Gas Dynamics In High-Power Impulse Magnetron Sputtering

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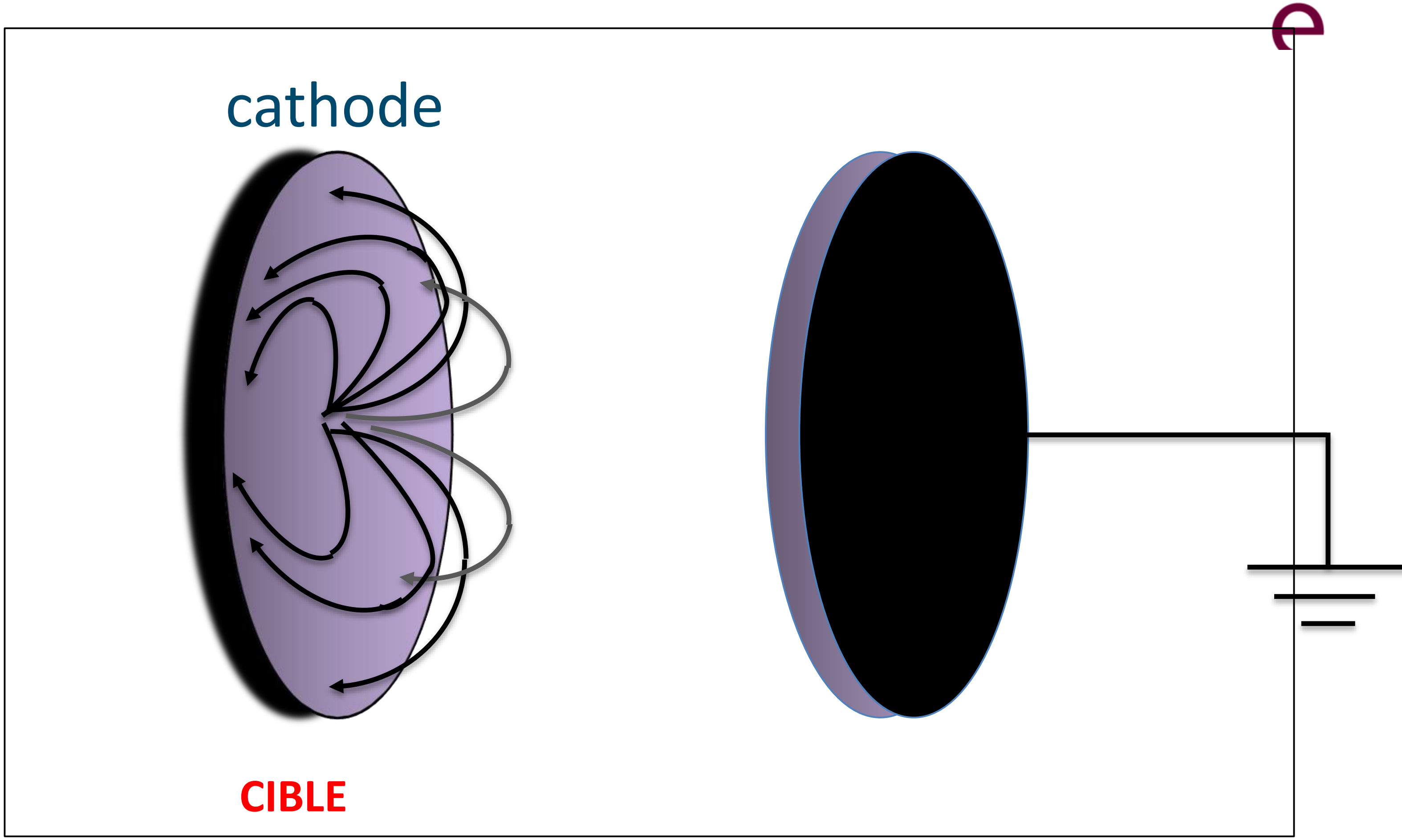
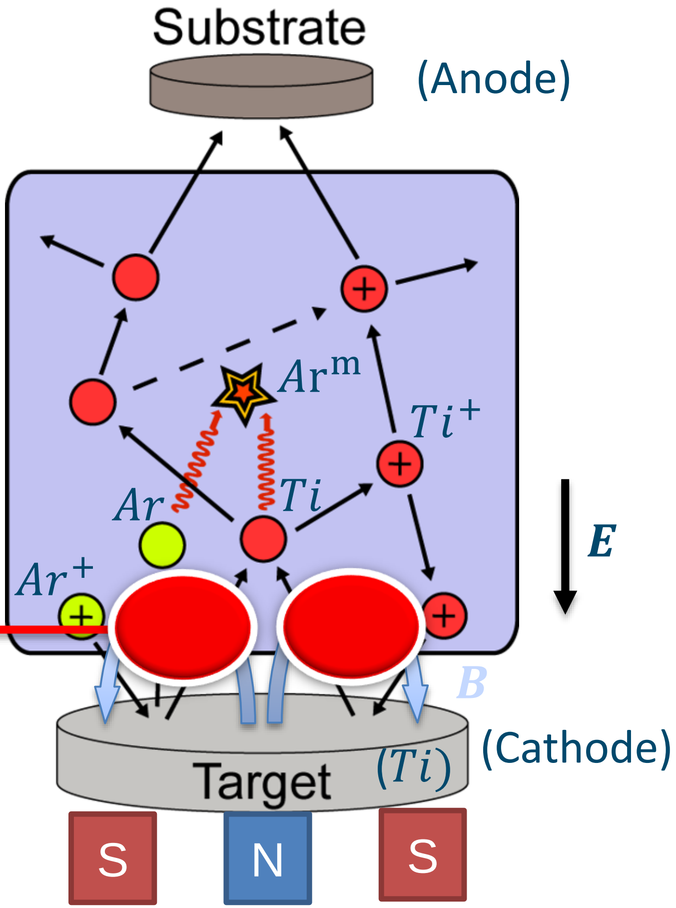
Magnetron discharge



(ions TARGET)

Sputtering:
using plasma to erode a target



High density Plasma

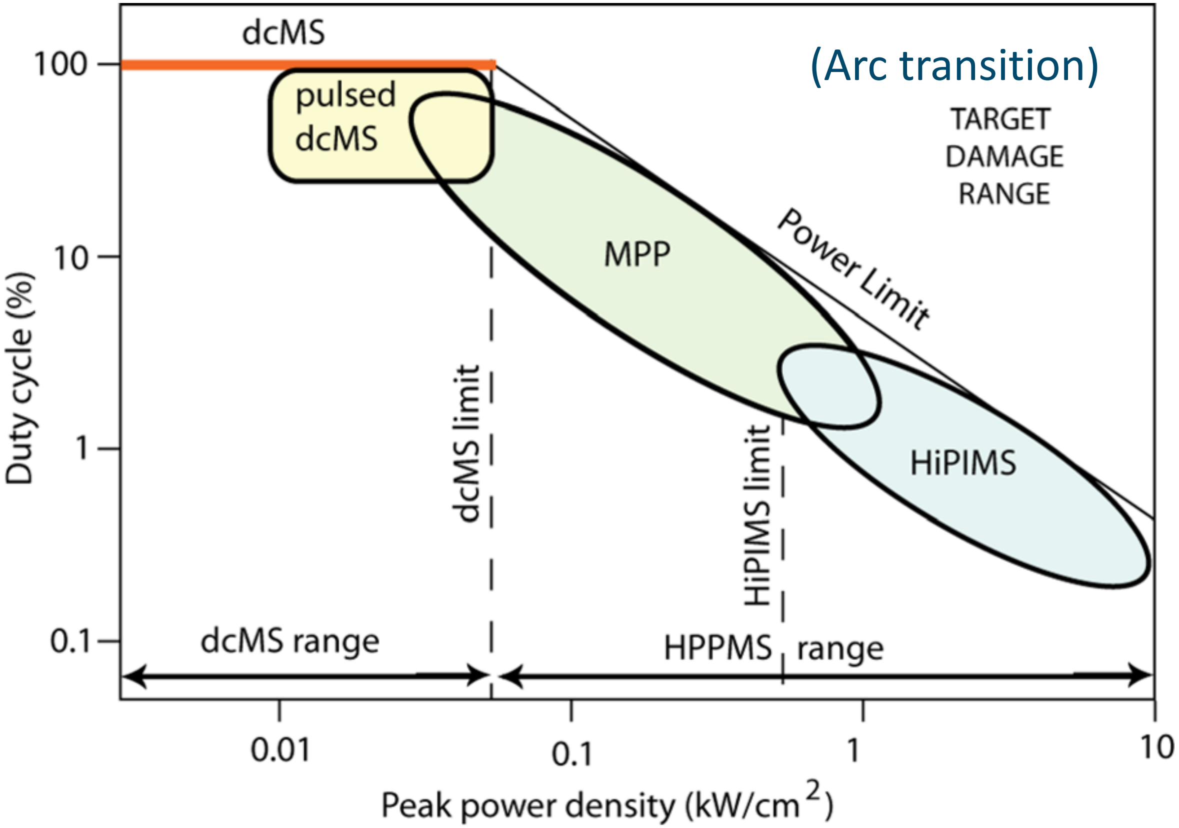
Ionization Region (IR)

Magnetron Sputtering:

- **B** field
- Confinement of e^-
- Low pressure & Better sputtering rate

High-Power Impulse Magnetron Sputtering (HiPIMS)

- As opposed to conventional dcMS
- low duty cycle and high peak power



Typical values:

