Different modes of a capacitively coupled radio frequency discharge in methane

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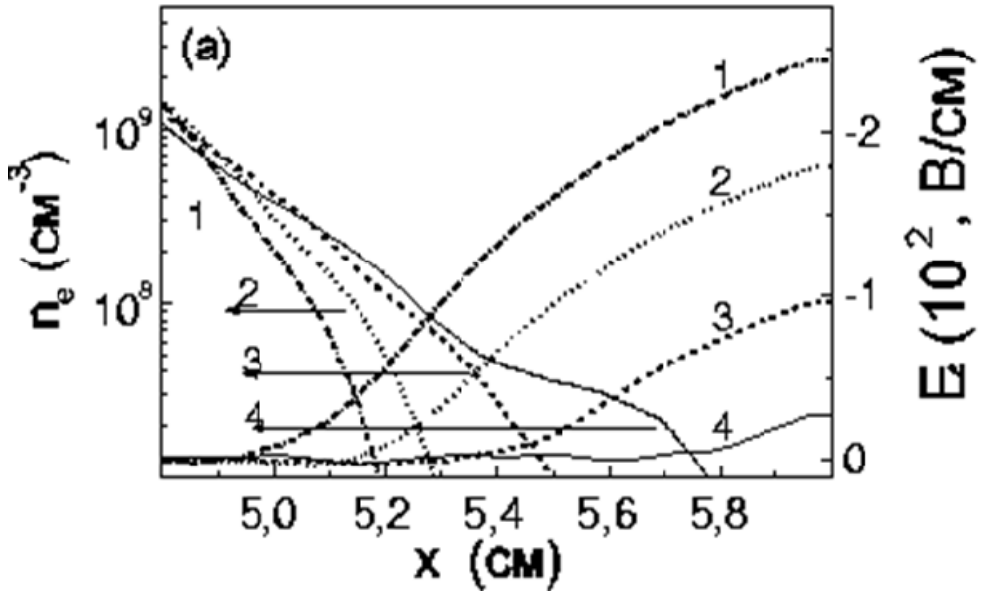
Outline

- Combined PIC MCC algorithm
- Transition between different modes of ccrf discharge in methane
- Phase diagram of ccrf discharge modes
- Hysteresis
- 2D PIC MCC simulation of transition between different modes of ccrf discharge in CH_4
- Conclusion

Why it is necessary to modify Particle in Cell Monte Carlo collision method?

Illustration of electron heating in the rf discharge sheath



electrode

(a) Distributions of electrical field E and electron density ne in different moments of rf cycle:
t=0.2T (1), 0.3T (2), 0.4T (3), 0.5T (4), where T – rf cycle time, P=75 mTorr, j=1 mA/cm2, d=6 cm

Combined model

Kinetic equations for electron and ion distribution functions:

$$\frac{\partial f_e}{\partial t} + \vec{v}_e \frac{\partial f_e}{\partial x} - \frac{e\vec{E}}{m} \frac{\partial f_e}{\partial \vec{v}_e} = J_e, \quad n_e = \int f_e d\vec{v}_e,$$

$$\frac{\partial f_i}{\partial t} + \vec{v}_i \frac{\partial f_i}{\partial x} + \frac{e\vec{E}}{M} \frac{\partial f_i}{\partial \vec{v}_i} = J_i, \quad n_i = \int f_i d\vec{v}_i,$$

Continuity equations for ion and electron densities and currents (see next slide)

Poisson equation:

$$\Delta\phi = 4\pi e(n_e' - n_i'), \quad E = -\frac{\partial \phi}{\partial x}.$$

Schweigert, Schweigert, *New combined PIC-MCC approach for fast simulation of a radio frequency discharge at low gas pressure.* Plasma Source Sci Technol., **13**(2), 315 (2004)

Continuity equations for ion and electron densities and currents

$$\frac{\partial n_e'}{\partial t} + \frac{\partial j_e'}{\partial x} = Q, \qquad \frac{\partial n_i'}{\partial t} + \frac{\partial j_i'}{\partial x} = Q,$$

$$\frac{\partial j_e'}{\partial t} = -\frac{\partial T_e' n_e'}{\partial x} - \frac{eE}{m} n_e' - \nu_e j_e' - Q_e, \qquad \frac{\partial j_i'}{\partial t} = -\frac{\partial T_i' n_i'}{\partial x} + \frac{eE}{M} n_i' - \nu_i j_i' - Q_i,$$

Poisson equation:

$$\Delta\phi = 4\pi e(n_e' - n_i'), \quad E = -\frac{\partial \phi}{\partial x}.$$

$$Q = N_g \int v_{ex} \sigma_i f_e \, \mathrm{d}\vec{v}_e$$

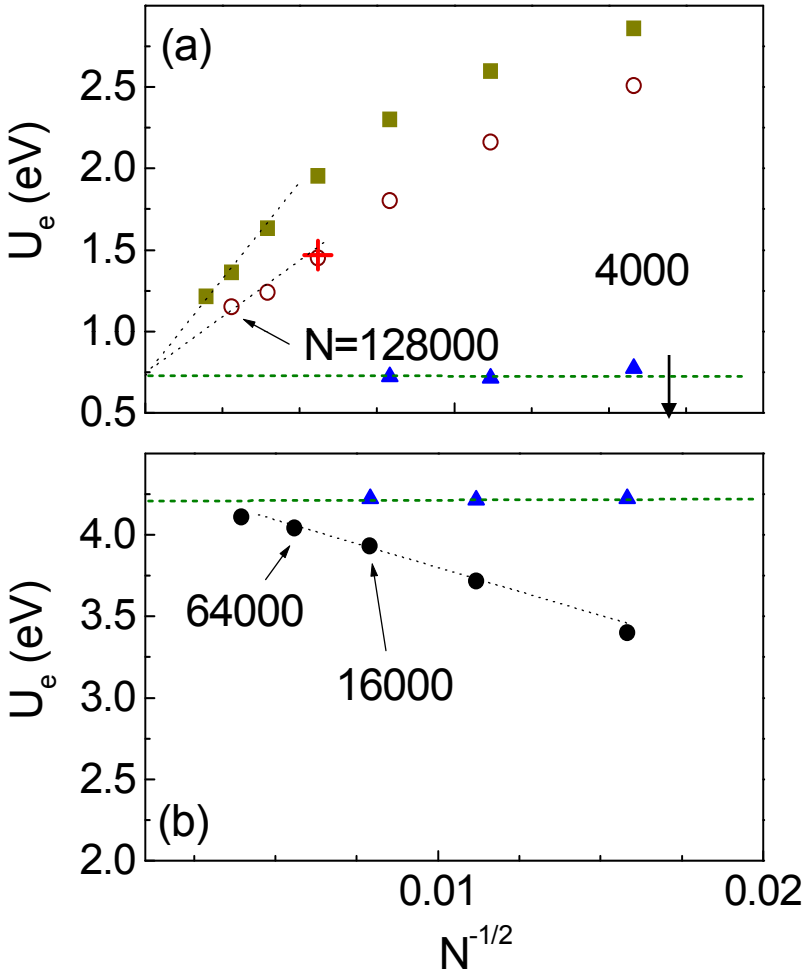
$$Q_e = N_g \int v_{ex} |\vec{v}_e| \sigma_t f_e \, \mathrm{d}\vec{v}_e - \nu_e \int v_{ex} f_e \, \mathrm{d}\vec{v}_e,$$

$$Q_i = N_g \int v_{ix} |\vec{v}_i| \sigma_r f_i \, \mathrm{d}\vec{v}_i - \nu_i \int v_{ix} f_i \, \mathrm{d}\vec{v}_i,$$

$$\nu_e = \frac{N_g \int |\vec{v}_e| \sigma_t f_e \, \mathrm{d}\vec{v}_e}{\int f_e \, \mathrm{d}\vec{v}_e}, \qquad \nu_i = \frac{N_g \int |\vec{v}_i| \sigma_r f_i \, \mathrm{d}\vec{v}_i}{\int f_i \, \mathrm{d}\vec{v}_i},$$

