On the cause of a double hysteresis during reactive magnetron sputtering

Koen Strijckmans,
Roeland Schelfhout, Diederik Depla

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GHENT UNIVERSITY

FACULTY OF SCIENCES

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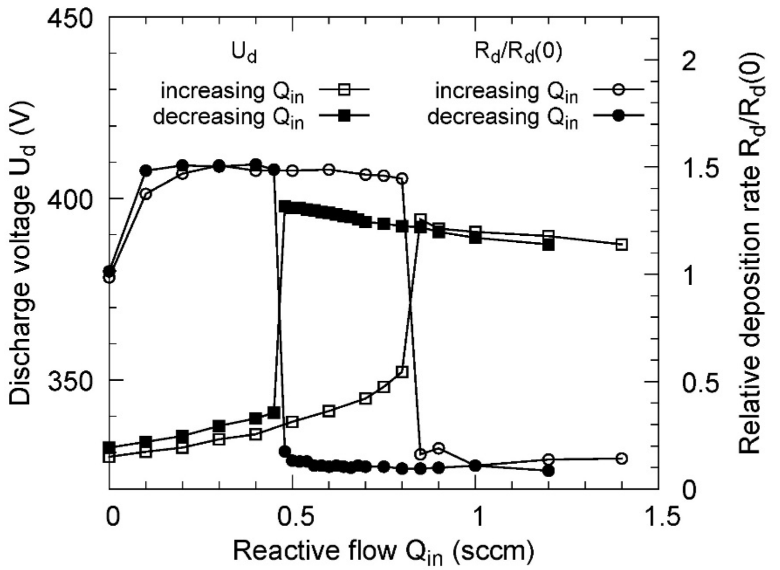
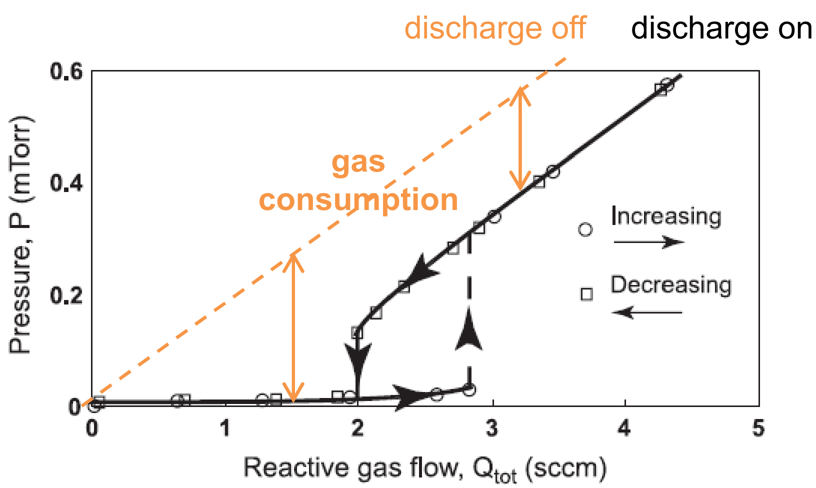
Outline

Hysteresis phenomena





<u>Direct controlled hysteresis experiment</u> = stepwise in/decrease of single **operation parameter**

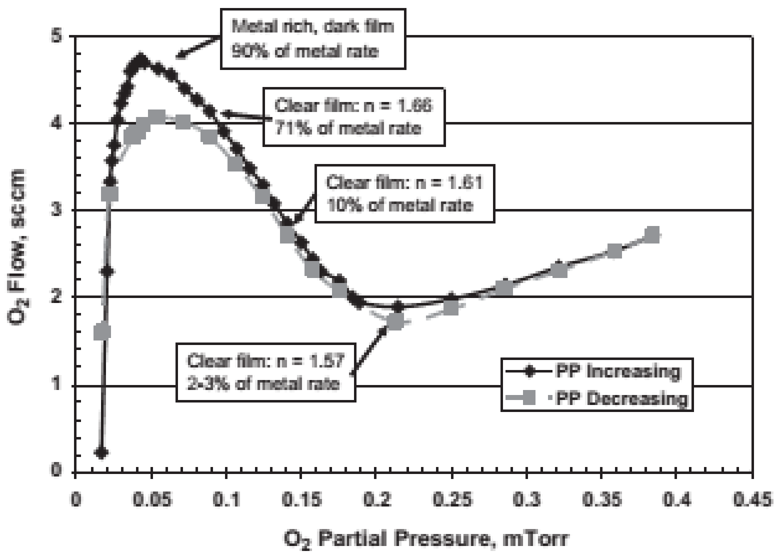
<u>Hysteresis</u> in

a) reactive gas pressure
b) discharge voltage
c) deposition rate

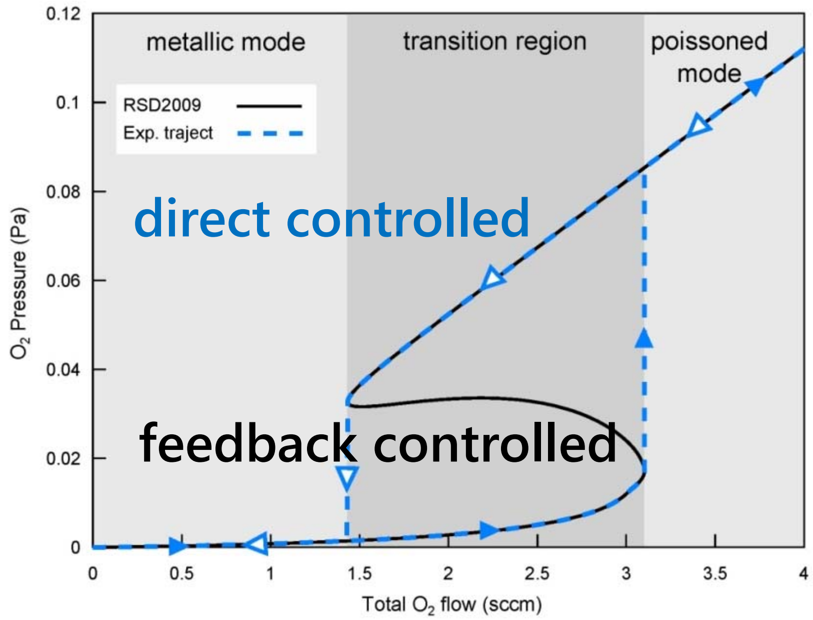
by **poisoning** (current = constant)

a) vanishing getter pump
b) changing Y_{SEE}
c) decreasing sputter yield ($Y_c << Y_m$)

Hysteresis phenomena



Sproul, Thin Solid Films, 491 (2005) 1-17



<u>Feedback controlled hysteresis experiment</u> =

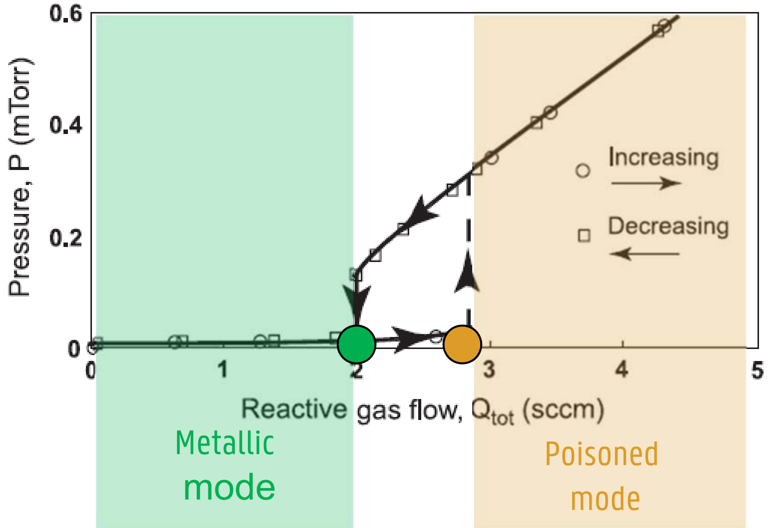
stepwise in/decrease of **variable** (e.g. p_r) by feedback controlled operation parameter