$U_D = 500\sim1000\text{V}$

$\lambda_{pw} \sim 100\ \mu\text{s}$

$P = 0.1\sim2\ \text{Pa}$

$n_e > 10^{18}\ \text{m}^{-3}$

$j > 0.6\ \text{A cm}^{-2}$

Outlook

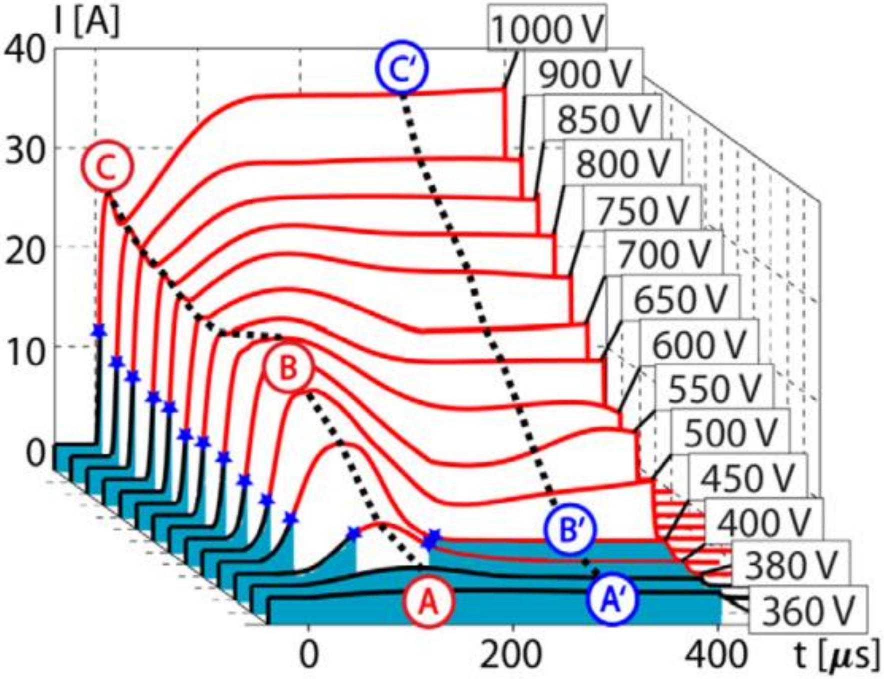
1. **HiPIMS plasma operation**
   - self-sputtering regime *versus* gas recycling regime
2. **Neutral gas dynamics in HiPIMS plasmas**
   - 0D IRM (Ionization Region Model)
   - 2D DSMC
3. **Ion dynamics in HiPIMS plasmas**
   - 0D IRM
   - 2D OHiPIC (Orsay High density Particle in Cell)
4. **Conclusions**

1- HiPIMS plasma operation

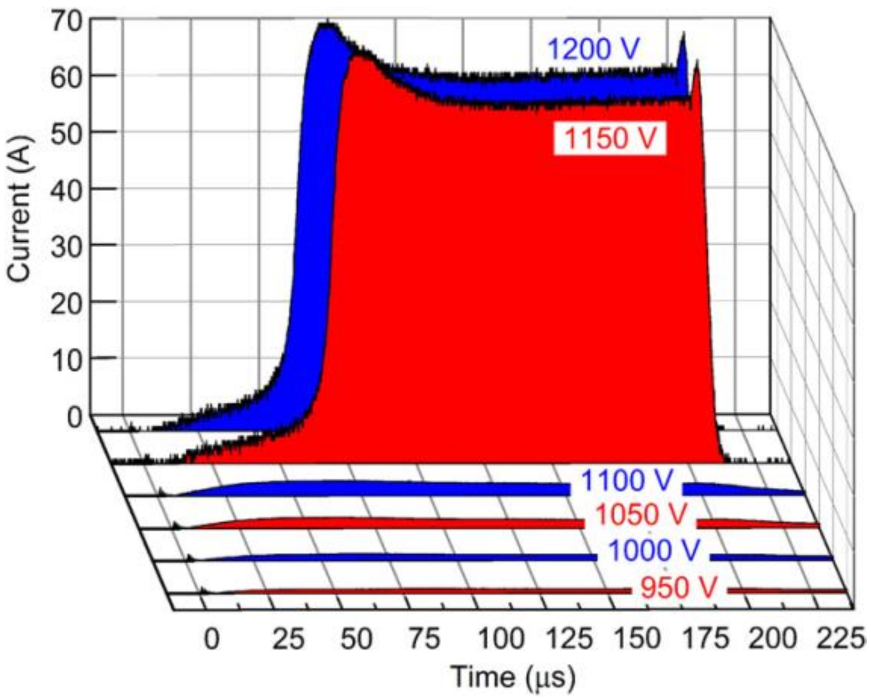
self-sputtering regime *vs.* gas recycling regime

I(*t*) *versus V* in HiPIMS?

Al target



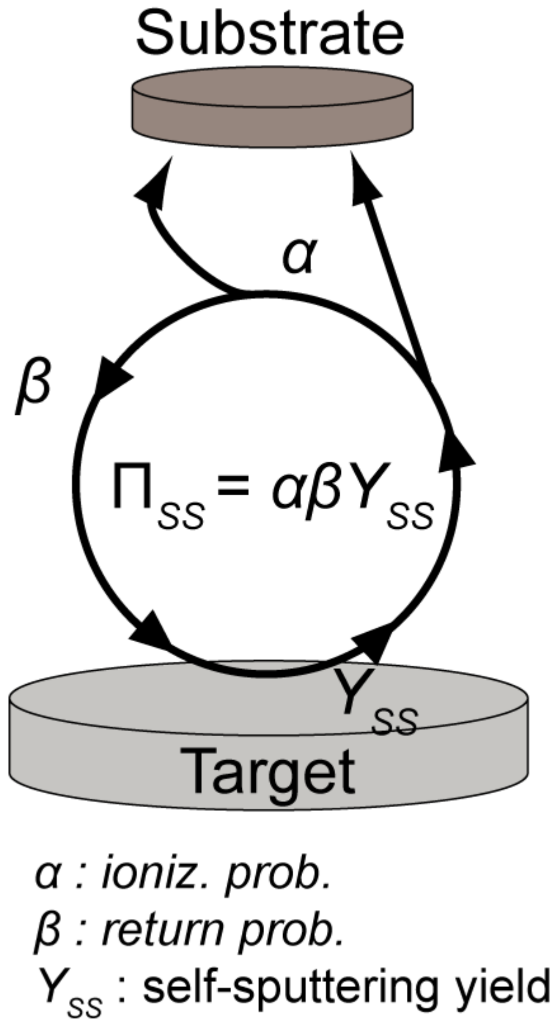
C target



Huo *et al*., PSST **23** 025017 (2014)

A. Anders *et al*., J. Phys. D. Appl. Phys. 45 012003 (2012)

Initial picture



α : ioniz. prob.
β : return prob.
Y_{SS} : self-sputtering yield

Self-sputtering parameter
Cu @ Ar

$\alpha_t = 0.8$,

$\beta_t = 0.7$,

and

$Y_{SS} = 0.5$

$$\pi_{SS} = \alpha_t \beta_t Y_{SS}.$$

If $\Pi_{SS} > 1$ ➡ Self-sputtering regime!

A. Anders *et al.*, J. Phys. D. Appl. Phys. 45 012003 (2012)

General picture

GIM – Generalized Ionization Model

Self-sputtering parameter

$\alpha_t = 0.8$,

$\beta_t = 0.7$,

and

$Y_{SS} = 0.5$

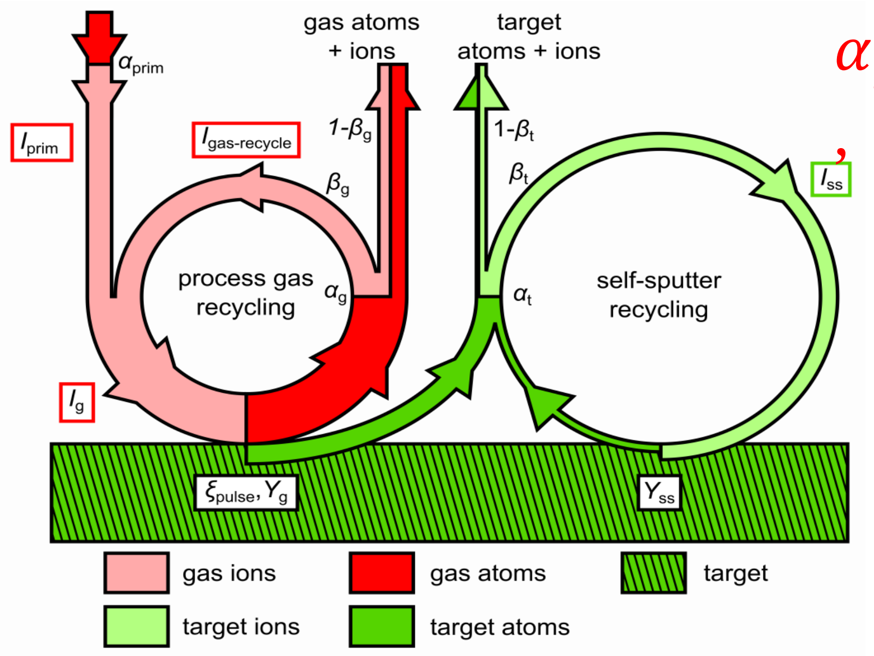
$$\pi_{SS} = \alpha_t \beta_t Y_{SS}.$$

Gas-sputtering parameter

$\alpha_{prim} = 1, \xi_{pulse} = 1,$

$\alpha_g = 0.7, \beta_g = 0.7, Y_g = 0.4$,

$$\pi_g = \alpha_g \beta_g \xi_{pulse}$$



A. Anders *et al.*, J. Phys. D. Appl. Phys. 45 012003 (2012)

Gudmundsson *et al.*, PSST **25** 065004 (2016)

Ions recycling

Gas-recycling

$$I_{\text{gas-recycle}} = I_{\text{prim}} \frac{\pi_g}{1-\pi_g}$$

$$0 < a < 1, \quad \sum_{n=1}^{\infty} a^n = a/(1-a)$$

***Target** current* by **gas ions**

$$I_g = I_{\text{prim}} + I_{\text{gas-recycle}} = I_{\text{prim}}\left(1 + \frac{\pi_g}{1-\pi_g}\right)$$