In usual fluid approach terms Q_e and Q_i are supposed to be zero, which is correct only for the constant scattering frequencies

Comparison of electron energy calculated with three different PIC-MCC methods:



Electron energy as function of ***N*** $^{-1/2}$*,*
where N is the number of psevdo particle

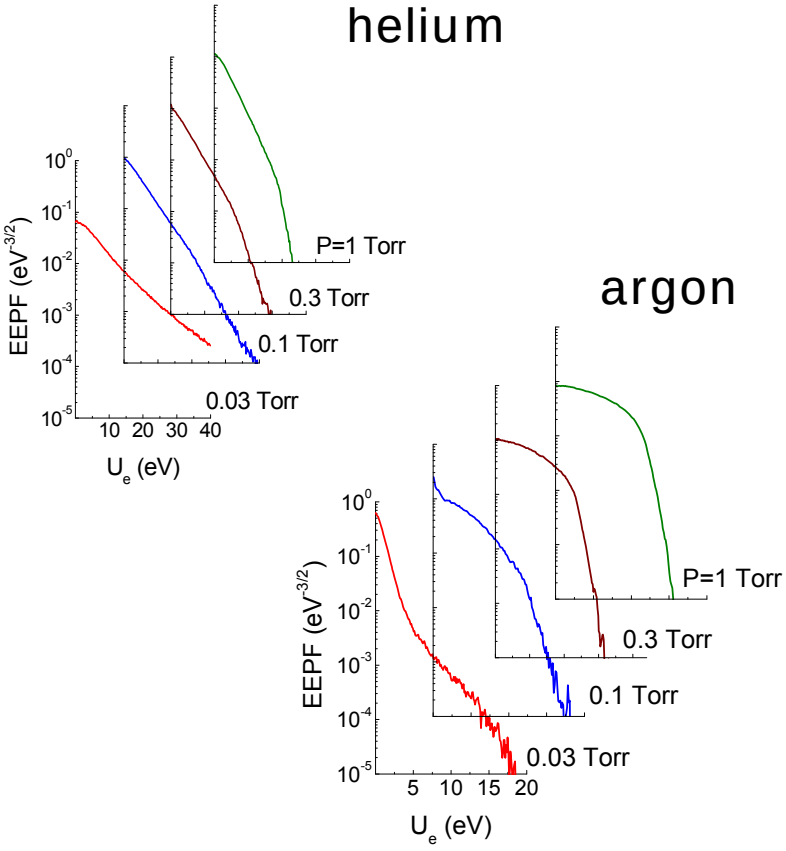
1) standard PIC-MCC,
2) PIC-MCC with space smoothing
3) combined PIC-MCC

In Figure:
P = 0.1 Torr ***(a)*** *and 0.3 Torr* ***(b),***
inter-electrode distance 6.7 см,
13,56 MHz discharge in argon,
j=2.65mA/cm^2

Standard PIC-MCC (■)
PIC-MCC with space smoothing (○)
combined PIC-MCC (▲)
┼ Birdsall, 1991.
- - - - - Godyak, 1986

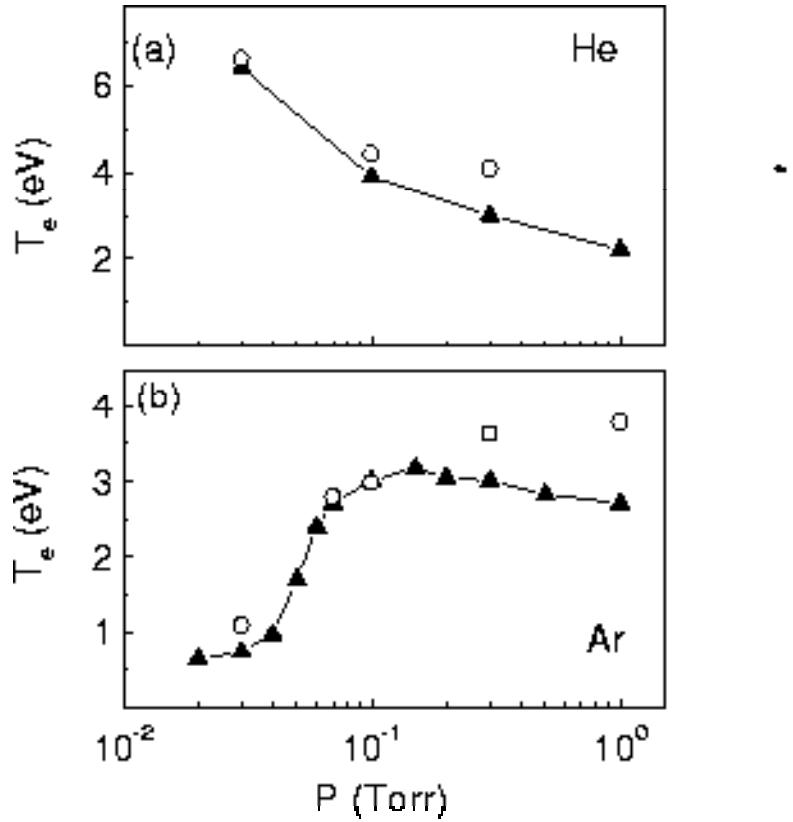
Electron energy distribution function

Simulation



○ - *experiment, Godyak, 1986, d=6.7 см, j=1мА/см^2*

▲ - *calculation*



Exp: Godyak et al, PSST (1992)
Sim: Schweigert, Schweigert, PSST,2004

Electron temperature in discharge center ver gas pressure in He and Ar

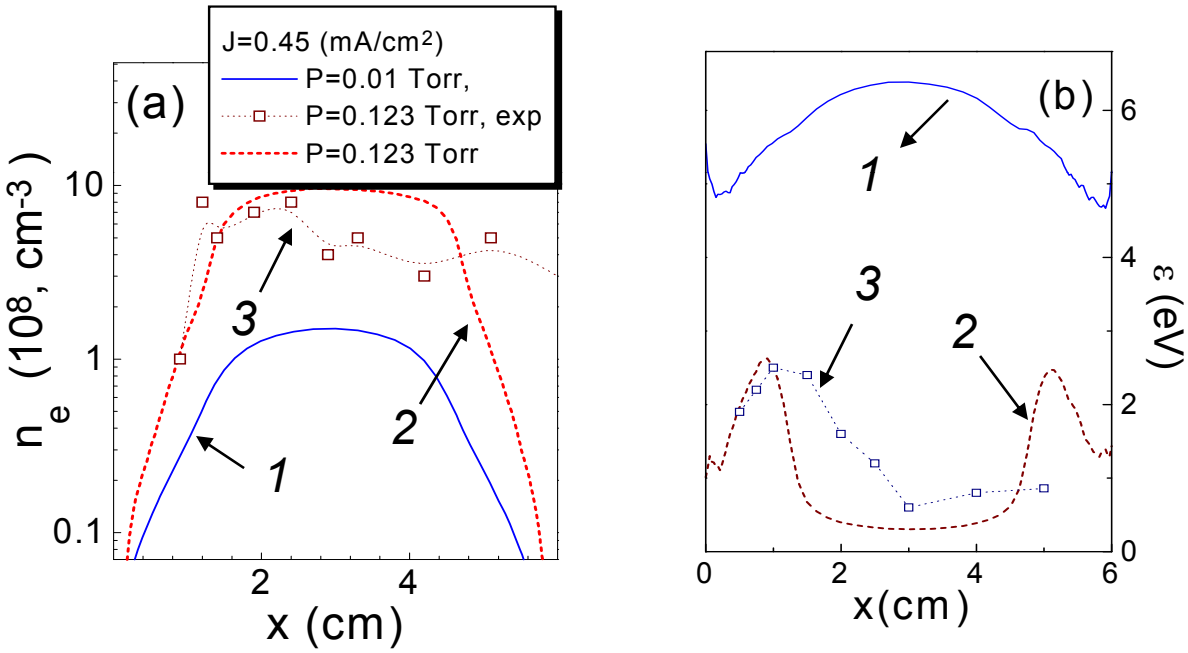
Transition between different regimes in methane. Simulations with combined PIC MCC.