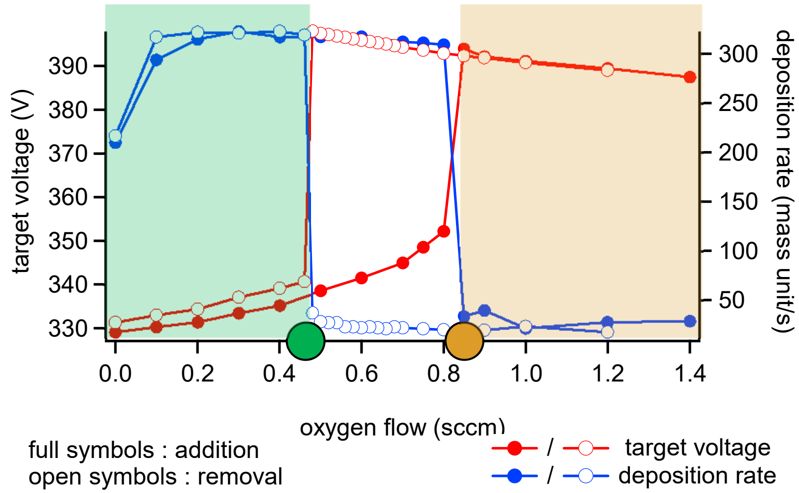
😐 S-shape

☹ instability transition region

☺ better film control / deposition rate

Hysteresis phenomena





First critical point

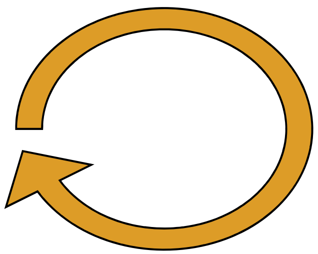
Second critical point

higher reactive gas pressure

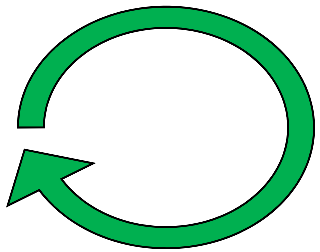
more target reaction

lower reactive gas pressure

less target reaction



avalanche effect



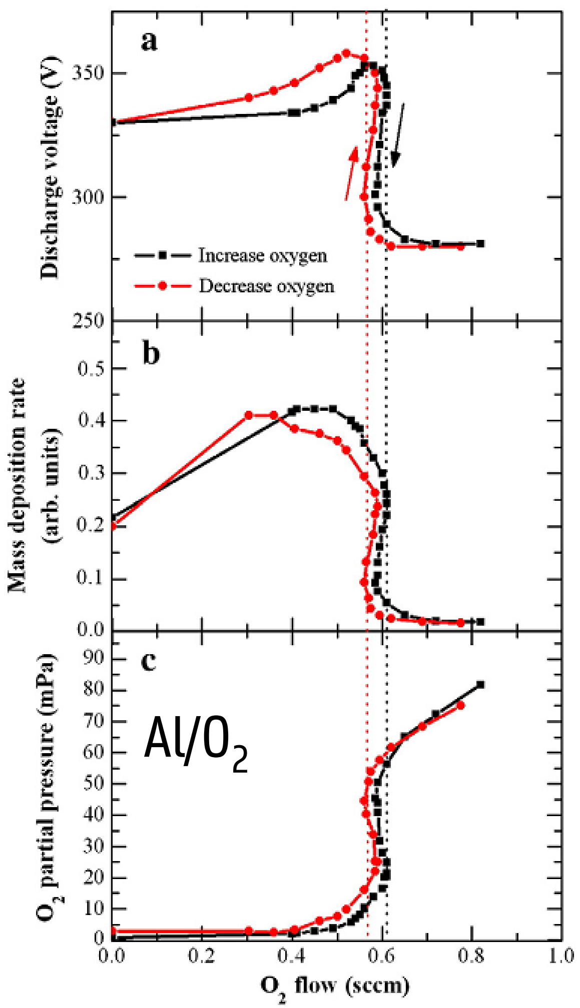
less reactive gas gettering by substrate

lower metal deposition rate

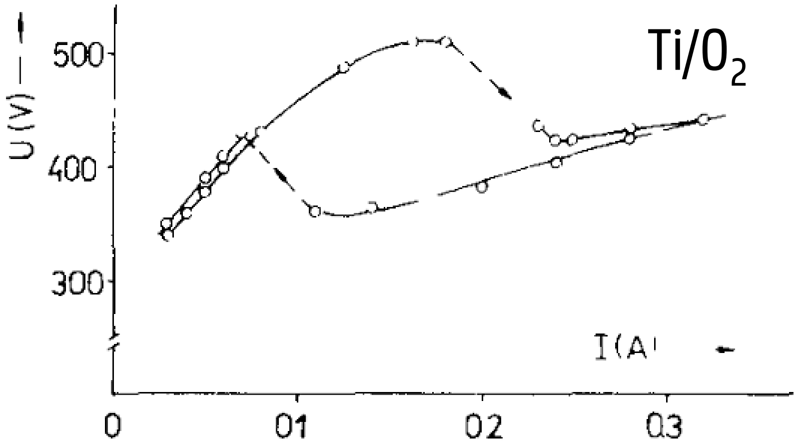
more reactive gas gettering by substrate

higher metal deposition rate

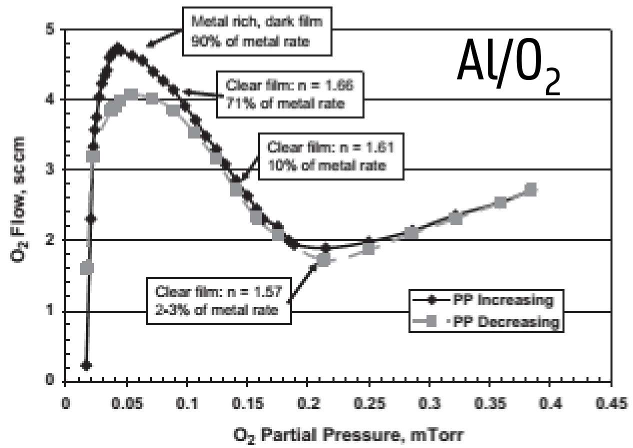
Double hysteresis?