Metal-recycling

$$I_{SS} = I_g\left(\frac{Y_g}{Y_{SS}} \frac{\pi_{SS}}{(1-\pi_{SS})}\right)$$

***Target** current* by **metal ions**

Total discharge current on the target I_D

$$I_D \approx I_i = I_{\text{prim}} + I_{\text{gas-recycle}} + I_{SS} = I_{\text{prim}}\left(1 + \frac{\pi_g}{1-\pi_g}\right)\left(1 + \frac{Y_g}{Y_{SS}} \frac{\pi_{SS}}{(1-\pi_{SS})}\right)$$

$$\equiv \boldsymbol{I_{prim}\Pi_{gas-recycle}\Pi_{SS-recycle}}.$$

Brenning *et al.*, PSST 26 (2017) 125003

Relative contribution to I_D

Dimensionless parameters

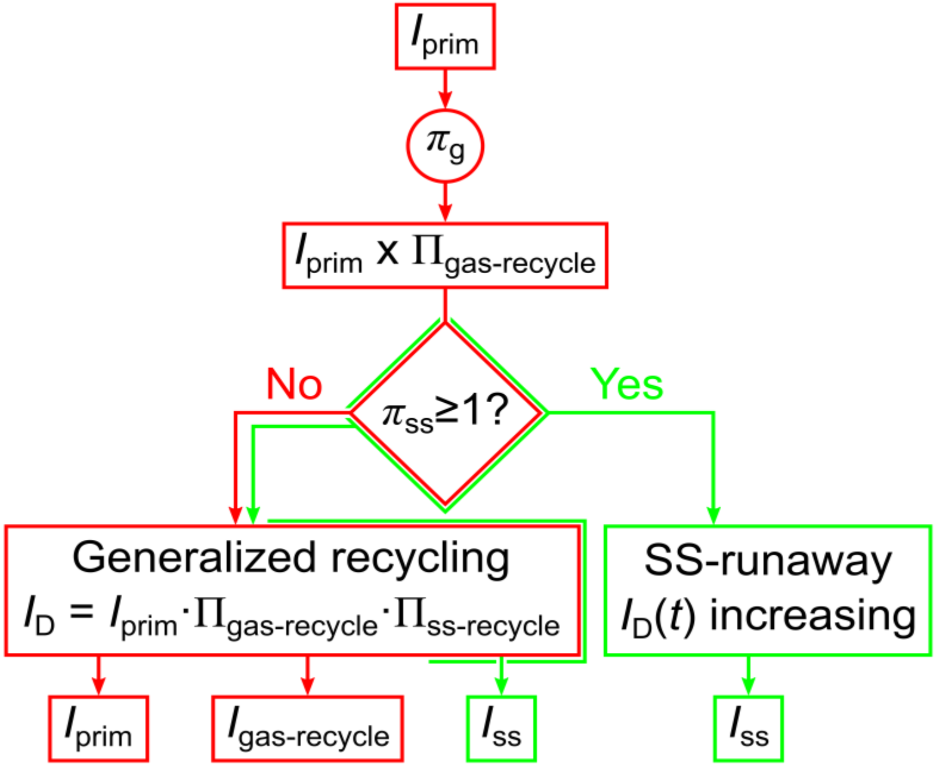
$$\frac{I_{\mathrm{gas-recycle}}}{I_{\mathrm{prim}}} = \frac{\alpha_{\mathrm{g}}\beta_{\mathrm{g}}}{1-\alpha_{\mathrm{g}}\beta_{\mathrm{g}}},$$

$$\frac{I_{\mathrm{SS}}}{I_{\mathrm{prim}}} = \left(1+\frac{\alpha_{\mathrm{g}}\beta_{\mathrm{g}}}{1-\alpha_{\mathrm{g}}\beta_{\mathrm{g}}}\right)\left(\frac{Y_{\mathrm{g}}}{Y_{\mathrm{SS}}}\frac{\alpha_{\mathrm{t}}\beta_{\mathrm{t}}Y_{\mathrm{SS}}}{(1-\alpha_{\mathrm{t}}\beta_{\mathrm{t}}Y_{\mathrm{SS}})}\right).$$

Critical current (empirical)

$$I_{\mathrm{crit}} = 0.38\, S_{\mathrm{RT}}p_{\mathrm{g}}$$

S_{RT} is the racetrack area in cm^2,
and p_{g} is the pressure in Pa

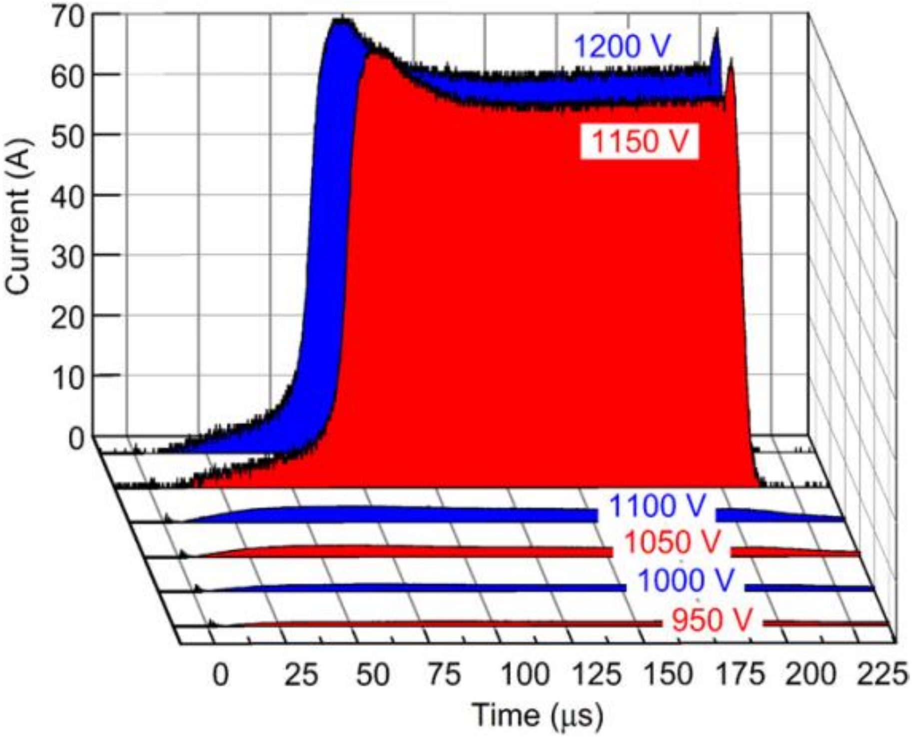


Huo *et al.*, PSST **23** 025017 (2014)

Brenning *et al.*, PSST 26 (2017) 125003

HiPIMS discharge regime analysis

Dimensionless contribution to I_D



Carbon

$$\boxed{I_{\mathrm{crit}} = 0.38\, S_{\mathrm{RT}} p_g = 3.9\ \mathrm{A}}$$

$$\Pi_{\mathrm{SS-recycle}} \leq \left(1 + \frac{Y_{\mathrm{g}}}{Y_{\mathrm{SS}}} \frac{Y_{\mathrm{SS}}}{1 - Y_{\mathrm{SS}}}\right)$$

$$= \left(1 + \frac{0.69}{0.52} \times \frac{0.52}{1-0.52}\right) = 2.4$$

$\Pi_{\mathrm{SS-recycle}} > 2$ means that the carbon-ion current I_{SS} is larger than the gas-ion current.

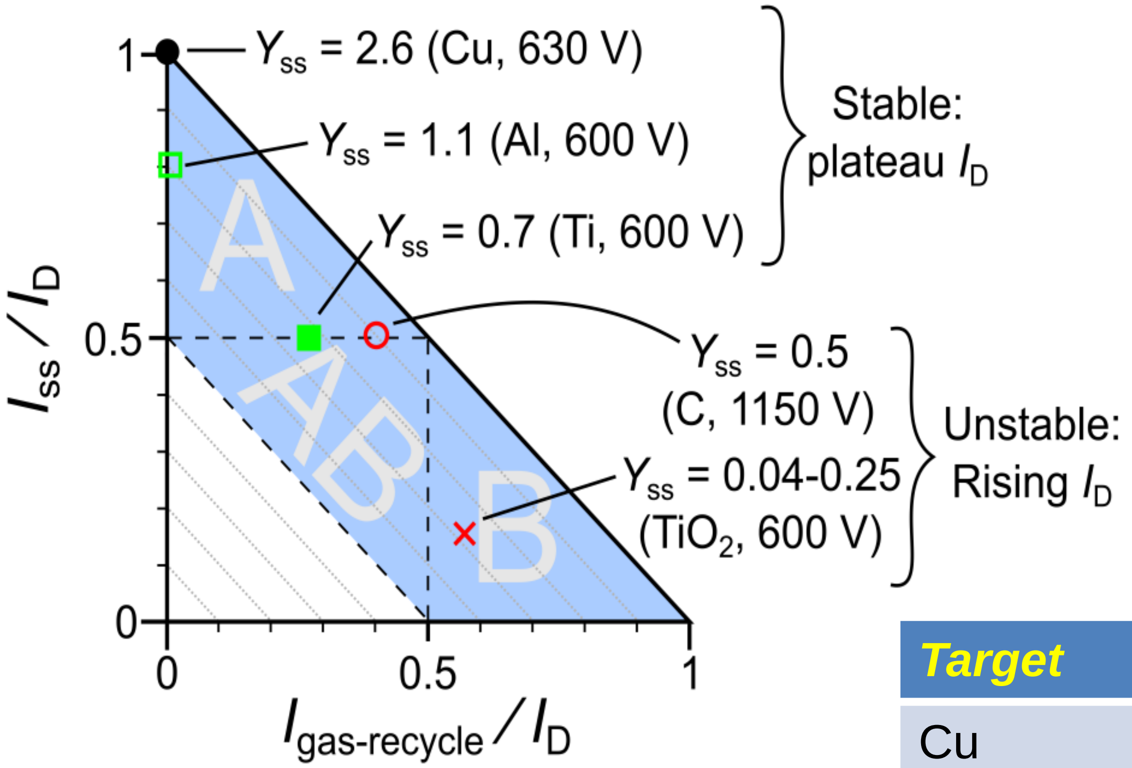
But I_D increases by **16** times!!!