Table 1. Electron–methane reactions involved in Monte-Carlo collision simulation.

	Reaction	Energy threshold (eV)
	Vibrational excitation	
1	$CH_4 + e = CH_4^* + e$	0.162
2	$CH_4 + e = CH_4^* + e$	0.361
	Dissociation	
3	$CH_4 + e = CH_3 + H + e$	8.0
4	$CH_4 + e = CH_2 + 2H + e$	8.0
	Ionization	
5	$CH_4 + e = CH_4^+ + 2e$	12.6
6	$CH_4 + e = CH_3^+ + H + 2e$	14.3

Two modes of discharge operation

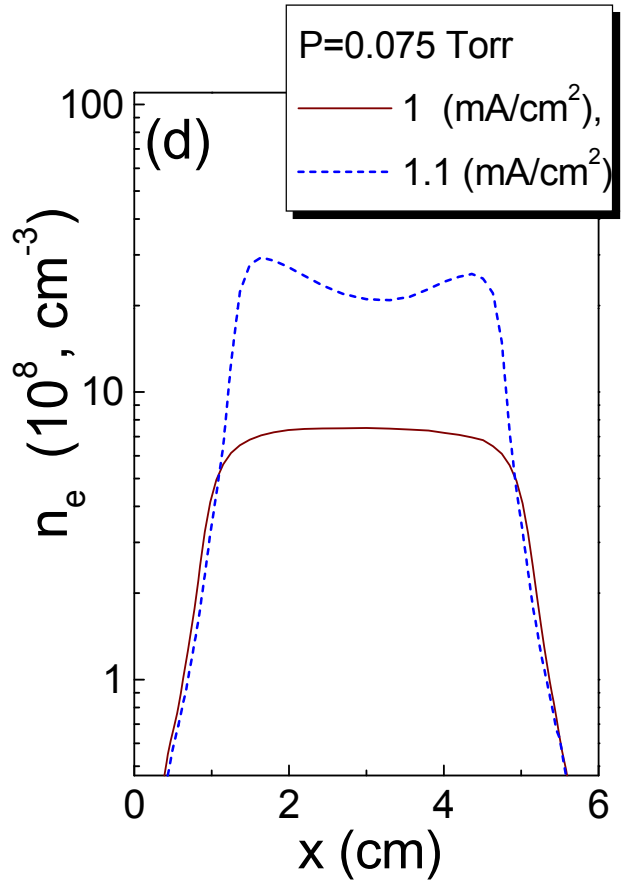
I. Schweigert, JETP 126, 4(10), (2004)
I. Schweigert, Phys. Rev. Lett. 92(15), 155001 (2004)



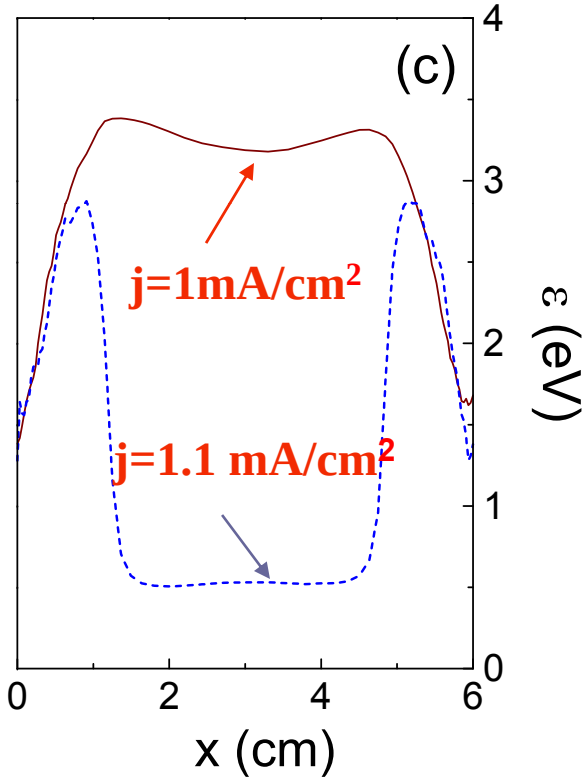
Averaged electron density (a) and electron energy (b) for P=10 mTorr (1) u 123 mTorr (2), j=0.45 mA/cm^2.
Volume mode is at P=10 mTorr (1), active sheath mode at P=123 mTorr (2), Sugai experiment, 1990 (3) P=123 mTorr.

Transition between two modes

Electron density



Electron energy



Small variation of discharge current (from 1mA/cm^2 to j=1mA/cm^2) induces transition between two modes