Aiempanakit, Thin Solid Films, 519 (2011) 7779-7784

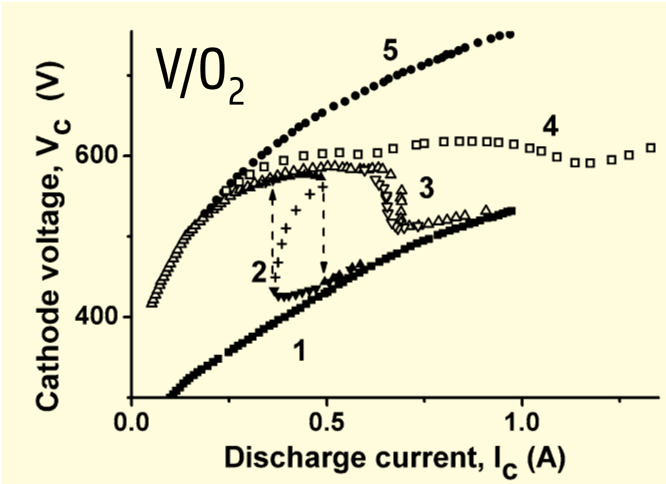


Steenbeck, *Thin Solid Films*, 92 (1982) 371-380

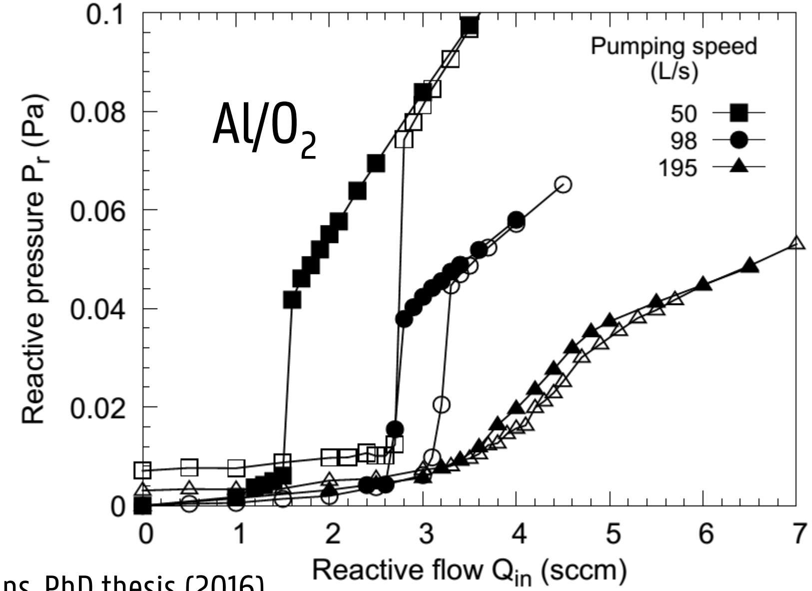


Sproul, Thin Solid Films, 491 (2005) 1-17

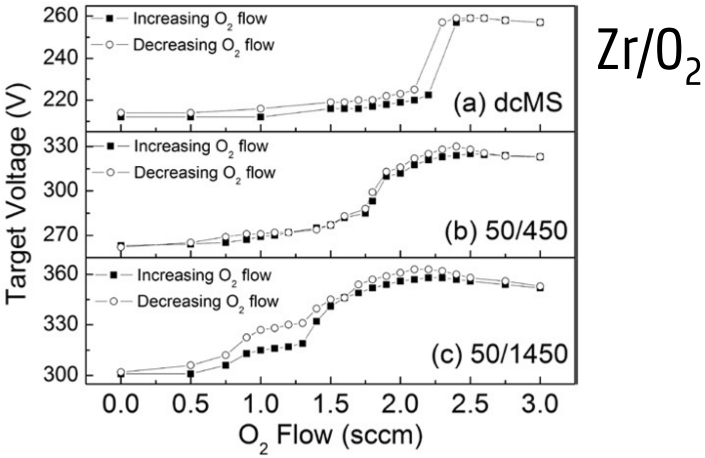
Double hysteresis?



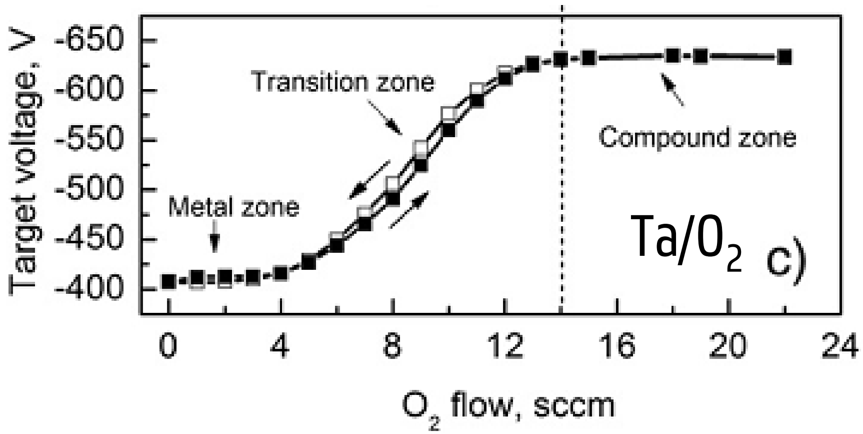
Marchenko, *Proc. SPIE* 7025, 70250D (2008)



Strijckmans, PhD thesis (2016)



Sarakinos, Surf. Coat. Technol.., 202 (2008) 5033-5035



Juškevičius, Thin Solid Films, 589 (2015) 95

Double S-curve: an 'simple' artifact?

… as these hystereses are measured sequential.
Several irreversible time-dependent or systematic effects can influence the hysteresis:

- change in discharge voltage due to target erosion
- chamber heating
- changing/ disappearing anode due to sputter deposition
- changing plasma potential
- chamber setup / magnetron type

→ challenge: Find an alternative measurement procedure, excluding most (all?) artifacts!

Outline

Experimental

Goal: eliminate ‘trivial’ causes of the double hysteresis

Means: original measure procedure of the double S-shaped hysteresis by scanning the 4-dimensional (I, V, p, Q) parameter space
☞ current I, voltage V, flow Q and pressure p