⬇

Gas recycled is needed!!!

Brenning *et al.*, PSST 26 (2017) 125003

Other targets HiPIMS



C @ 1150 V does **not** operate in **self-sputtering regime**

Target	$I_{gas-recycle}/I_D$	I_{SS}/I_D
Cu	0	1
Al	0.2	0.8
Ti	0.27	0.5
TiO_2	0.54	0.16

Brenning *et al.*, PSST 26 (2017) 125003

Conclusion on HiPIMS operation

- High Current operation is not necesserily due to the self-sputtering in noble gas operation

- In reactive mode :

  * Metal mode: self-sputter recycling dominates, but only half of the discharge current is carried by recycled metal ions (Ti^+). Ar^+ ion current recycled even smaller is present

  * Poisoned mode: gas recycling is the dominant process

  2/3 of Ar^+ ions are recycled

- During gas recycling T_e increases and then I_D.

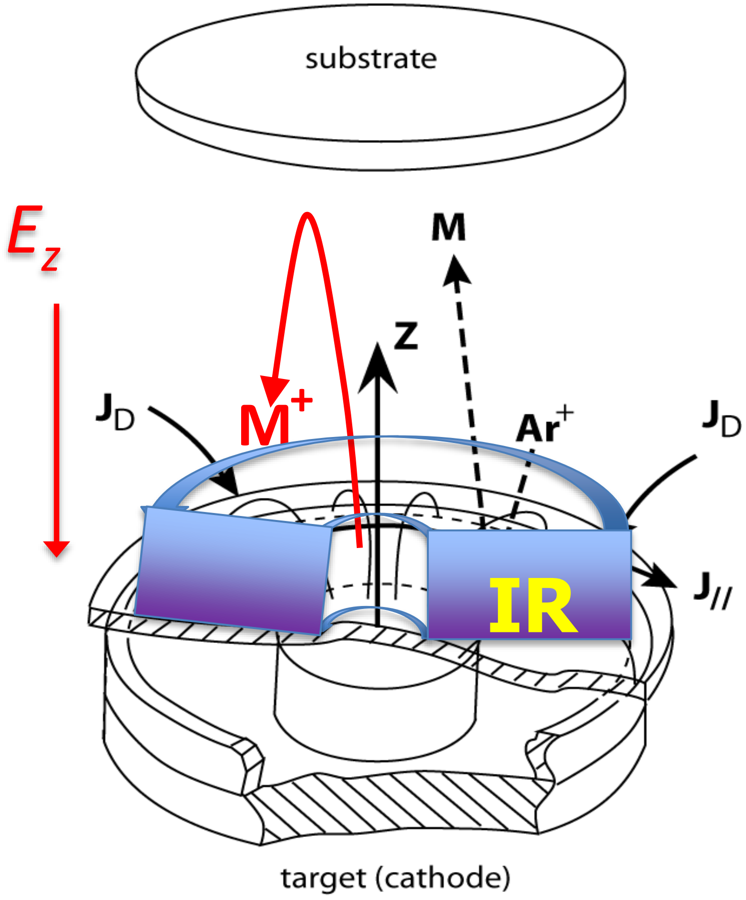
- The self-sputtering mode can really lead to the discharge current runaway!

2 - Neutral gas dynamics in HiPIMS plasmas

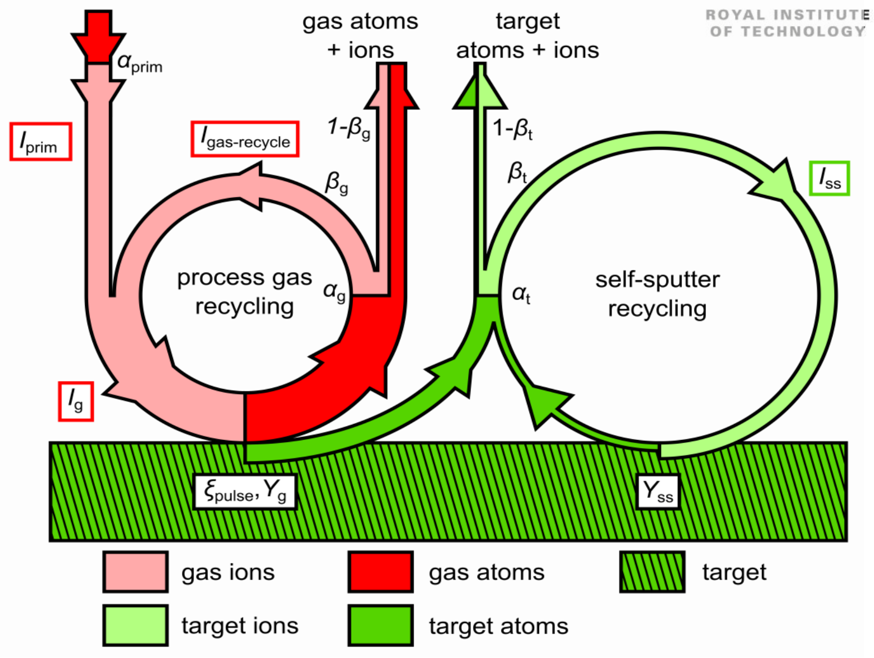
IRM (Ionization Region Model)

- ✓ 0D plasma modeling
- ✓ 2D DSMC

Background of IRM
(Ionization Region Model)
0D HiPIMS Modeling



Back-attraction + gas recycling + self-sputtering



N. Brenning *et al*., *PSST*. **21** 025005 (2012)

Strong E_z → Steep potential hill for M^+

β F_{PWR}

IRM approach

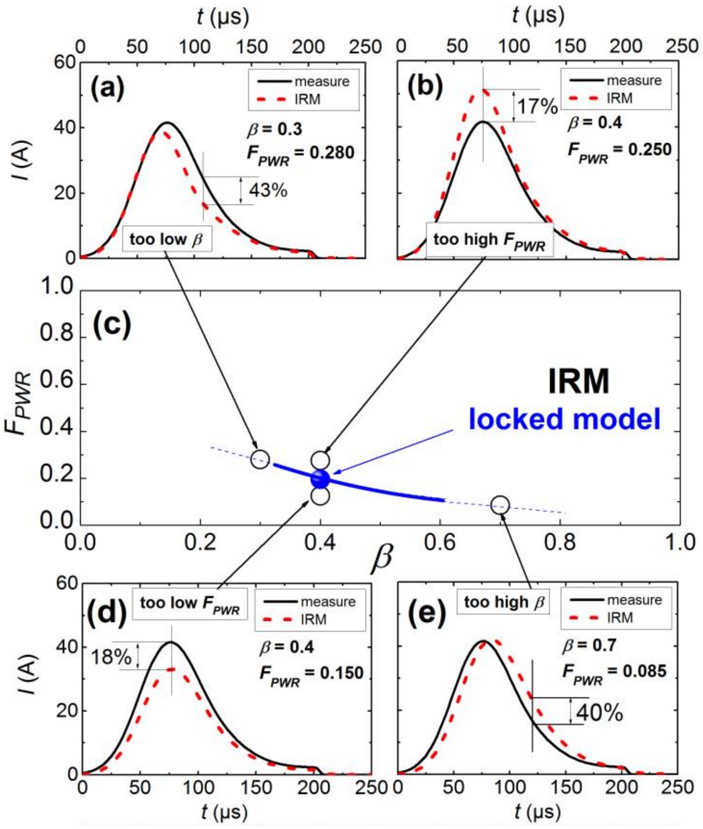
IRM assumptions

- ***e- are in Maxwell equilibrium (T_e)***
- ***Averaged plasma parameters over the IR volume***

- *β and F_{PWR} are locked by experimental **$U_D(t)$** and **$I_D(t)$**;*

IRM INPUT

- *U*(*t*), *I*(*t*);
- *Gas (Ar) T_{Ar}, p => n_{Ar}*
- *Target : Ti, γ*



M. Raadu *et al.*, *P S S T*. **20** 065007 (2011)

C. Huo *et al.*, *P S S T* **21** 045004 (2012)

Species included in R-IRM

Reactive - Ionization Region Model

Ti target in **Ar/O_2** mixture

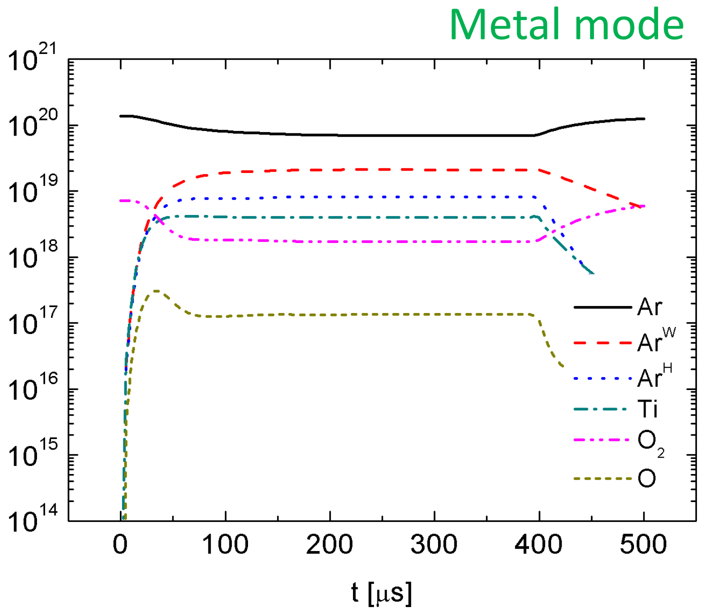
Global model: Balance equations for the main plasma species