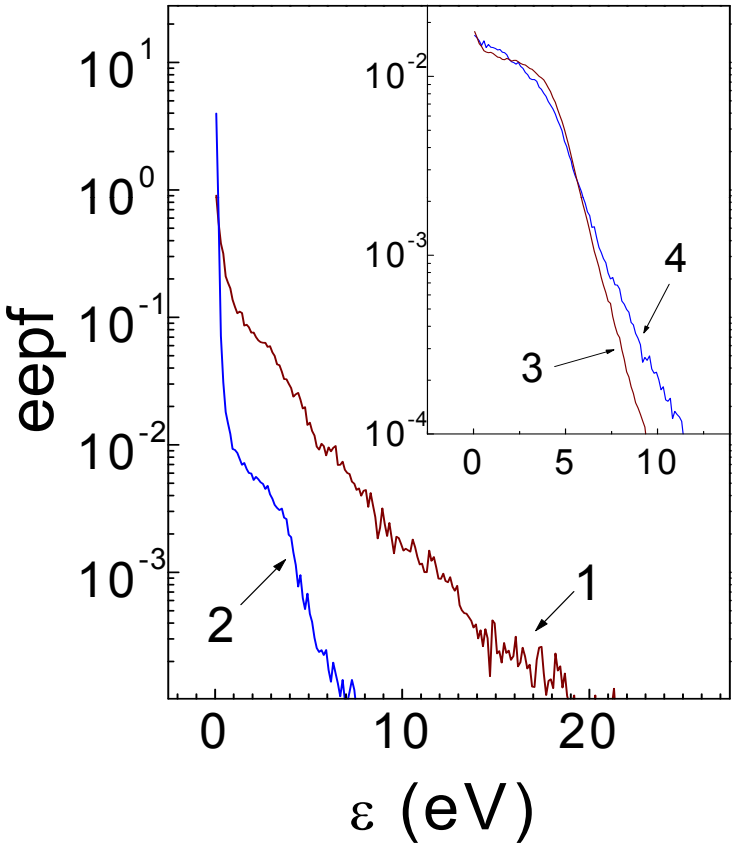
Electron energy probability function

In figure: EEPF in AS mode
j=1.1 mA/cm^2 (AS)
at x=0.86 cm (1)
at x=3 cm (2),

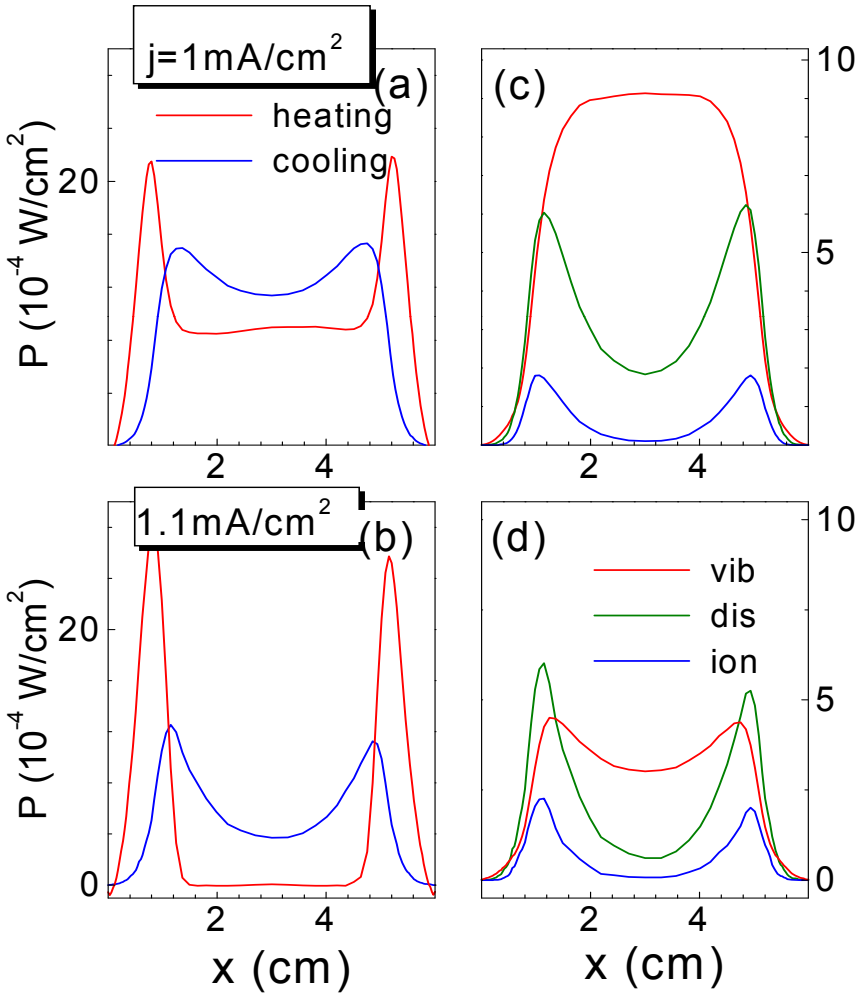
In figure insert: EEPF in VD mode
j=1 mA/cm^2 (VD)
at x=1.3 cm (3) and
at x=3 cm (4) (insert)

P=0.075 Torr, d=6 cm

1, 3 in the sheath, 2, 4 in the center of discharge



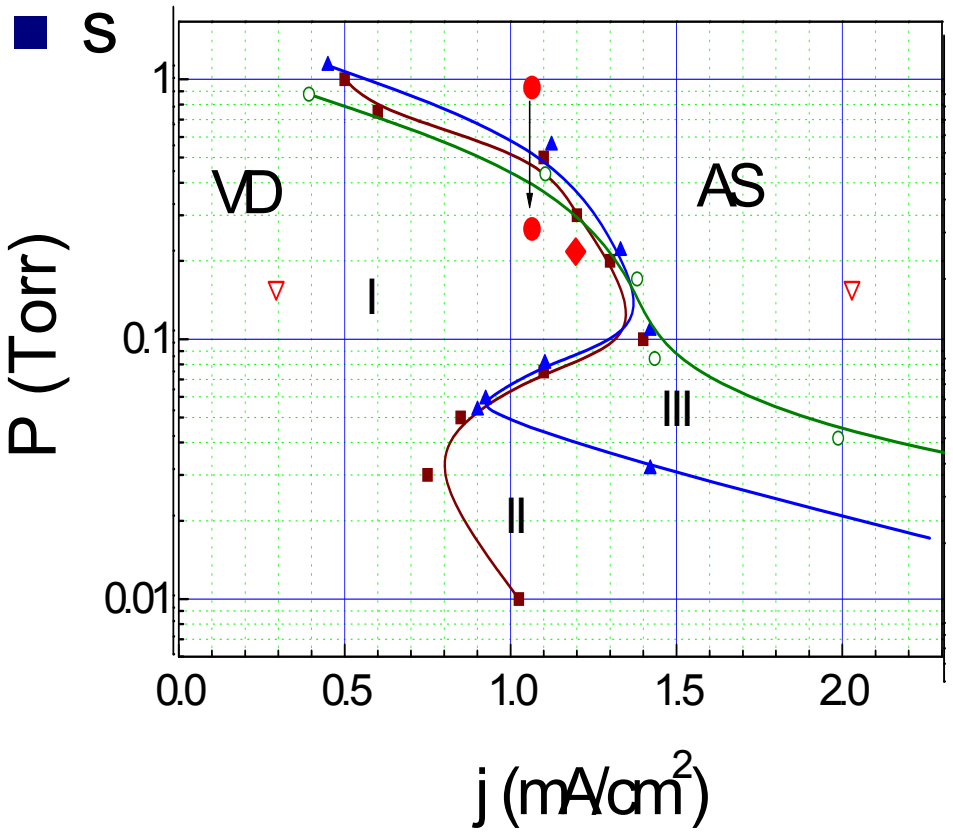
Formation of radicals CH_2, CH_3



Electron heating in electrical field E (a), (b) energy input in excitation of vibration states of CH_4 molecules (red), dissociation (green) and ionization (blue) (c), (d).

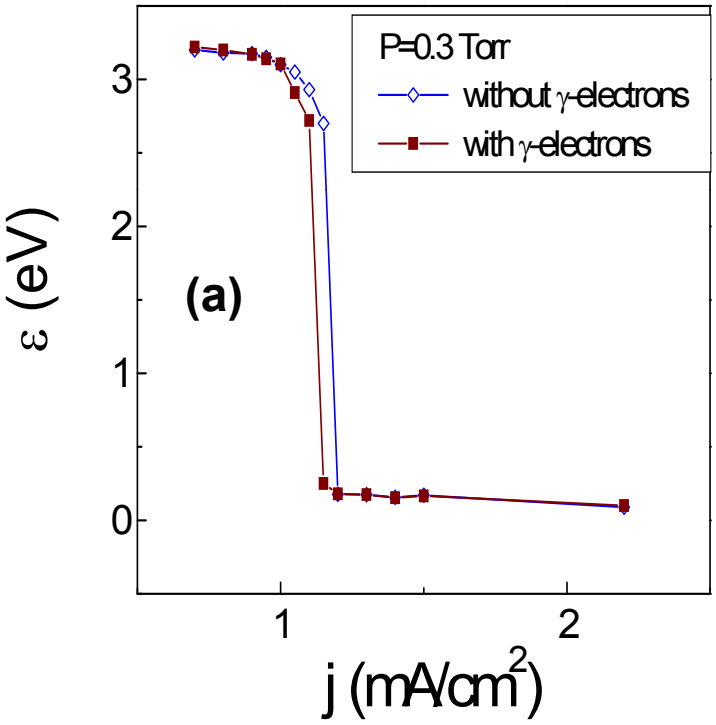
Current density:
j = 1 mA/cm² (a),(c),
1.1 mA/cm² (b),(d).
P= 75 mTorr.