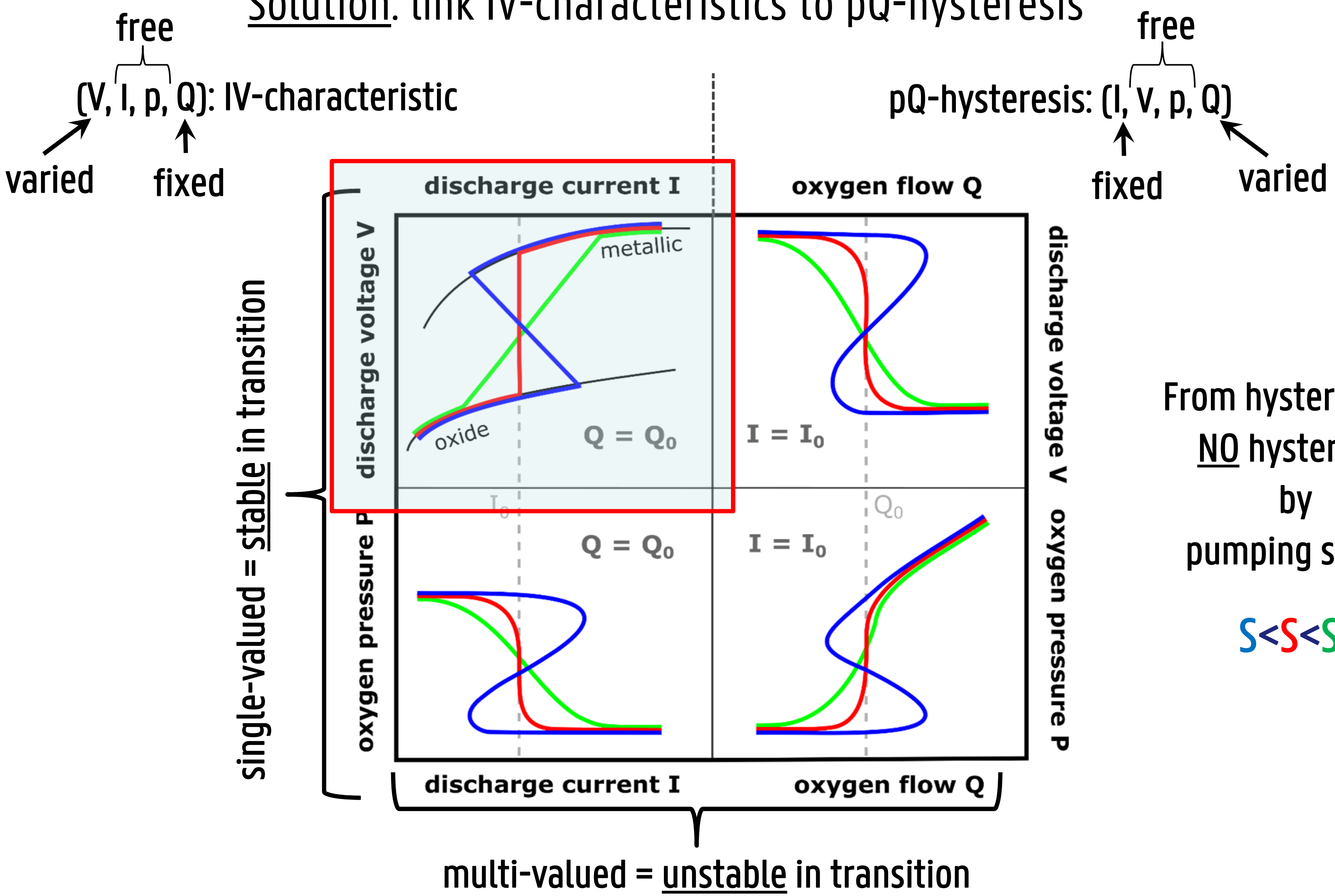
Benefits: EXCLUDE unwanted causes like

- target erosion
- chamber heating
- ...

Setup: Al 2 inch planar target in (0.2 x 0.2 x 0.4 m^3) chamber
Ar (0.4 Pa) / O_2 (varied) atmosphere
30 L/s pumping speed

(I, V, p, Q) parameter space

Solution: link IV-characteristics to pQ-hysteresis



From hysteresis to NO hysteresis by pumping speed

S<S<S

IV-characteristic

Intrinsic stability of the IV-characteristic

Increase in V

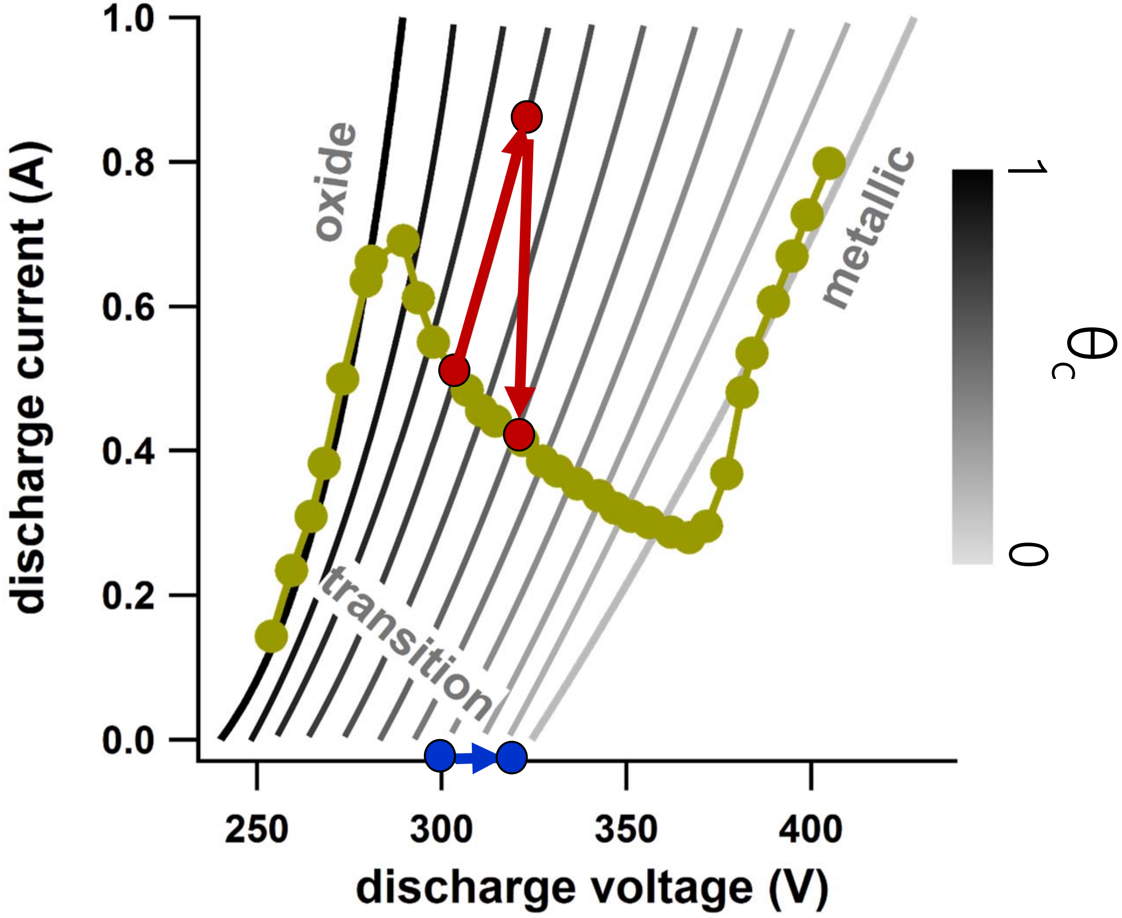
I increases along IV-line

target gets less poisoned

decrease in current I

target gets more poisoned

stops decrease in current I
stabilize to new IV point

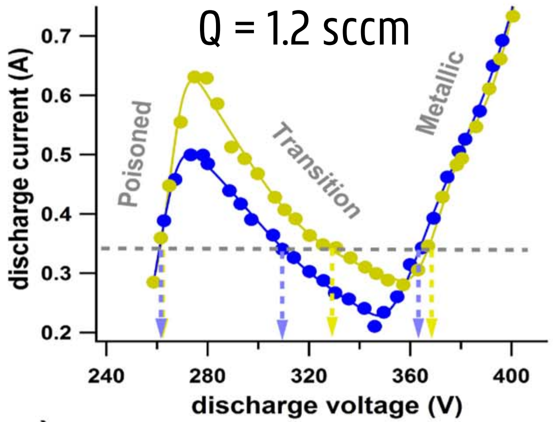


(I, V, p, Q) parameter space

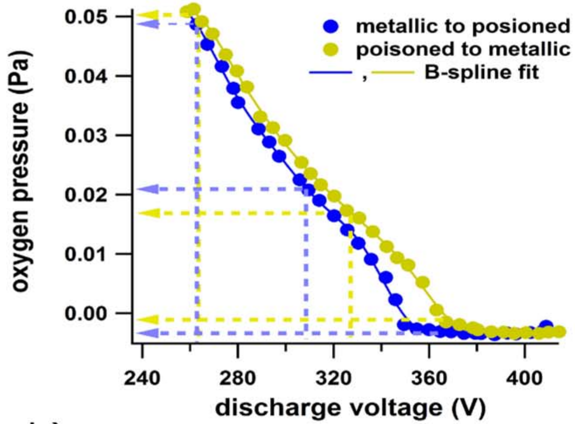
Solution: link IV-characteristics to pQ-hysteresis