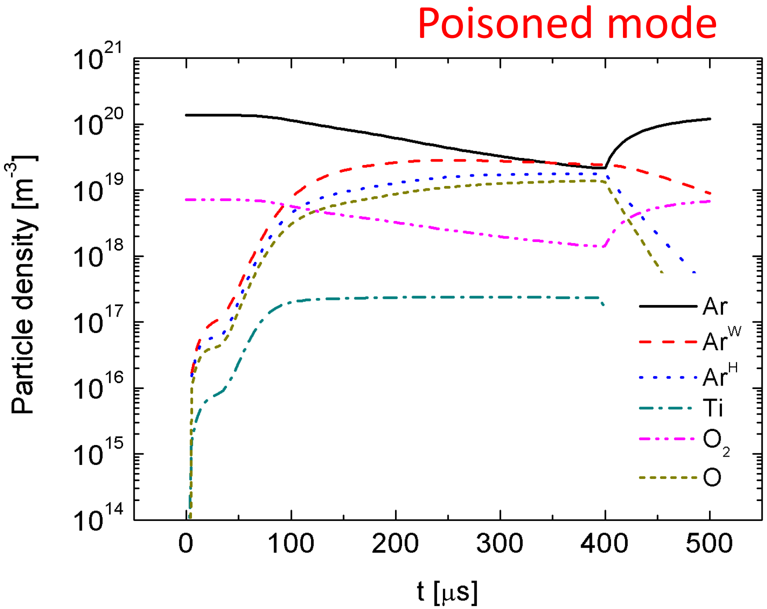
- **Electrons** (hot & cold)
- Argon atoms in the **ground state**
  - Warm argon atoms diffusing from the target at target temperature
  - Hot Ar atoms sputtered out at a few eV
- *Metastable* Ar, Ar^m ($1s_5$ and $1s_3$) (11.6 eV)
- *Argon ions* Ar^+ (15.76 eV)
- Titanium *atoms* Ti
- Titanium *ions* Ti^+ (6.83 eV), Ti^{2+} (13.58 eV)
- Oxygen molecules in the **ground state O_2**
- *Metastable* oxygen molecules $O_2(a^1delta)$ (0.98 eV) and $O_2(b^1sigma)$ (1.627 eV)
- Oxygen atoms in the **ground state $O(^3P)$**
- *Metastable* oxygen atoms $O(^1D)$ (1.96 eV)
- Positive *ions* O_2^+ (12.61 eV) and O^+ (13.62 eV)
- *Negative ions* O^-

R-IRM results - neutrals

Metal mode

- Gas rarefaction Ar ~ 50%
- Gas ***depletion*** O_2 ~ 75%
- $n_{Ti} > n_{O2}$





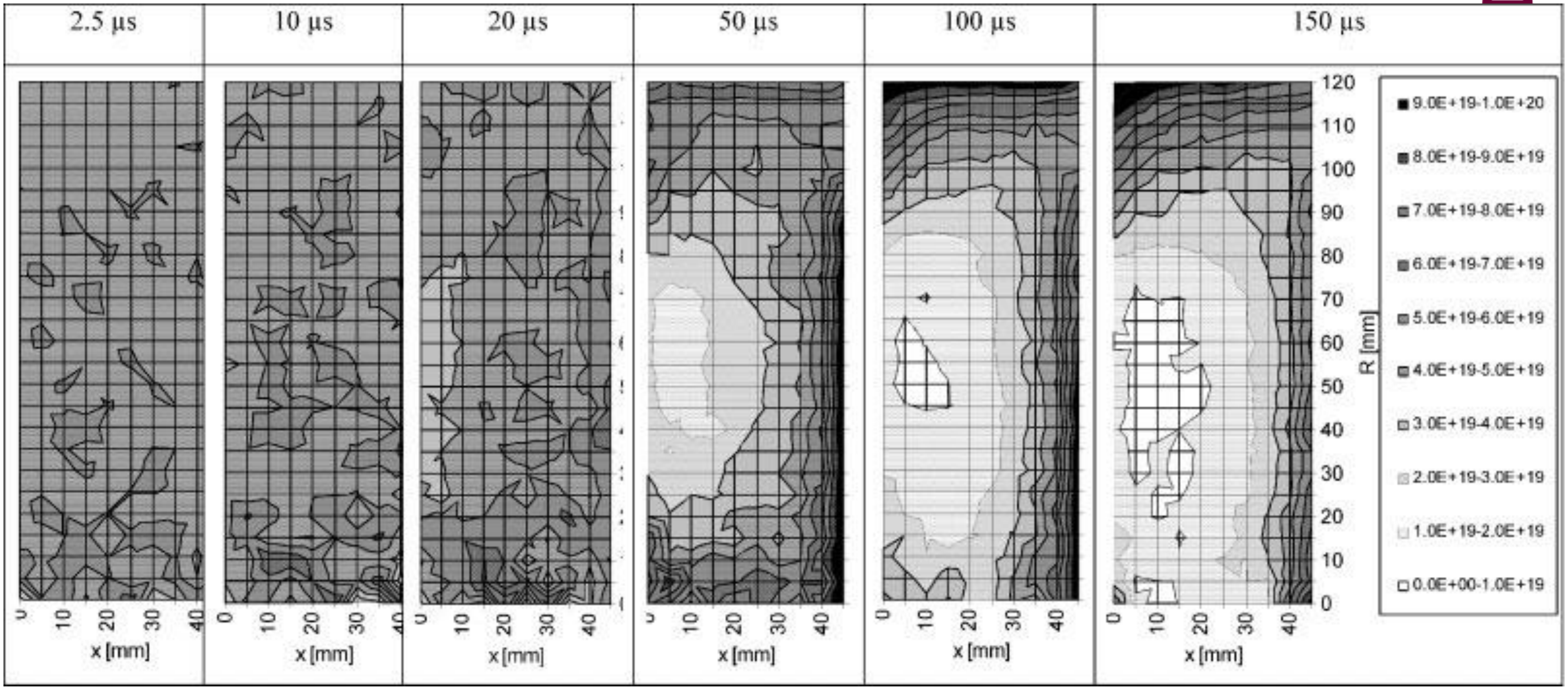
Poisoned mode

- Gas rarefaction Ar ~ 85%
- Gas ***depletion*** O_2 ~ 80%
- $n_{Ti} < n_{O2}$

J.T. Gudmundsson *et al.*, PSST. **25** 065004 (2016)

Gas rarefaction in HiPIMS by DSMC

Direct Simulation Monte Carlo

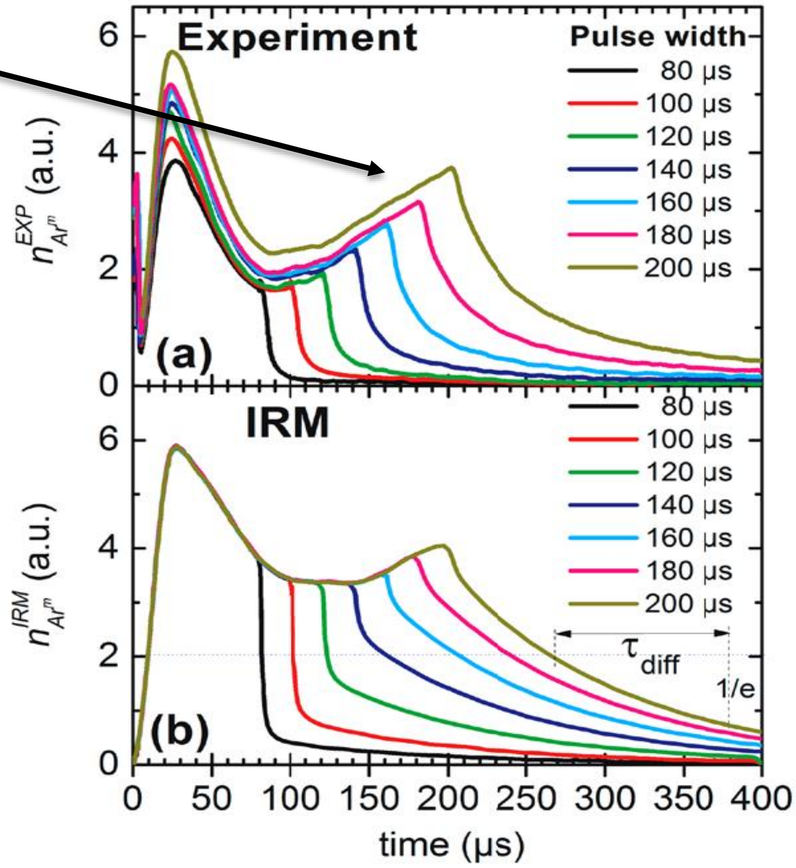


- Neutral plasma gas rarefaction occurs after 50 µs !

Kadlec. P Phys Polym 4 (2007) S419

Gas rarefaction in HiPIMS by IRM

Argon metastable



- Neutral plasma gas rarefaction occurs after 50 µs !

Exp: Vitelaru *et al.* . PSST 21 (2012) 025010

IRM: Stancu *et al.* . PSST 24 (2015) 045011

What can we learn on neutral gas dynamics in HiPIMS from 0D modeling ?

- Current ion composition !
- High current on the target means high erosion !
- Energetic particles coming from the target drag the gas out of the ionization region (wind effect) !
- Gas refilling from the undisturbed volume !