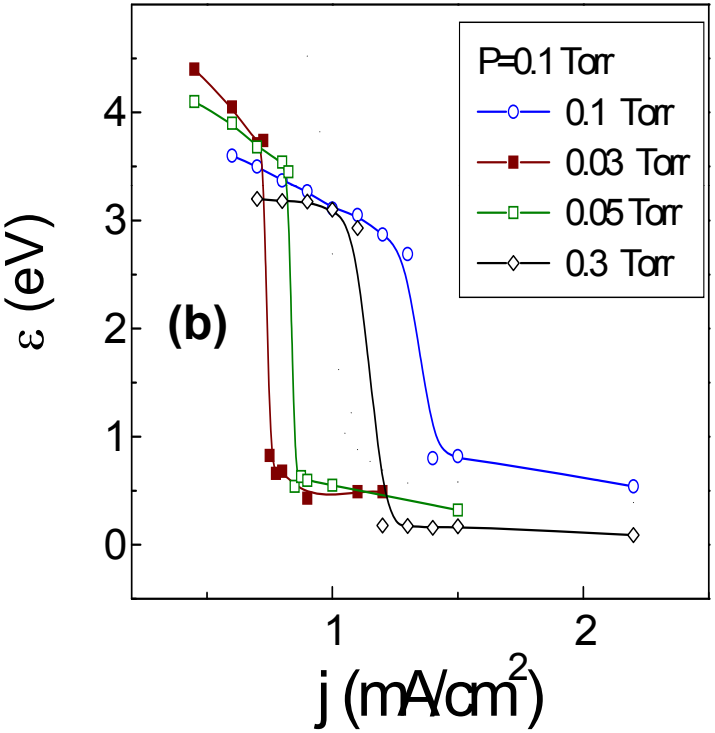
Phase diagram of different regimes in ccrf discharge in methane



Lines separate VD and AS modes for different inter-electrode gaps:
d = 6 cm - (■)
d = 4 см – (▲)
d = 3 см – (○)
Symbols are comparison with other authors:
V. Ivanov, 2002 (▽)
Nagayama, 1998 (♦),
Bera, 1999 (●)

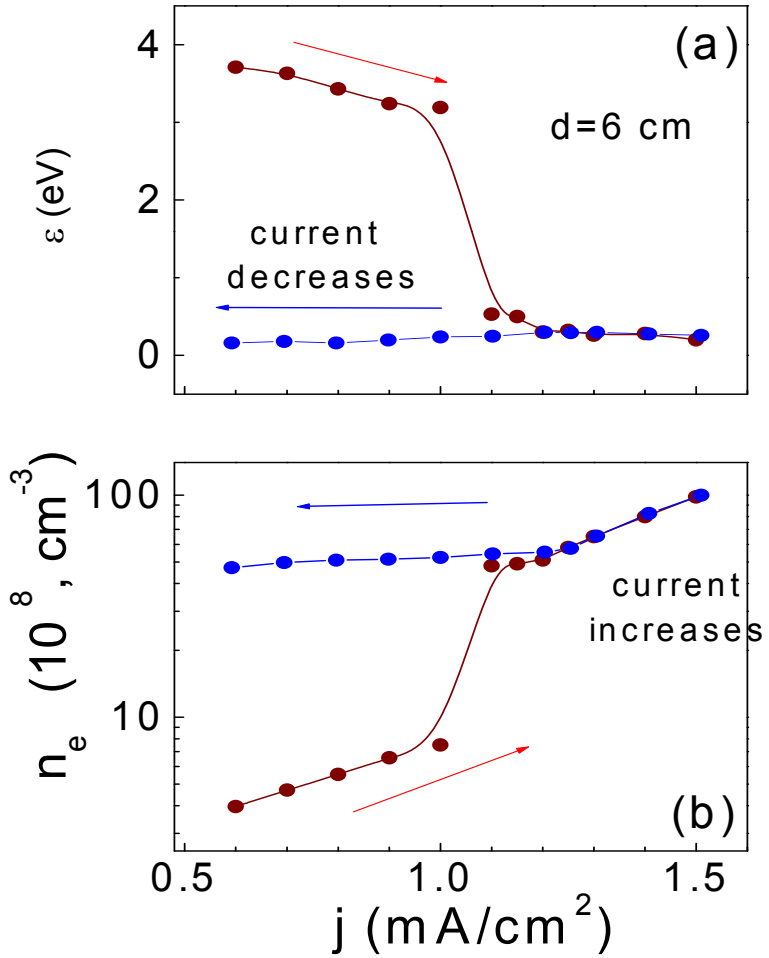
Is α-γ mechanism responsible for transition between different modes?





Electron energy in the center of discharge as function of discharge current for γ=0 and 0.2 (a) and for different gas pressures for γ=0 (b).

Hysteresis in ccrf discharge in methane



Electron energy (a) and electron density (b) in center of discharge as function of discharge current for 75 мTopp. Hysteresis was observed in experiment of C. Bohm, J. Perrin, 1991 в SiH_4 ccrf discharge.

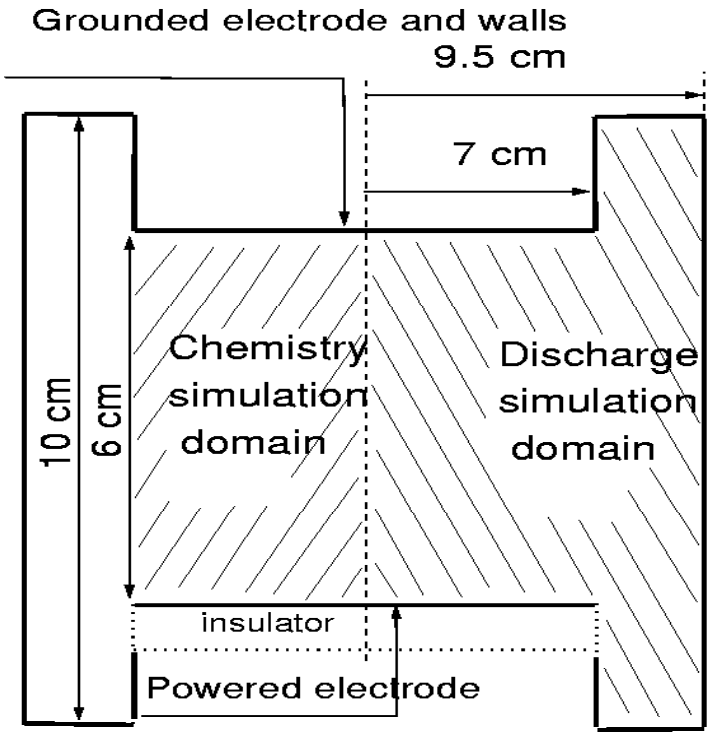
Two dimensional fast PIC MCC simulations

Acceleration algorithm by V. Schweigert, Plasma Source Sci. Technol. 8, B1,(1999)

I.V. Schweigert, A. Alexandrov, *Transition Between Different Modes of a Capacitively Coupled Radio Frequency Discharge in CH4 in One and two dimensional PIC-MCC simulations. IEEE Transaction on Plasma Science*, 33, 615-622 (2005)

A. Alexandrov, I.V. Schweigert, *Two-dimensional PIC-MCC simulations of a capacitively coupled radio frequency discharge in methane.* PSST, 14, 209-218 (2005)

Simulation of ccrf discharge in methane in reactor (2D axially symmetrical model)



Reactor geometry and calculation cell

Kinetic equations for electron and ion distribution functions:

$$\frac{\partial f_e}{\partial t} + \vec{v}_e \frac{\partial f_e}{\partial \vec{r}} - \frac{e\vec{E}}{m} \frac{\partial f_e}{\partial \vec{v}_e} = J_e, \quad n_e = \int f_e d\vec{v}_e,$$

$$\frac{\partial f_i}{\partial t} + \vec{v}_i \frac{\partial f_i}{\partial \vec{r}} + \frac{e\vec{E}}{M} \frac{\partial f_i}{\partial \vec{v}_i} = J_i, \quad n_i = \int f_i d\vec{v}_i,$$