Voltage controlled operation
at fixed oxygen flow

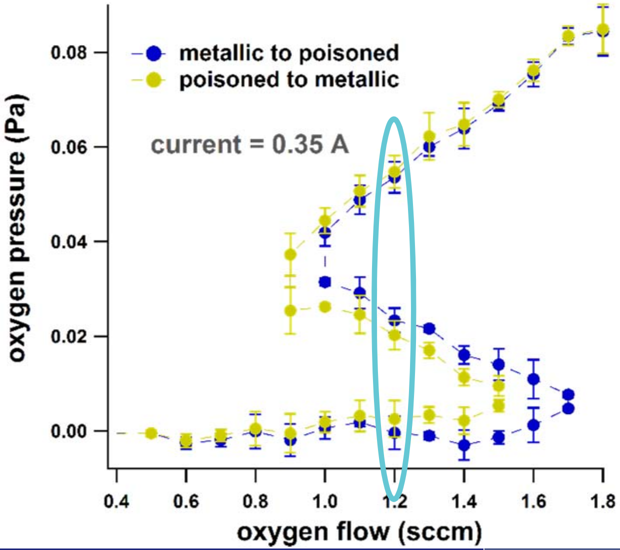
☞ stable access to transition mode
☞ fast stabilization times
☞ low voltage (poisoned mode)
↔ high voltage (metallic mode)



a)

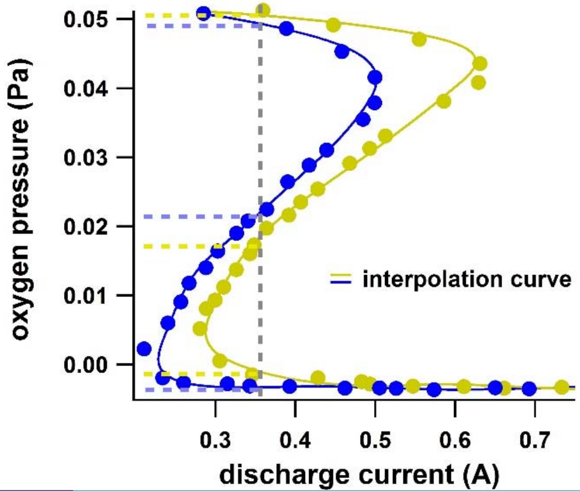


b)



Result:
pQ-point

for fixed
I = 0.35 A

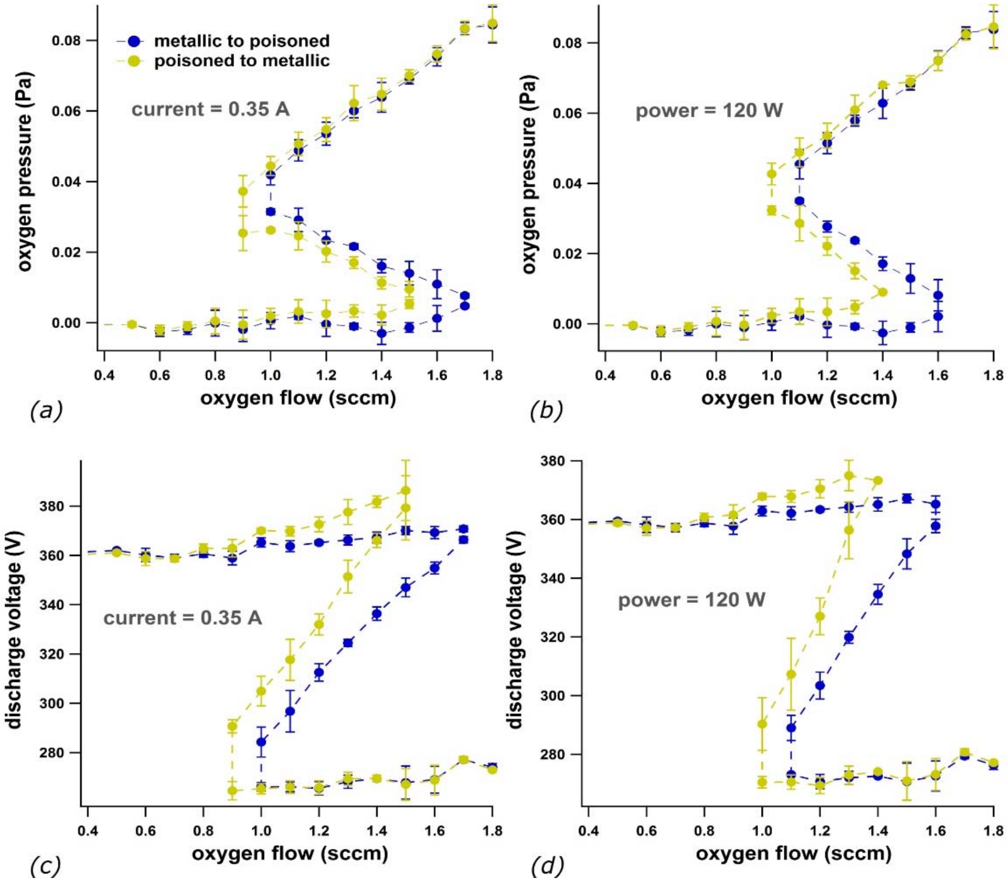


A double S-curve, is it real?

Reconstruction of pQ and VQ-hysteresis

Every IV-characteristic with fixed Q is measured randomly

→ sequential time-dependent effects are excluded



Reveals a significant different process curve depending on process history

Impact on voltage controlled feedback mechanism?

Schelfhout et al.,
Appl. Phys. Lett 109, 111605 (2016)

Double S-curve: NOT a 'simple' artifact

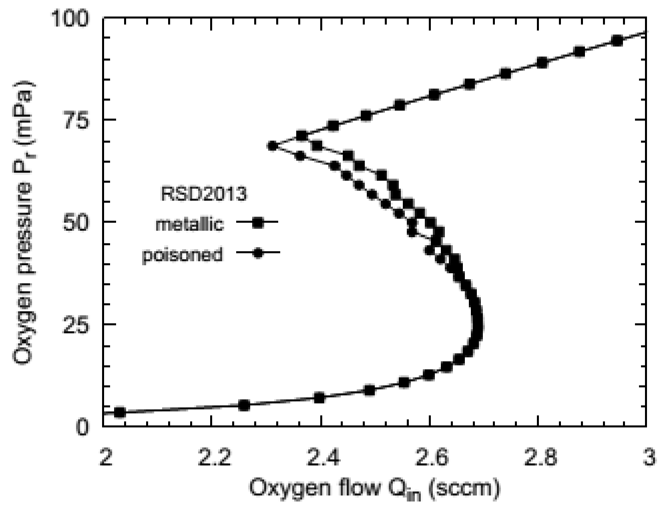
Which irreversible time-dependent/systematic effects did we exclude?

- change in discharge voltage due to target erosion
  - 👌 ± 5 s stabilization time
  - 👌 target in reference condition
- chamber heating
  - 👌 fast/random measurements
- changing/ disappearing anode due to sputter deposition
  - 👌 stainless steel brush
- changing plasma potential
  - 👌 Langmuir probe measurements
- chamber setup / magnetron type
  - 👌 rotatable magnetron

Outline

Can we understand this?

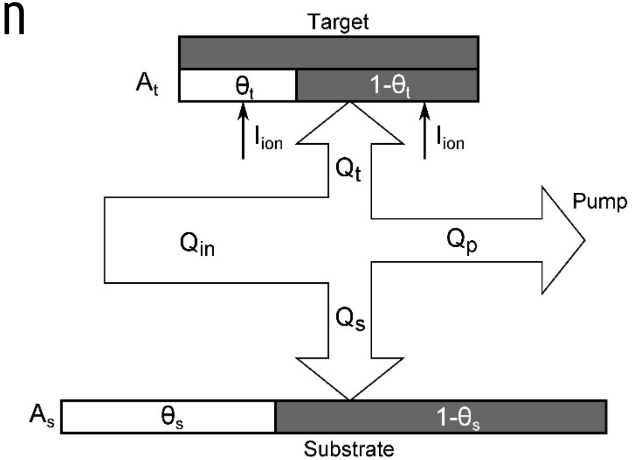
YES, even our Reactive Sputter Deposition (RSD) model was predicting this.