3 - Ion dynamics in HiPIMS plasmas

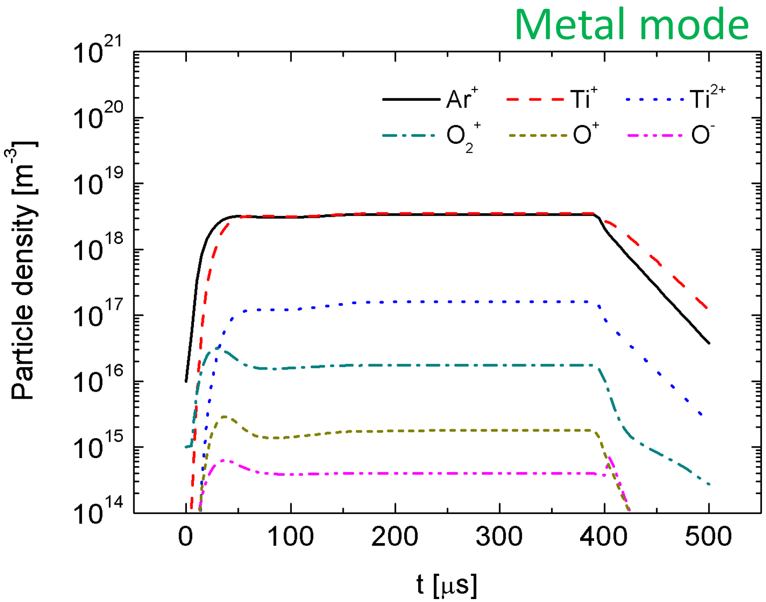
OHiPIC (Orsay High density Particle in Cell)

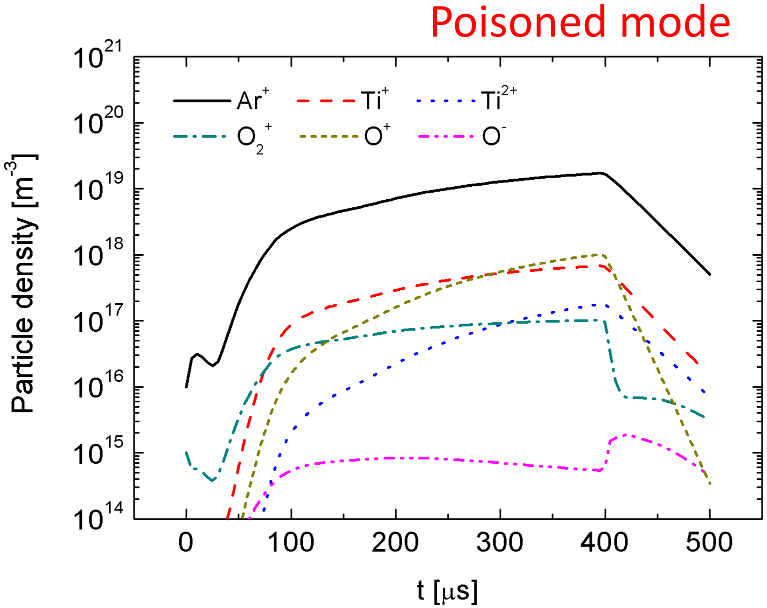
- ✓ 0D IRM modeling
- ✓ 2D plasma modeling

R-IRM results - ions

Metal mode

- Ar^+ and Ti^+ ions dominate
- Ti^{2+} ions have an order of magnitude lower density
- The O_2^+ and O^+ ion densities much lower





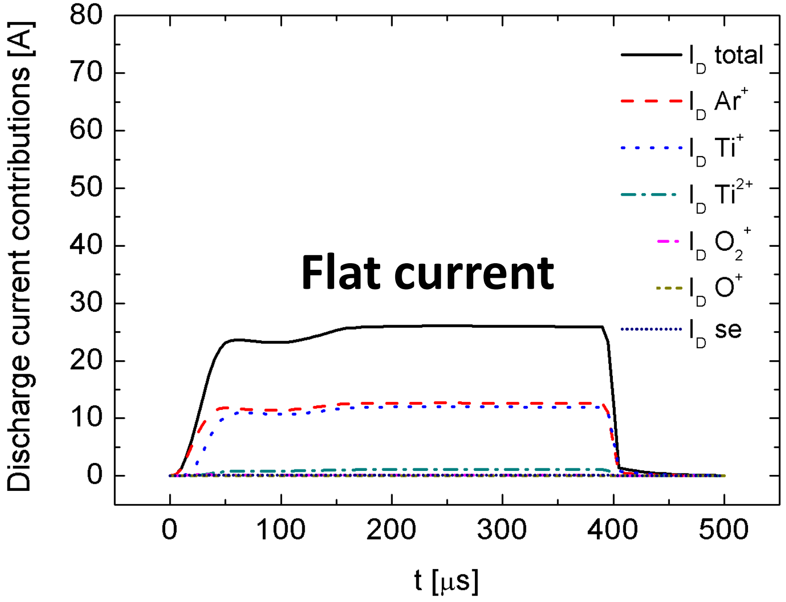
Poisoned mode

- Ar^+ ions dominate the discharge
- Ti^+ and O^+ similar densities
- The Ti^{2+} ion density low, but increases towards end of pulse

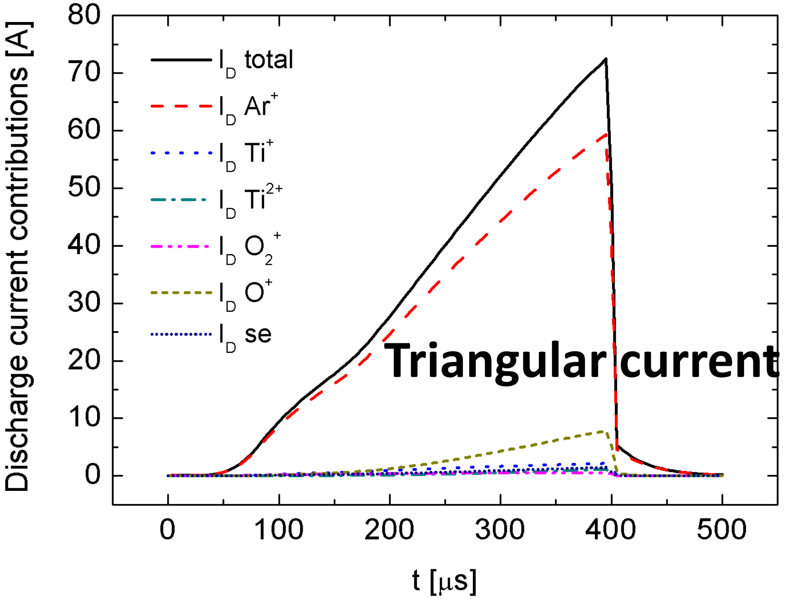
Lundin *et al.*, J. Appl. Phys. 121(17) (2017) 171917

R-IRM results
current contributions

Metal mode



Poisoned mode



Metal mode

- 50/50 of Ar$^+$ and Ti$^+$
- Gas-sustained self-sputtering
- 4% Ti^{2+}

Poisoned mode

- Mainly Ar$^+$ (80%)
- 10% O$^+$

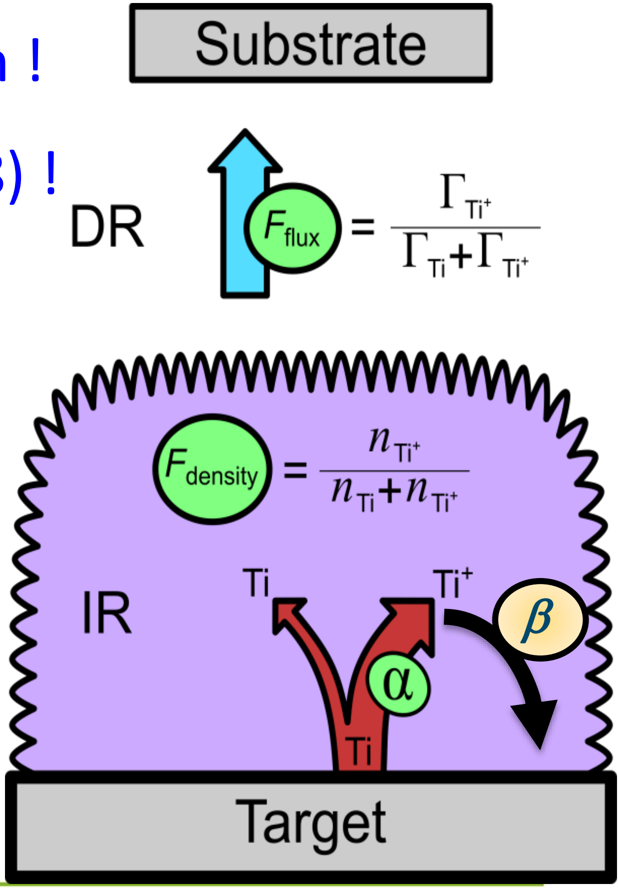
Gas-sustained SS: C. Huo et al., PSST 23 (2014) 025017

Lundin *et al.*, J. Appl. Phys. 121(17) (2017) 171917

What can we learn on ions dynamics in HiPIMS from 0D modeling ?

- Current rise means gas / metal ionization !
- Ion back-attraction fraction (β = 0.1 – 0.8) !
- Estimation of the ion flux towards the substrate !

$$F_{flux} = \frac{\int_{\text{pulse}} \Gamma_{\text{Ti}^+}(\text{t})\text{dt}}{\int_{\text{pulse}} \left(\Gamma_{\text{Ti}^+}(\text{t}) + \Gamma_{\text{Ti}}(\text{t})\right)\text{dt}}$$



2D **OHIPIC** model

Orsay HIgh density plasma Particle-In-Cell model

Debye length in HiPIMS

$n_e > 10^{13}$ cm^{-3} > **10^{19} m^{-3}** => $\lambda_e \approx$ **10 µm** (T_e = 4eV)

Geometry (*x*, *z*), periodic in *y*

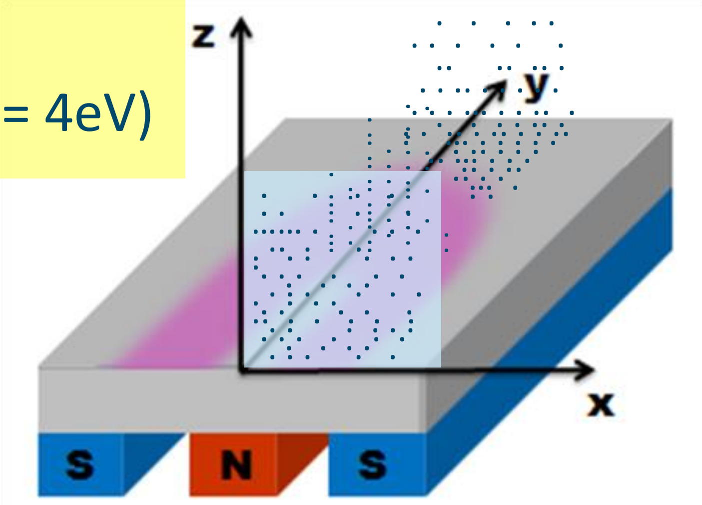
- Cell dimensions: **Δx, Δz = 10 µm !!!**
- Simulation volume: 2 x 2.5 cm^2
- Grid: **> 10^6** nodes
- 10^7 macro particles; **3D** trajectories

Control parameters

- Time step: $\Delta t = 5 \times 10^{-12}$ s ÷ 5×10^{-11} s
- Simulated real time: **15 µs** !!!