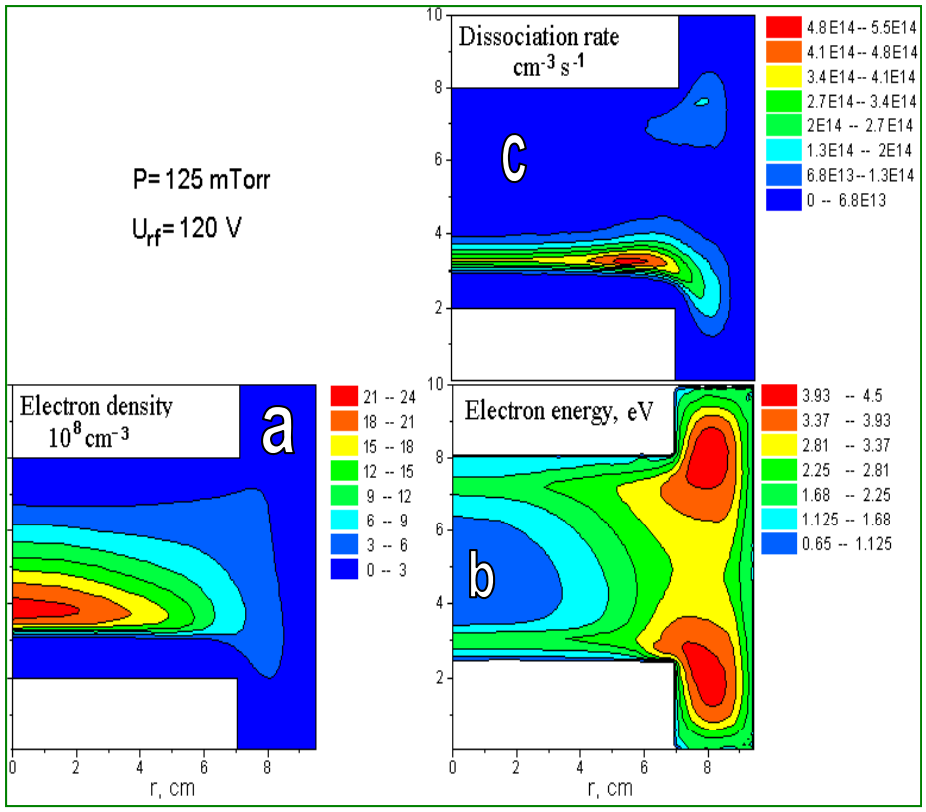
Poisson equation:

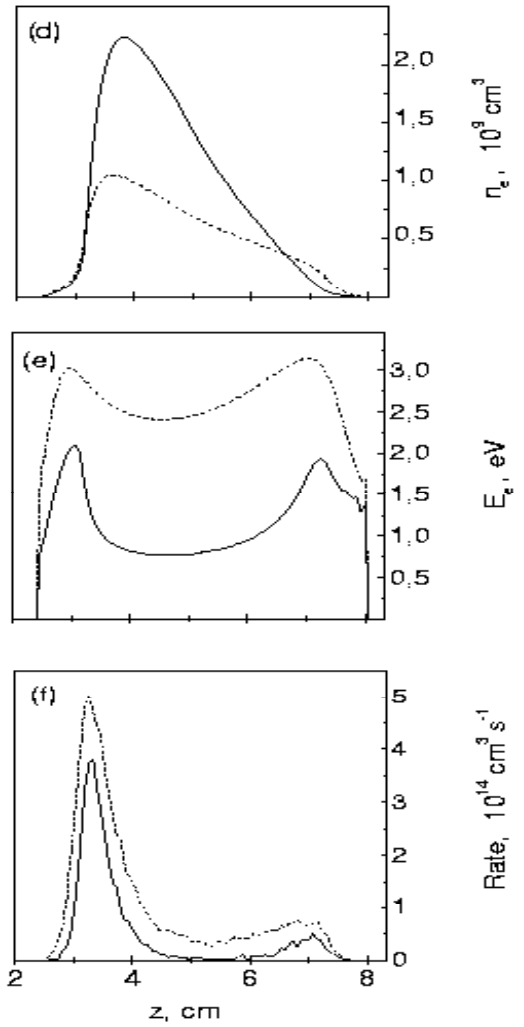
$$\Delta\phi = 4\pi e(n_e' - n_i'), \quad E = -\frac{\partial \phi}{\partial \vec{r}}.$$

Table 2. Chemical reactions taken into account in the g
chemistry model.

N	Reaction	Rate constant k_N ($m^3\ s^{-1}$)
	Electron–methane	
1	$CH_4 + e = CH_3 + H + e$	Obtained by MCC
2	$CH_4 + e = CH_2 + 2H + e$	Obtained by MCC
3	$CH_4 + e = CH_4^+ + 2e$	Obtained by MCC
4	$CH_4 + e = CH_3^+ + H + 2e$	Obtained by MCC
	Ion–methane	
5	$CH_4 + CH_4^+ = CH_5^+ + CH_3$	1.5×10^{-15}
6	$CH_4 + CH_3^+ = C_2H_5^+ + H_2$	1.2×10^{-15}
	Radical reactions	
7	$CH_3 + CH_3 = C_2H_6$	8×10^{-17}
8	$CH_3 + H = CH_4$	1.38×10^{-16}
9	$CH_2 + H = CH + H_2$	2.7×10^{-16}
10	$CH_2 + CH_4 = CH_3 + CH_3$	1.5×10^{-18}
11	$CH_2 + CH_2 = C_2H_2 + H_2$	5.3×10^{-17}
12	$CH_2 + CH_3 = C_2H_4 + H$	10^{-16}
13	$CH + CH_4 = C_2H_4 + H$	10^{-16}
14	$CH + CH_4 = C_2H_5$	10^{-16}
15	$C_2H_5 + H = CH_3 + CH_3$	6×10^{-17}
16	$C_2H_5 + H = C_2H_4 + H_2$	3×10^{-18}
17	$C_2H_5 + CH_3 = C_3H_8$	4.2×10^{-18}