Strijckmans, PhD thesis (2016)

… but what is the RSD model all about?

- (semi-)analytical model focusing on the description of the process curve
- based on balancing of <u>sputtered</u> and <u>reactive</u> material
- a Berg-like model with a quite <u>advanced target</u> description



RSD model in one page

System part	Resolved variable		Model approach
Chamber	P Q_p	reactive partial pressure gas flow to pump	one-cell
Target •Surface • Subsurface	Q_t θ_m θ_c θ_r $n_m(x)$ $n_r(x)$	gas flow consumption metallic fraction chemisorbed fraction reacted fraction metal concentration reactive gas concentration	one-cell uniform current multi-cell non-uniform current depth profile SRIM implantation
Substrate	θ_s Q_s	chemisorbed fraction gas flow consumption	one-cell multi-cell SIMTRA profile

5 BALANCE equations ⇔ 5 ODE's

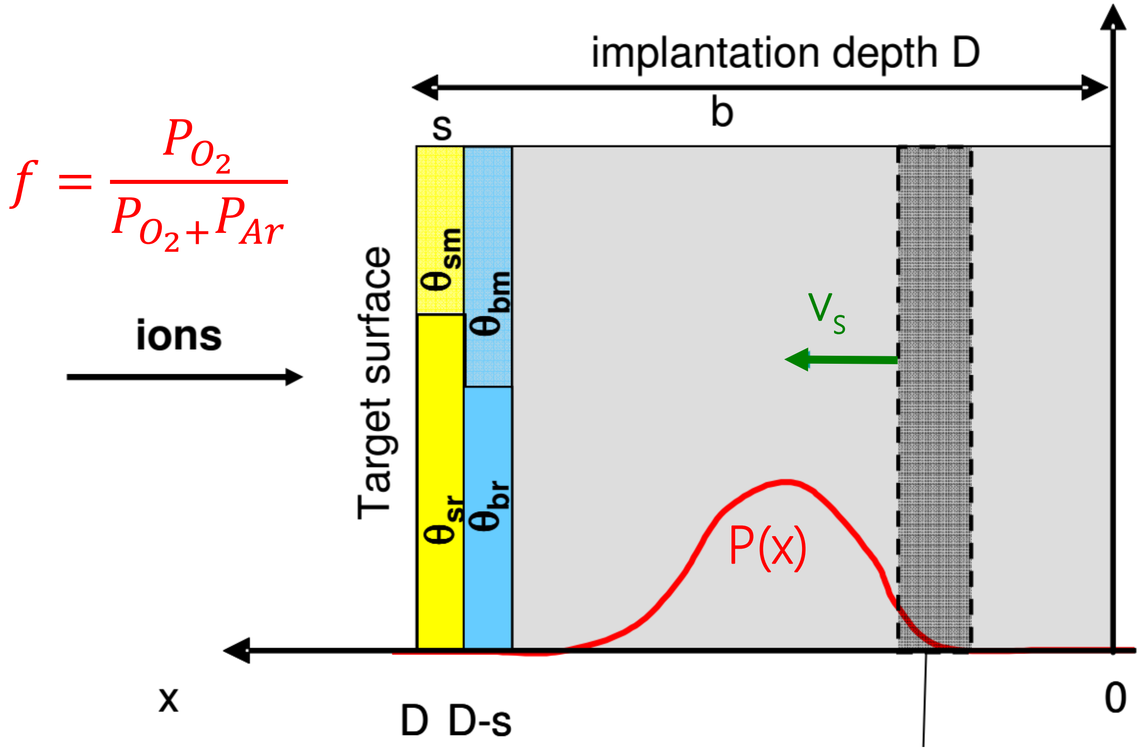
$$0 = f(y) \leftrightarrow \frac{dy}{dt} = f(y)$$

steady state ⇔ time

2 ODE's ⇔ 2 PDE's

$$0 = f(y, \frac{\partial y}{\partial x}) \leftrightarrow \frac{\partial y}{\partial t} = f(y, \frac{\partial y}{\partial x})$$

Zooming in on the RSD model



Target bulk:

- Reactive ion implantation (f)
- Transport to the surface by erosion (v_s)
- Chemical reaction with compound formation (k)

$$v_s \frac{dn_R(x)}{dx} = -k n_R(x) n_m(x) + 2 f j_{ion} p(x)$$

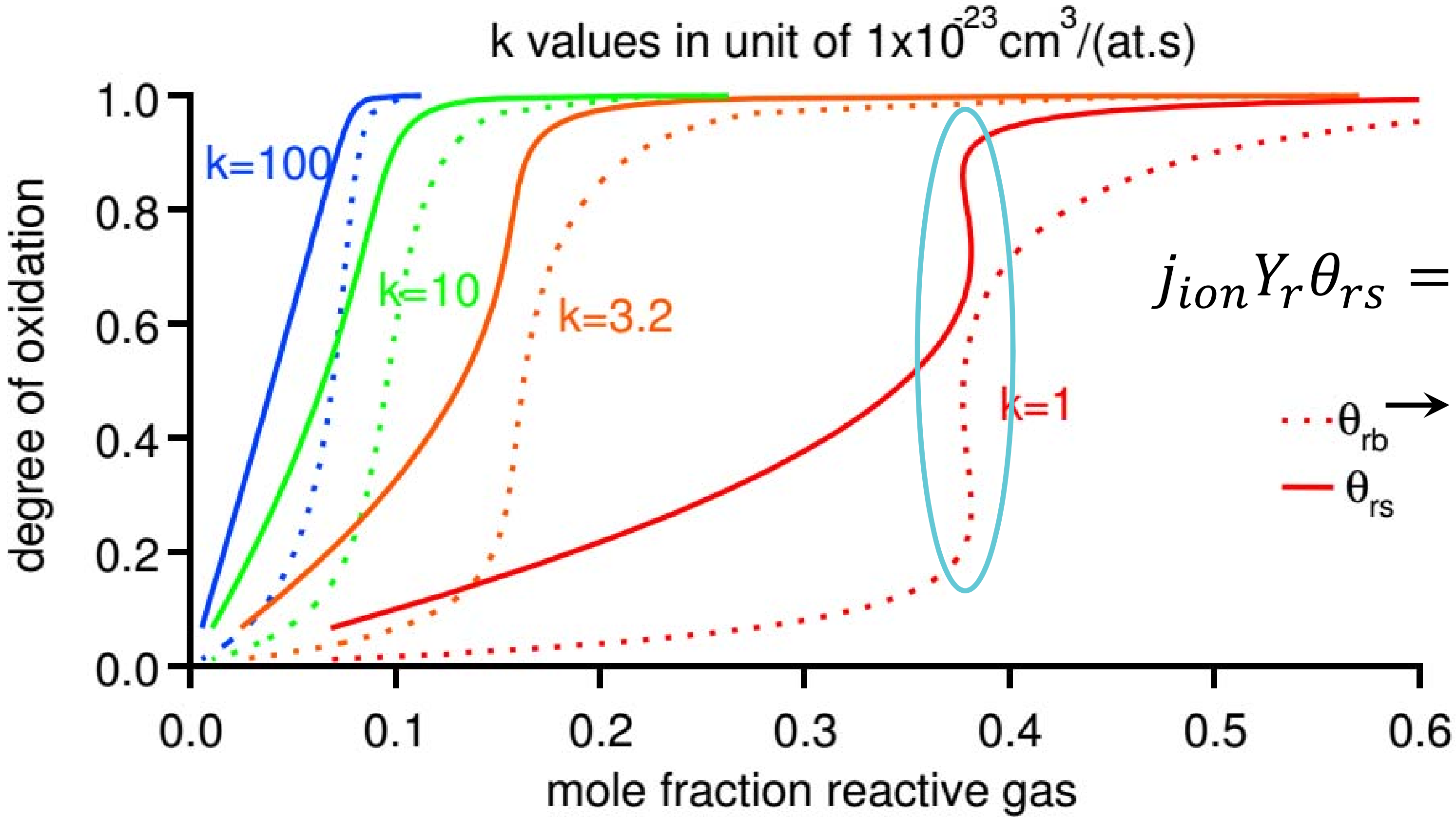
$$v_s \frac{dn_m(x)}{dx} = -k n_R(x) n_m(x)$$

2 coupled ODE's

Depla et al., J. Phys. D: Appl. Phys. 40 (2007) 1957–1965

The steady state RSD model

Playing around with the model!