Simplified gas kinetics

- Ar^m, Ar^+ produced by e^- impact

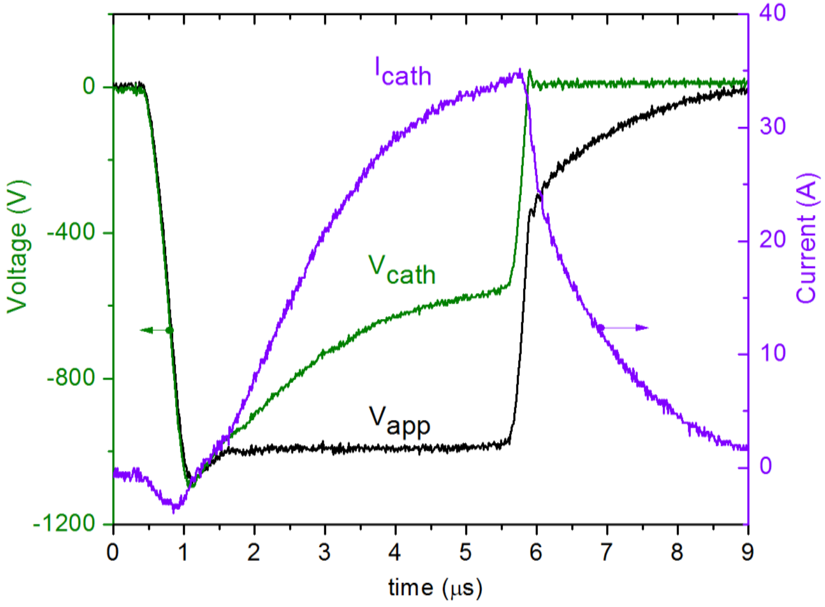


HiPIMS current
experiment *versus* simulation

OHIPIC results compared to HiPIMS experiment

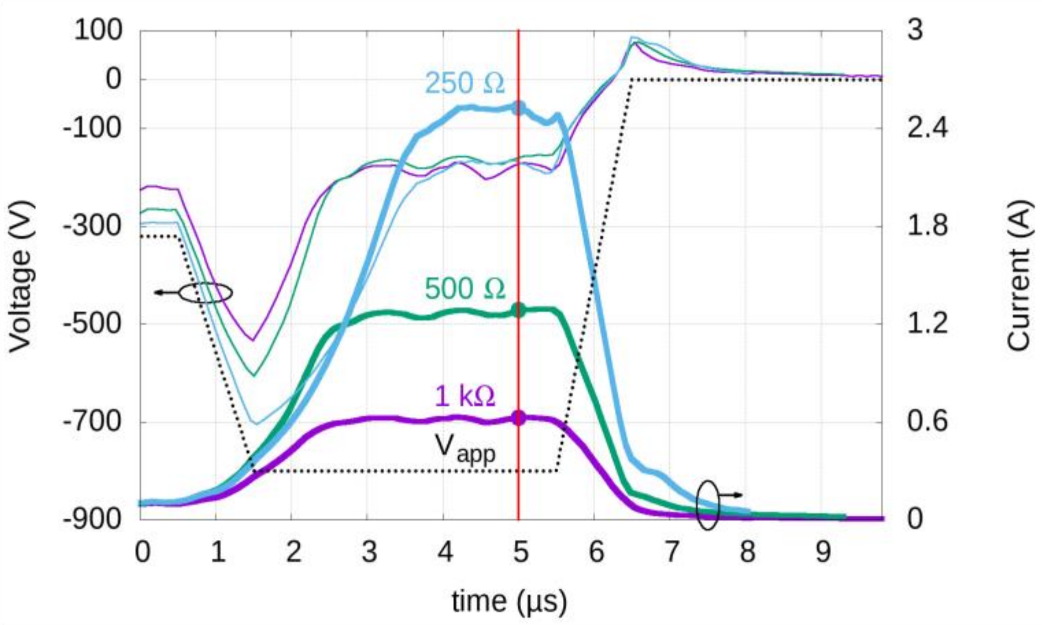
Experiment

using short pulsed HiPIMS



OHIPIC model

simulated discharge current & voltage

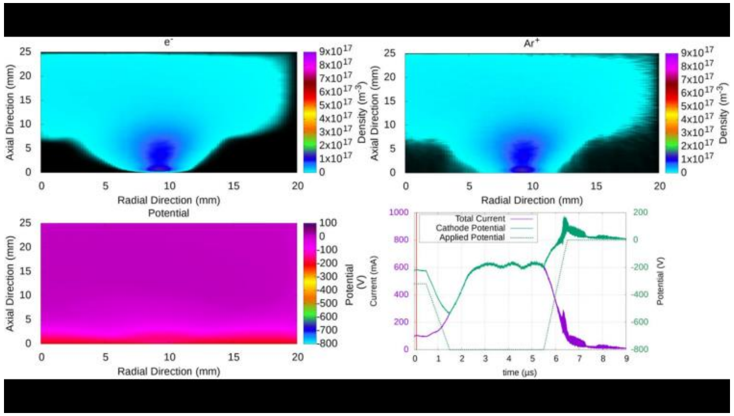


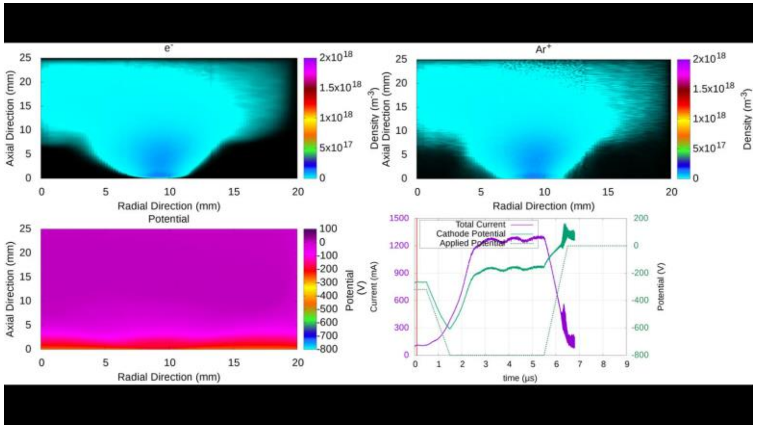
Revel *et al,* PSST (2018) *on line*

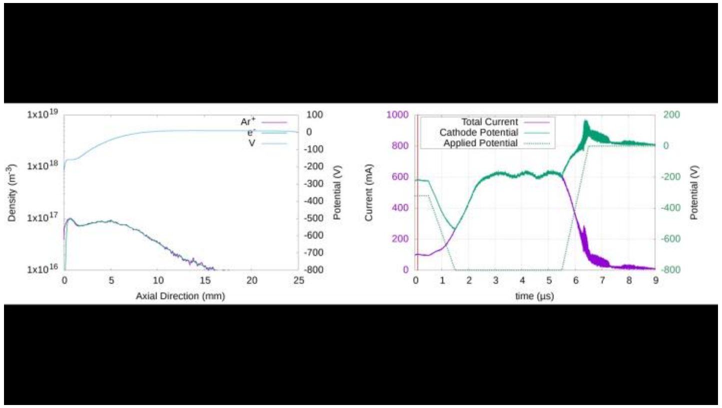
HiPIMS Plasma Evolution

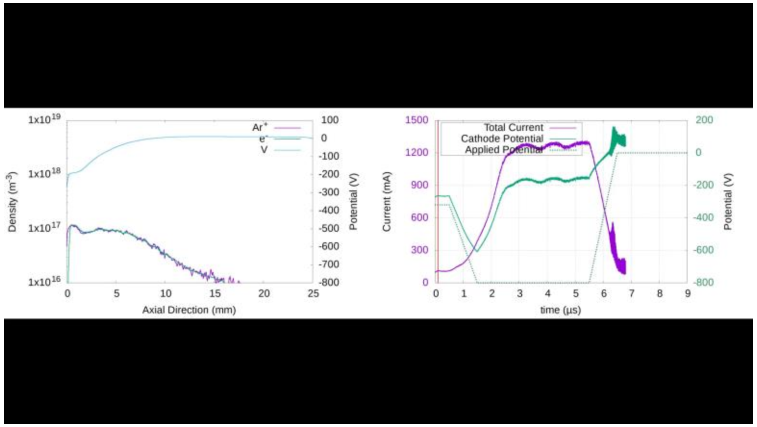
R = 1 kΩ

R = 500 Ω



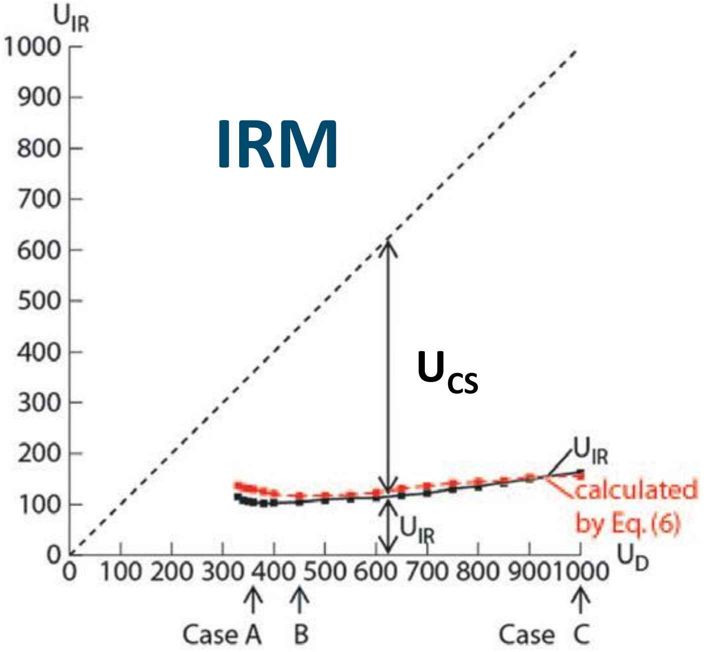




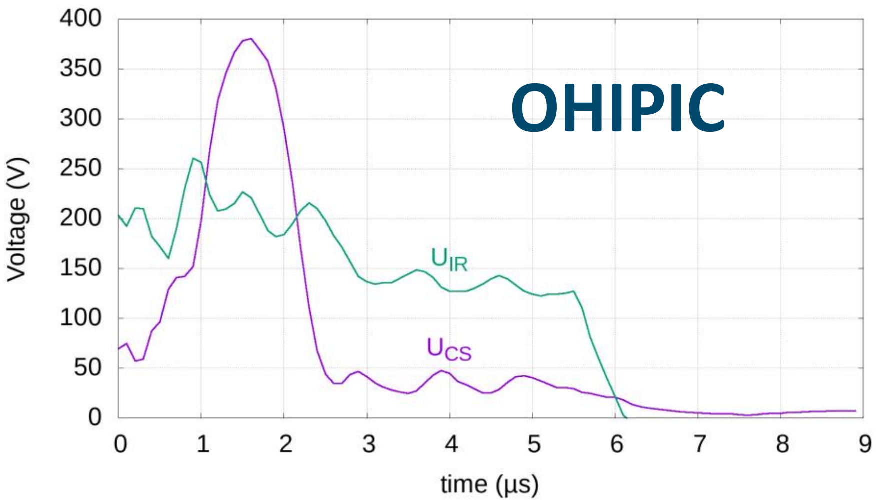


Revel *et al*, PSST (2018) *on line*

Voltage distribution over plasma regions



Huo *et al.* PSST 22 (2013) 045005

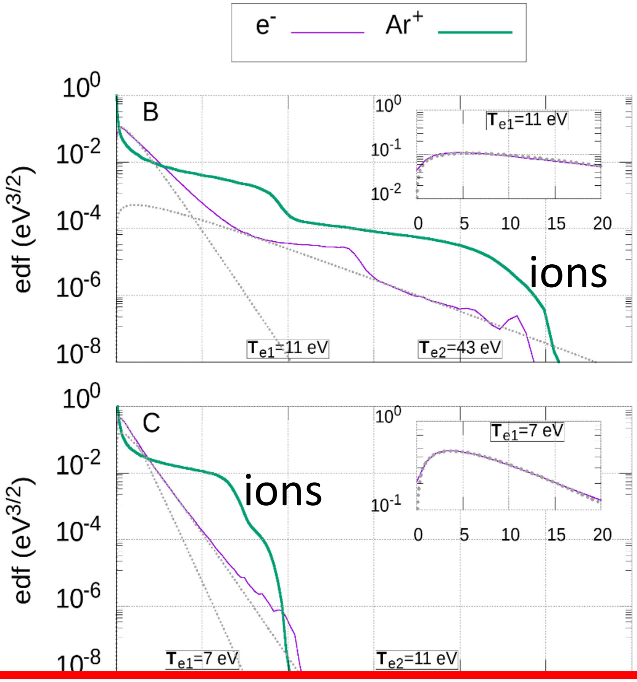


Revel, Minea, Costin PSST (2018) *on line*

- ✓ **Dischrage voltage splits between the cathode sheath U_{CS} and the Ionization Region (IR): $U_D(t) = U_{CS} + U_{IR}$**
- ✓ ***$U_{CS} > U_{IR}$ @ pulse beginning or high I_D, else $U_{CS} < U_{IR}$***

To take home!

eedf and *iedf* by OHIPIC



Current increase

Current plateau

To take home!

- ✓ ***eedf*** **shows two electron populations,**
  one thermalized and one following the cathode voltage
- ✓ **Plasma ions energy is uniformly distributed**
  over the $U_D(t)$ range

Revel, Minea, Costin PSST (2018) *on line*

What we learn from the ion dynamics by 2D OHIPIC

- The time delay in the current rise is due to the build-up of the space charge (ionization)
- Ion erosion is radially larger in HiPIMS than in DC mode
- Ion energy covers all the interval between 0 and the applied voltage
- Ion are accelerated in the voltage drop over the ionization region and further in the sheath
- Ion flux at the cathode and substrate can be evaluated

General Conclusion
on gas dynamics from HiPIMS modeling

✓ The power balance between *sheath* and *IR* as well as the *ion back-attraction* are effectively captured by global **0D IRM**

✓ **Global HiPIMS** plasma behavior is understood, either for noble gas or reactive mixtures

✓ 2D particle modeling give access to charged species evolution (densities, fluxes), but also ***eedf*** and ***iedf***

✓Good coherence between **IRM** and **OHIPIC** allows to apprehend for the first time the spokes by modeling

✓ Gas rarefaction can be captured by IRM and DSMC, but it plays for pulses longer than 50 µs or for very high currents

PLATHINIUM