Active sheath mode

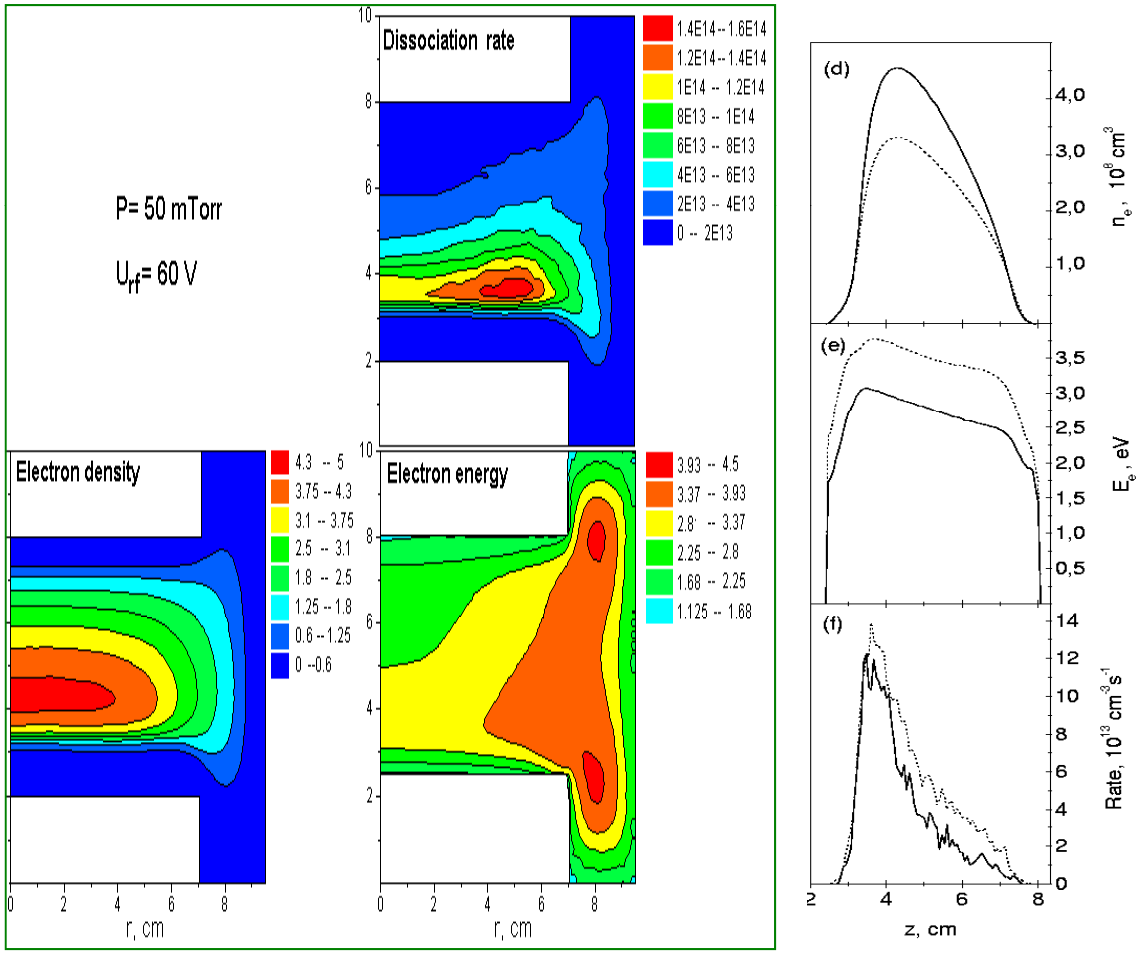




P=123 mTorr, U=120 V

(a, d) electron density, (b,e) electron energy, (c,f) CH4→CH3+H,.
In Figs (d-f) at r=0 (solid), r=6 cm (dashed)

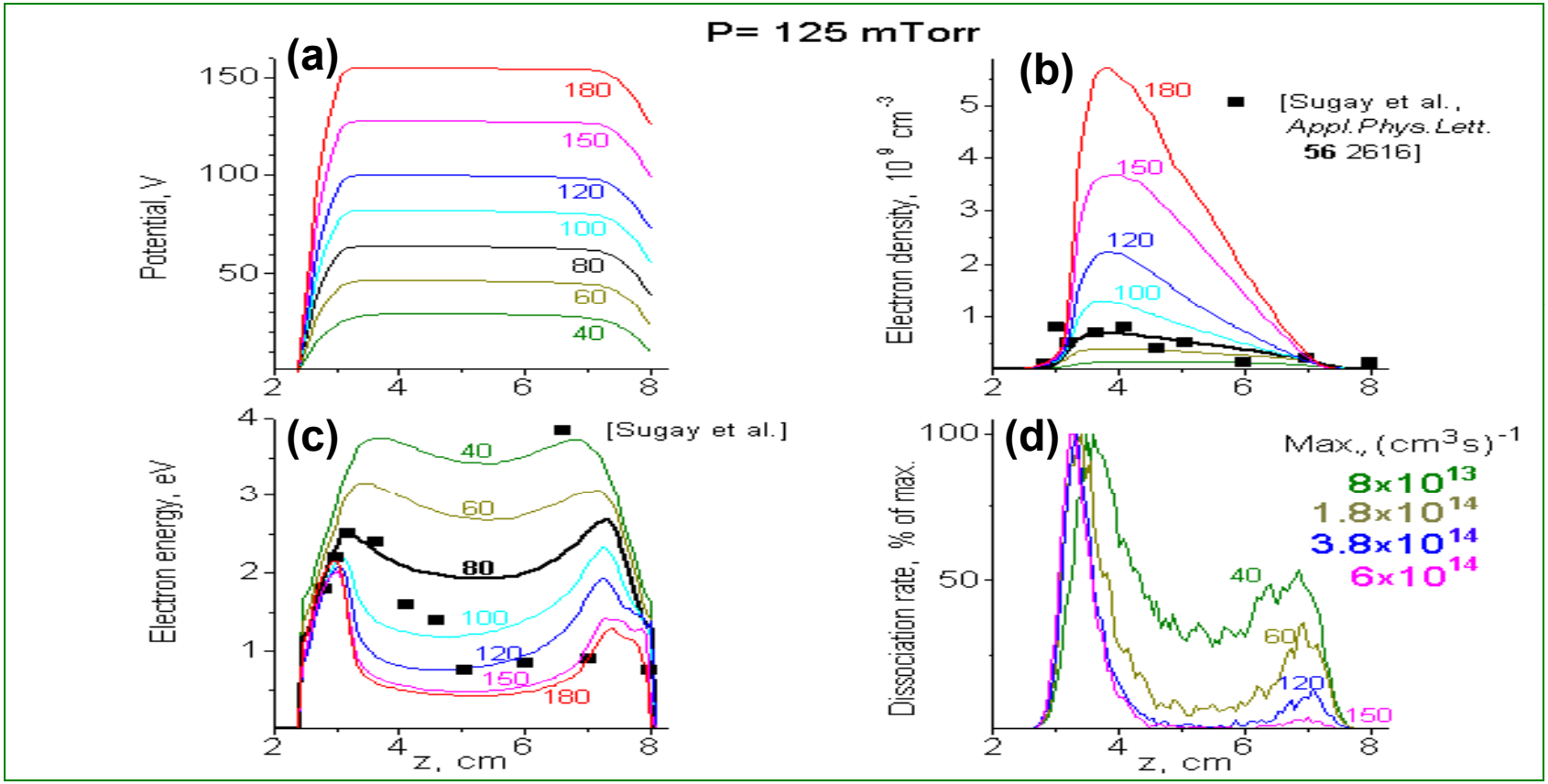
2D distributions of plasma parameters in volume dominated mode