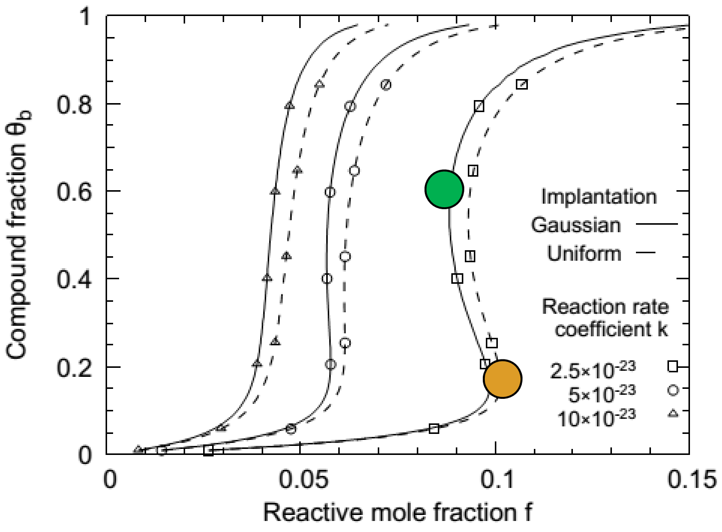


$$j_{ion} Y_r \theta_{rs} = v_s \theta_{rb} n_0$$

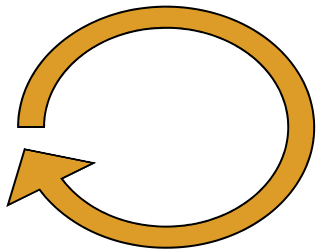
$$\theta_{rb} \rightarrow 1 - \frac{n_m}{n_{m,0}}$$

There's an abrupt change in the target state as a function of reactive gas fraction

Pressure hysteresis phenomena



First critical point



Avalanche effect develops by two mechanisms:

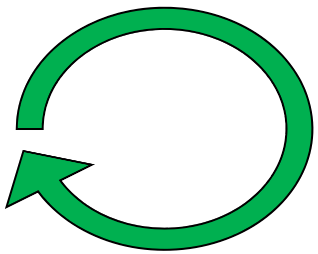
1. Implantation term

$$\frac{dn_R(x)}{dx} = \frac{2fj_{ion}}{v_s} p(x)$$

2. Reaction term

$$\frac{dn_R(x)}{dx} = -\frac{k}{v_s} n_M(x) n_R(x)$$

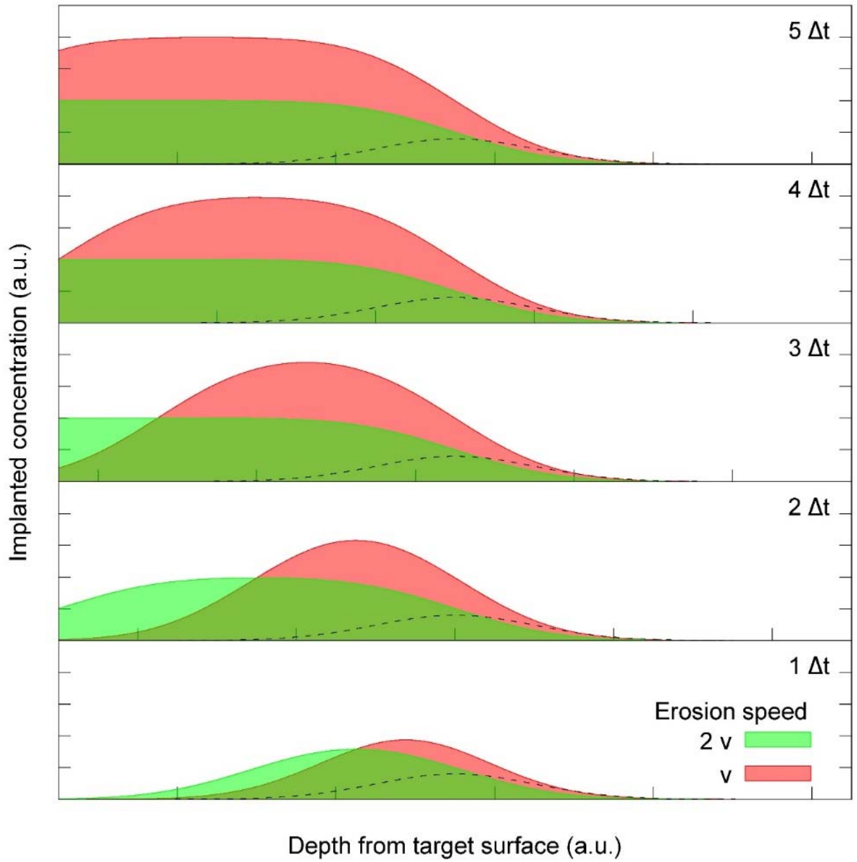
Second critical point



The two critical mechanisms

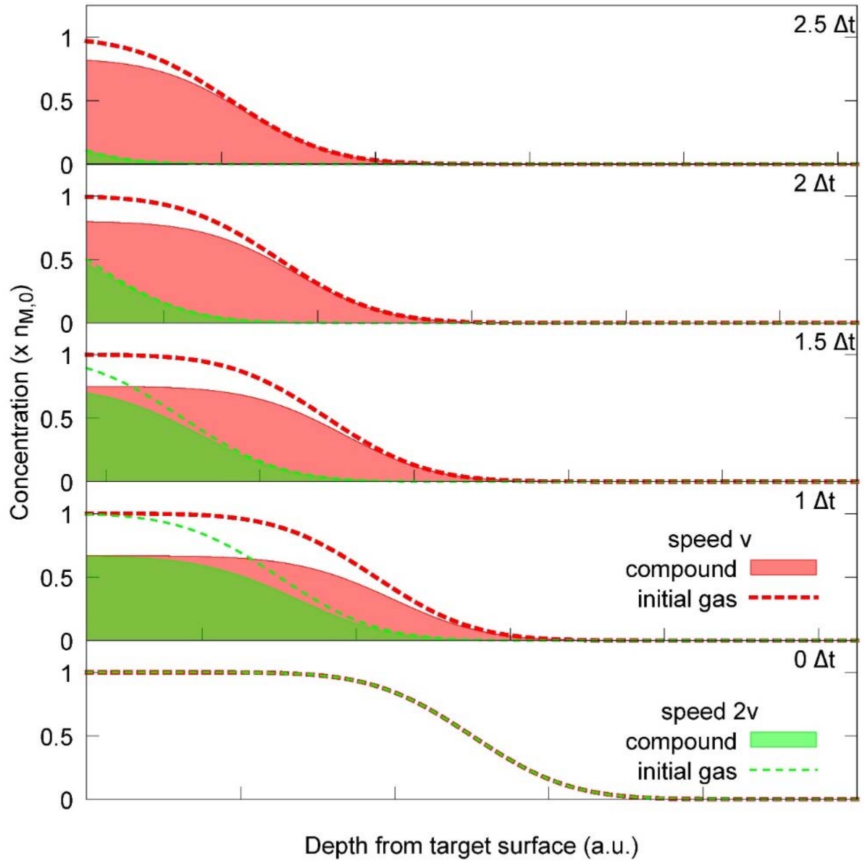
Gas buildup by Gaussian implantation

$$\frac{dn_R(x,t)}{dt} = \frac{2fj_{ion}}{v_s}p(x)$$



Reaction (same initial gas concentration)

