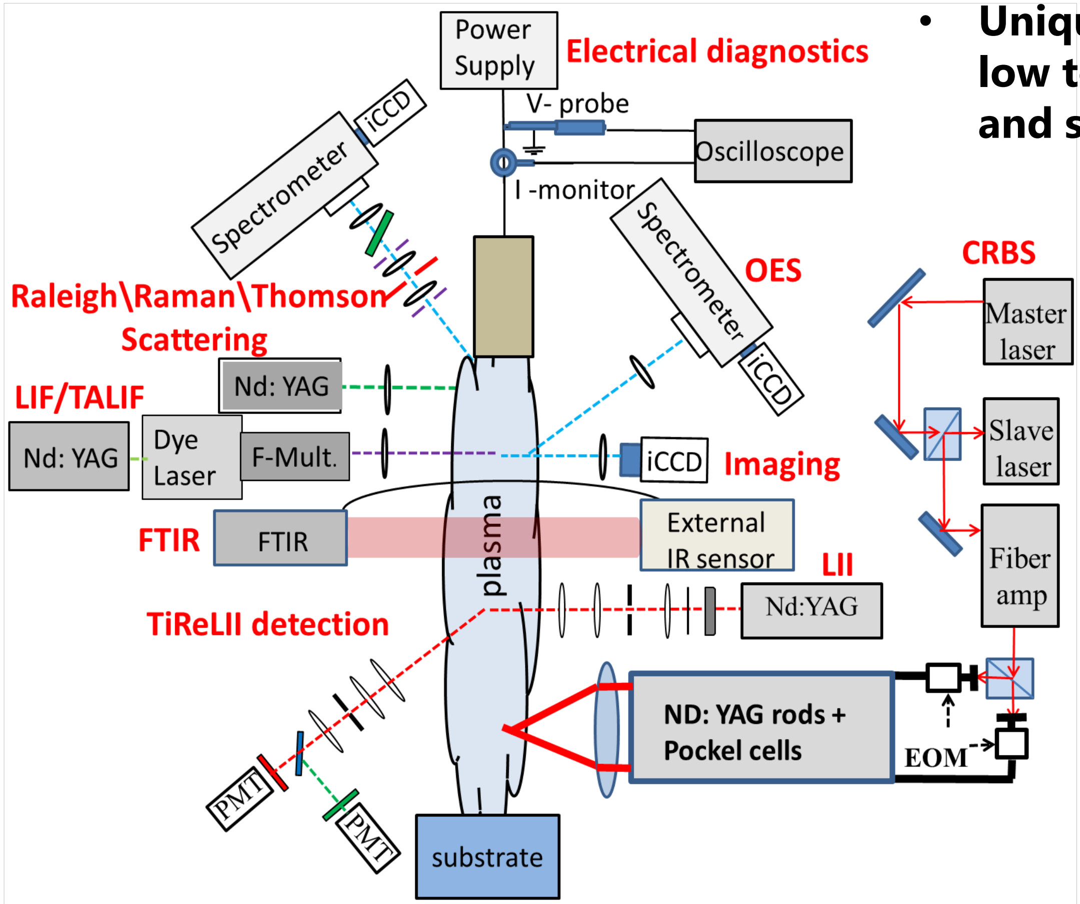
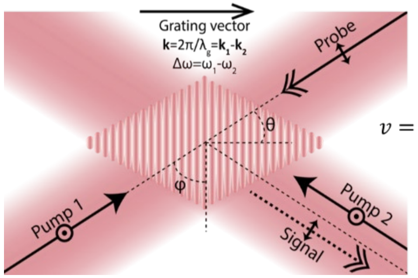
PCRF LTP diagnostics available at PPPL



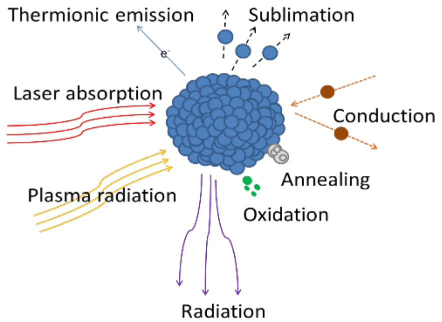
- **Unique combination of ns/DC diagnostics of low to high pressure plasmas, nanoparticles and secondary electron emission & charging**

CRBS



$$v = \frac{\Delta f \lambda_{pump}}{\sin \varphi}$$

LII



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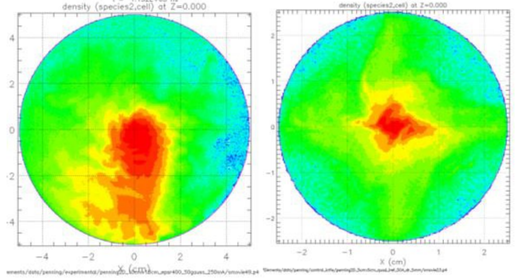
+ Princeton University Diagnostics, see talk by A. Dogariu

Computational Tools for LTP Modeling

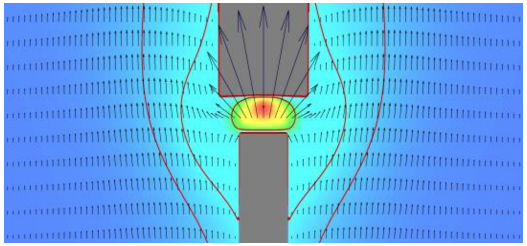
- **Particle-in-cell codes (2D EDIPIC, 3D PPPL-modified LSP)**
  - state of the art collision models and plasma-surface interaction, validated by numerous benchmarks

E-beam plasma



- **Fluid codes (3D ANSYS)**
  - implemented sheath models, MHD effects, surface interface

High-pressure arc



- **Molecular Dynamics (DFT-TB)**
  - DFT codes: full and tight binding approximation, CMD (classical potentials), KMC –kinetic Monte Carlo, and thermodynamic code for chemical composition.

Nanoparticles growth