P=50 mTorr, U=60 V

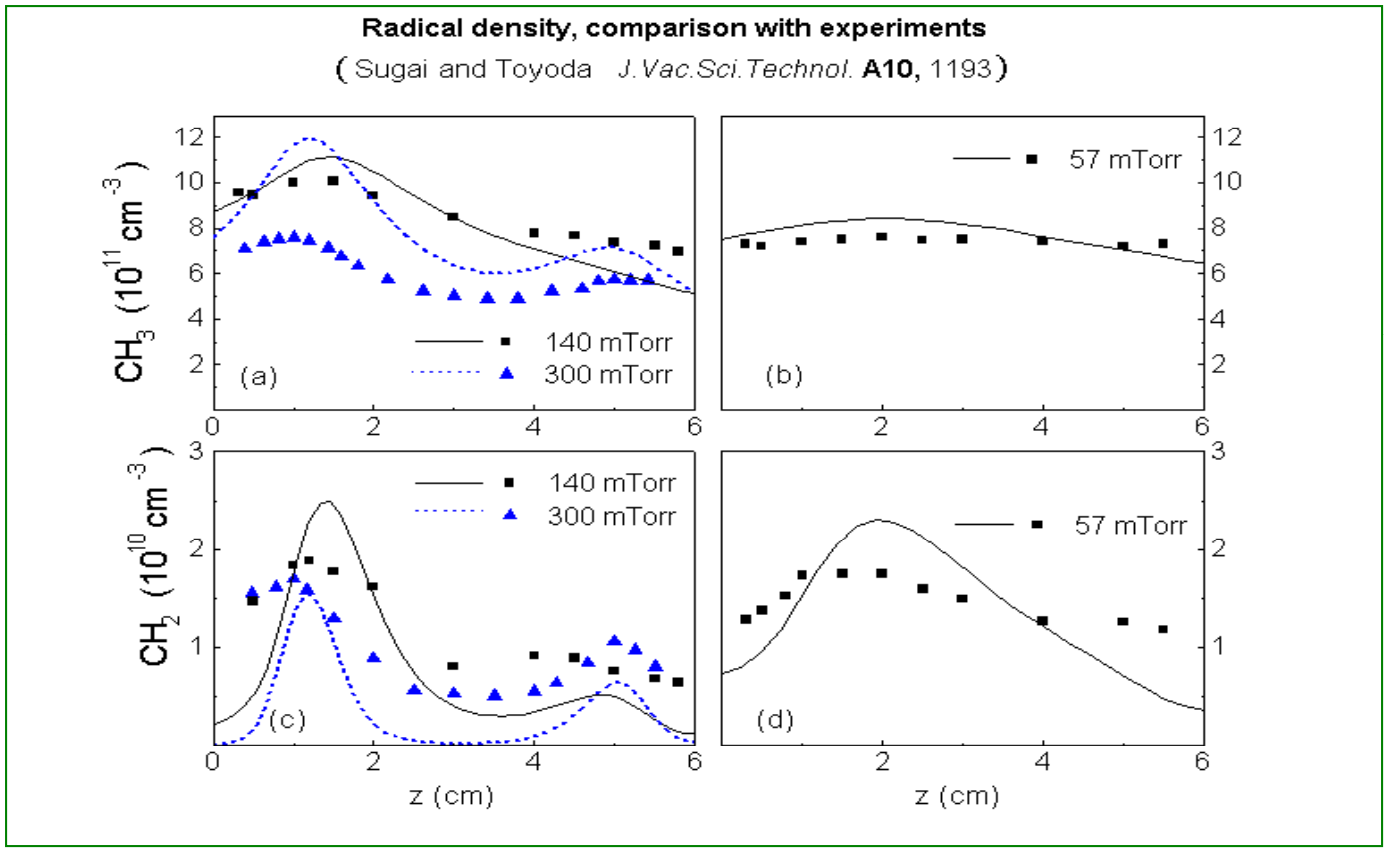
(a, d) electron density, (b,e) electron energy, (c,f) CH4→CH3+H. In Figs (d-f) at r=0 (solid), r=6 cm (dashed)

Transition between different modes in plasma reactor



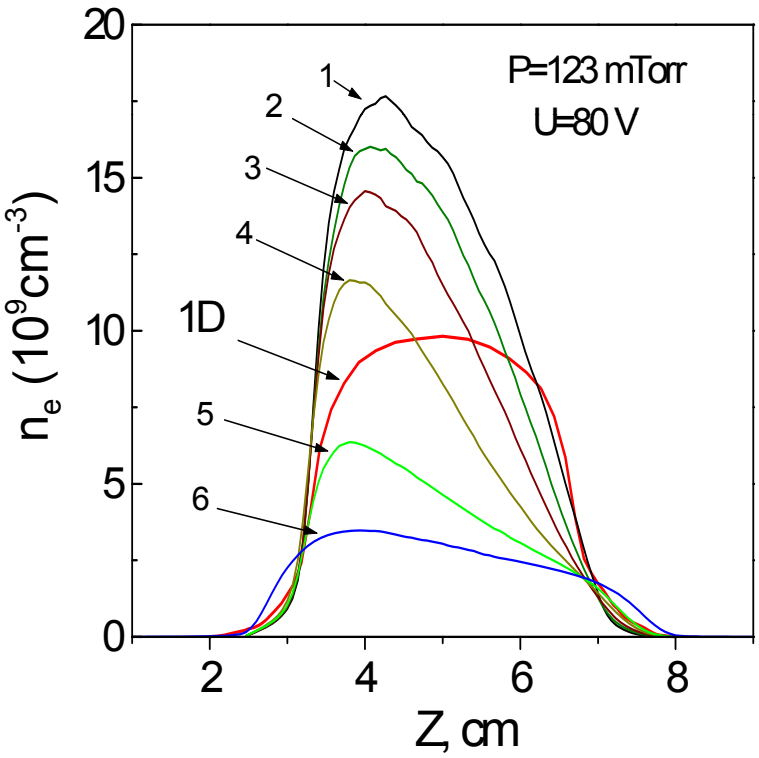
Averaged over rf cycles at r=0: (a) potential, (b) electron density (c) electron energy, (d) dissociation rate $CH_4 \rightarrow CH_3+H$ at P=123 mTorr, U=180-40 V, symbols - exp Sugai, 1990

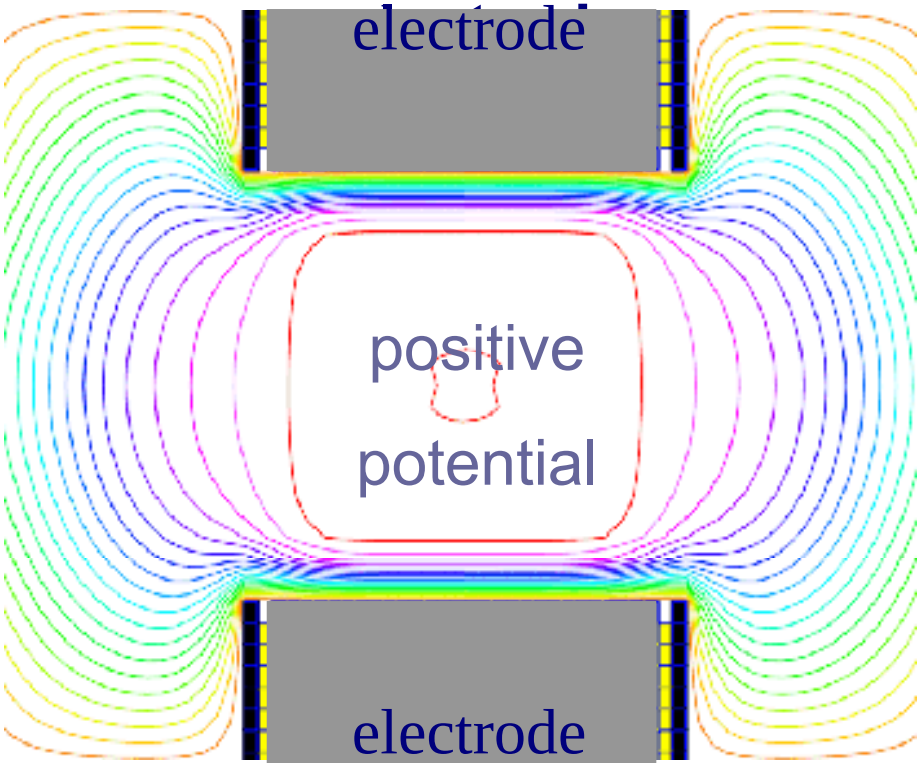
Radical distribution



Distribution of density of radical CH_3 u CH_2 for different gas pressures. Symbols experiment of Sugai, 1990

Comparison of 1D and 2D results





Conclusion

- Combined PIC MCC was developed for efficient simulation of low gas pressure discharge (in particular for gases with a deep Ramsauer minimum in the elastic scattering cross sections)
- We studied two regimes of the ccrf discharge glow in methane and constructed the P-j phase diagram to show the location of volume dominated and active sheath regimes.The critical values for transition have first been calculated for a wide range of j and P.
- It is shown that the transition between two modes of the CH_4 discharge is not related with α-γ transition as in the argon discharge.
- We have found the hysteresis in discharge behavior.