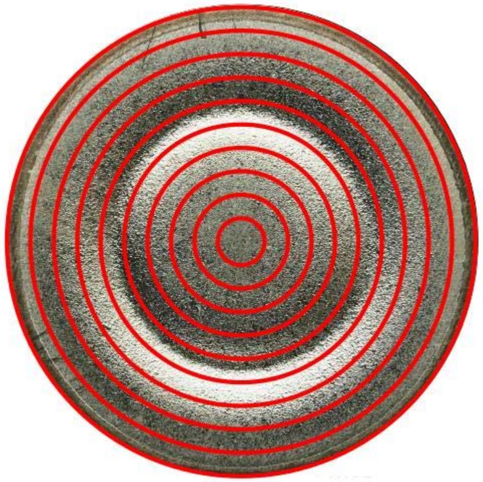
$$\frac{dn_R(x,t)}{dt} = -\frac{k}{v_s}n_M(x,t)n_R(x,t)$$

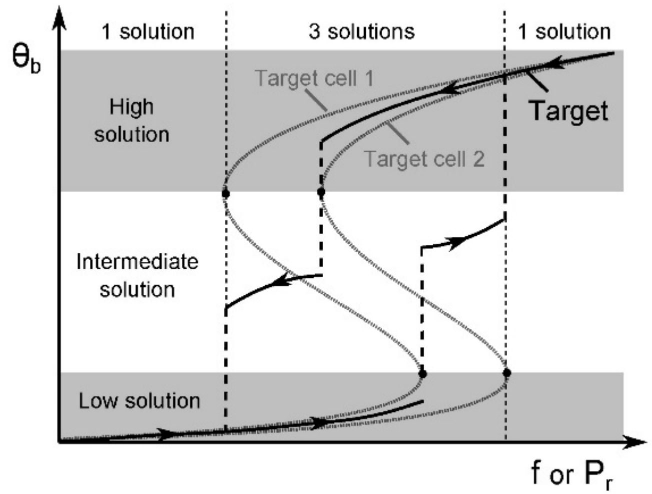


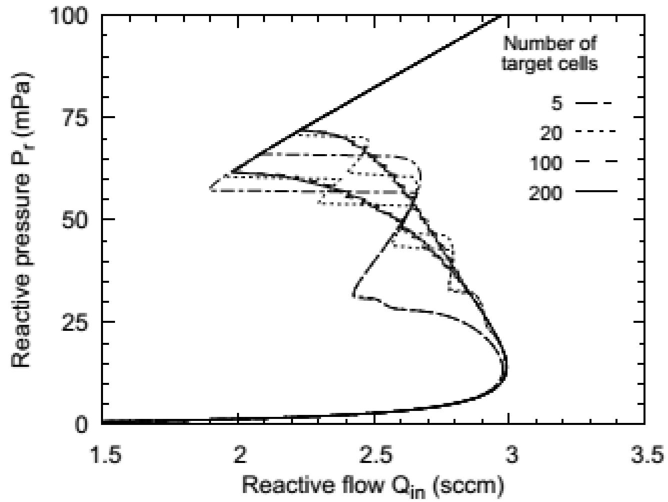
From one- to multi-cell target

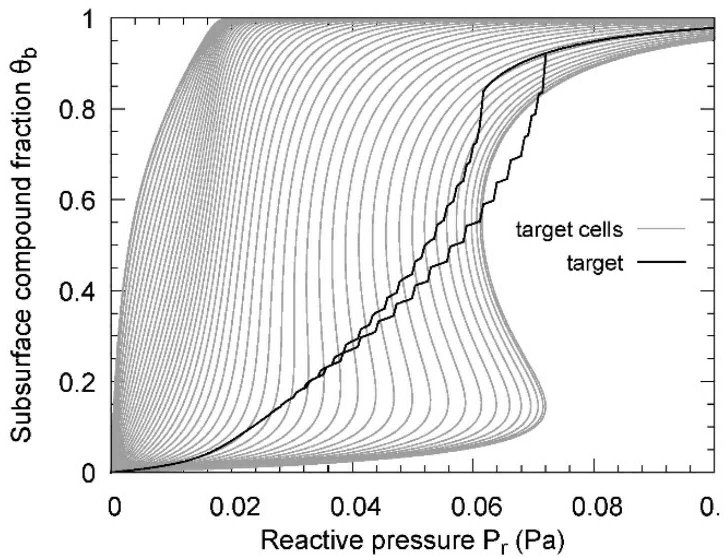
Current on target is non-uniform!

Radial current profile must be included in the model.









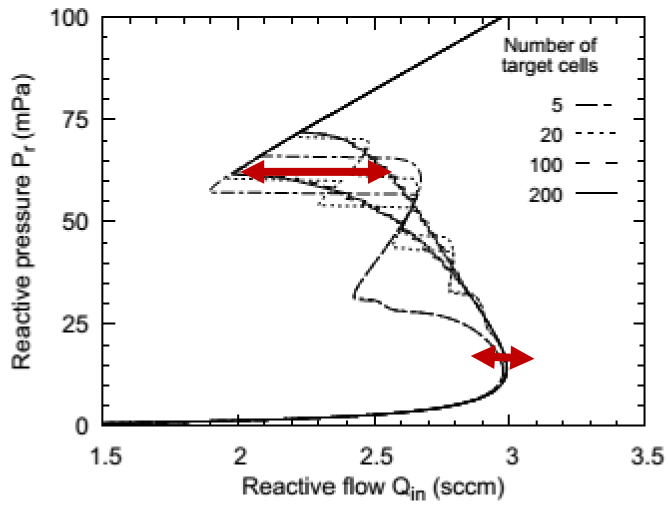
Combination of multiple individual target cells gives us the double S-shape.

Are we there yet?

… one can question if the proposed reaction mechanisms are the only cause of the double S-shaped hysteresis?

We think there is more because …

- Modelled hysteresis is double but separation seems to weak at first critical and to strong at second critical point



- Implanted reactive gas concentration will influence the sputter yield by
  - differences in collision cascade
  - diluting the metal concentration

Outline

Conclusion

- Although present in literature, the phenomena of a **double S-shaped hysteresis** is largely **ignored**.
- A **novel experimental procedure** to retrieve this double S-shaped hysteresis eliminating several possible artifacts is proposed for the Al/O_2 system.
- The **origin** of this additional critical behavior can be linked with the **implantation** and **reaction** of reactive gas.
- The RSD model predicted this **avalanche effect** as a function of the **mole fraction**.
  .... but the experimental match is not (yet) perfect.
- **Efforts in modelling** hope to unravel the complete story!

Acknowledgements

Contributing colleagues:

Roeland Schelfhout
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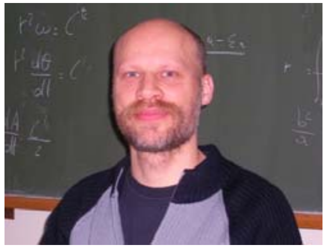
Diederik Depla
... for the positive guiding



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