PLASMA THIN FILM INTERNATIONAL UNION MEETING

23-27 September 2019

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Magnetron Ion processing & Arc Technologies European Conference

MAGNETRON

CIP2017

Colloquium International on Plasma processes

PLASMA

ITFPC17

Innovations in Thin Film Processing and Characterization

THIN FILM

PLATHINIUM
PLASMA THIN FILM INTERNATIONAL UNION MEETING

Thank you!


kia ora
ありがとう
barka
tau
bedankt
arigatô
bayarlalaa
děkuji
rahmat
grazzi
мерси
tack
sulpáy
akun
ačiū
dakujem
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djiere dieuf
tanemirt
mersi
diolch
dhanyavad
shukriya
spas
teşekkü ederim
gràcies
welalin
enkosi
mochchakkeram
sukriya
merci
chnorakaloutioun
dank je
terima kasih
gratias ago
তোমাকে ধন্যবাদ
mamnun
misaotra
kop khun
najis tuke
teşekkür ederim
obrigado
matondo
paldies
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taiku
хвала
blagodaram
didi madloba
mahalo
gràçie
tapadh
chokrane
grazie
asante
thank you
mèsi
ευχαριστώ
mauruuru
takk
謝謝
Баярлалаа
xièxie
dankie
sobodi
dankon
manana
vinaka
danke
go raibh maith agat
tenki
kiitos
спасибо
murakoze

ACCESSIBILITY

- ✓ Direct flights from Paris, Bruxelles, London, Barcelona…
- ✓ 17 km away from Nice International Airport
- ✓ 2 stations : 1 for high speed trains (TGV) and 1 for Regional trains
- ✓ Highway A8 "la Provençale « Exit 44 »

Recycled ions in poisoned mode

Reactive HiPIMS (Ti @ Ar/O_2)

Gas recycling

$$\Pi_{gas} = \alpha_{GA,recycle}\beta\xi_{pulse} = 0.75 \times 0.90 \times 1 \approx 0.68$$

$$\Pi_{gas-recycle} \approx 3.08$$

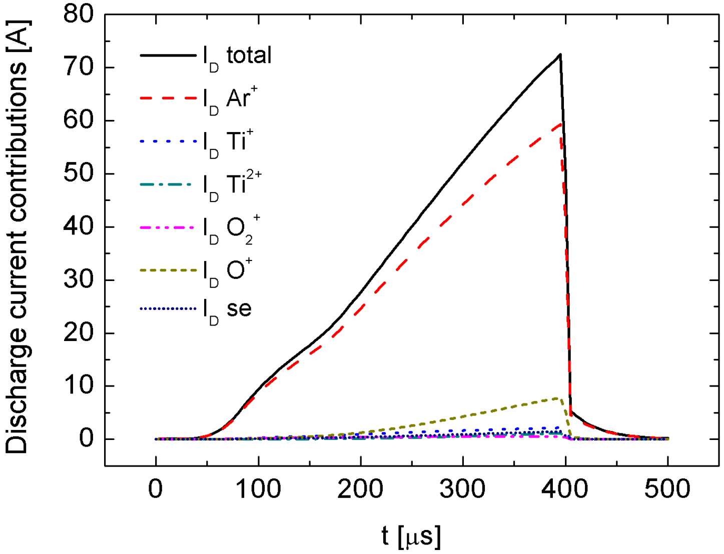
Self-sputter recycling

$$\Pi_{SS} = \alpha_{O}\beta Y_{SS} = 0.35 \times 0.90 \times 0.25 \approx 0.08$$

$$\Pi_{SS-recycle} \approx 1.09$$

$$\Pi_{SS} = \alpha_{Ti}\beta Y_{SS} = 0.97 \times 0.90 \times 0.04 \approx 0.03$$

$$\Pi_{SS-recycle} \approx 1.04$$



Discharge dominated by process gas recycling.

Our Ar/O_2/Ti process has in principal no upper limit→ Current increases

High Power Impulse Magnetron Sputtering (**HiPIMS**)

SHORT & FAST Pulsed generator concept which uses

- **preionization** to guarantee the fast rise time of the current,
- fast fall time of the discharge voltage at the switch-off

- ✓ **Average Power 100 W**
- ✓ **Pulse width: ~10 µs**
- ✓ **Pulse Power ~ 1 MW!**
- ✓ **U_{max} ~ 1 kV**
- ✓ **I_{max} : 10-300 A**

Ganciu *et al*, World Patent No. WO 2005/090632.

Ganciu *et al*, US Patent No. 7, 927, 466 B2 (19 April 2011)

Kouznetsov , U. S. Patent No. 6,296, 742 B 1 (**2001**)