# On the role of recycling in high power impulse magnetron sputtering discharges

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#### Introduction

 Magnetron sputtering has been a highly successfull technique that is essential in a number of industrial applications



- Conventional dc magnetron sputtering (dcMS) suffers from a low degree of ionization of the sputtered material
- High power impulse magnetron sputtering (HiPIMS) provides a highly ionized material flux, while being compatible with conventional magnetron sputtering deposition systems



#### High power impulse magnetron sputtering discharge

- High ionization of sputtered material requires very high density plasma
- In a conventional dc magnetron sputtering discharge the power density (plasma density) is limited by the thermal load on the target
- High power pulsed magnetron sputtering (HPPMS)
- In a HiPIMS discharge a high power pulse is supplied for a short period
  - Iow frequency
  - Iow duty cycle
  - low average power



Gudmundsson et al. (2012) JVSTA 30 030801

- Power density limits
  - $p_{t} = 0.05 \text{ kW/cm}^{2} \text{ dcMS}$  limit
  - $p_t = 0.5 \text{ kW/cm}^2 \text{ HiPIMS limit}$



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# Ionization region model studies of HiPIMS discharges



- The ionization region model (IRM) was developed to improve the understanding of the plasma behaviour during a HiPIMS pulse and the afterglow
- The main feature of the model is that an ionization region (IR) is defined next to the race track
- The IR is defined as an annular cylinder with outer radii  $r_{c2}$ , inner radii  $r_{c1}$  and length  $L = z_2 - z_1$ , extends from  $z_1$  to  $z_2$  axially away from the target



The definition of the volume covered by the IRM From Raadu et al. (2011) PSST 20 065007



- The temporal development is defined by a set of ordinary differential equations giving the first time derivatives of
  - the electron energy
  - the particle densities for all the particles
- The species assumed in the of-IRM are
  - cold electrons e<sup>C</sup> (Maxwellian), hot electrons e<sup>H</sup> (sheath acceleration)
  - argon atoms Ar(3s<sup>2</sup>3p<sup>6</sup>), warm argon atoms in the ground state Ar<sup>W</sup>, hot argon atoms in the ground state Ar<sup>H</sup>, Ar<sup>m</sup> (1s<sub>5</sub> and 1s<sub>3</sub>) (11.6 eV), argon ions Ar<sup>+</sup> (15.76 eV)
  - titanium atoms Ti(a<sup>3</sup>F), titanium ions Ti<sup>+</sup> (6.83 eV), doubly ionized titanium ions Ti<sup>2+</sup> (13.58 eV)
  - aluminium atoms Al(<sup>2</sup>P<sub>1/2</sub>), aluminium ions Al<sup>+</sup> (5.99 eV), doubly ionized aluminium ions Al<sup>2+</sup> (18.8 eV)

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- The model is constrained by experimental data input and fitted to reproduce the measured discharge current and voltage curves, *I*<sub>D</sub>(*t*) and *V*<sub>D</sub>(*t*), respectively
- Two model fitting parameters were found to be sufficient for a discharge with Al target
  - *V*<sub>IR</sub> accounts for the power transfer to the electrons
  - β is the probability of back-attraction of ions to the target

From Huo et al. (2017) JPD 50 354003

Experimental data from Anders et al. (2007) JAP 102 113303





- A non-reactive discharge with 50 mm diameter Al target
- Current composition at the target surface



- When the discharge is operated at 400 V the contributions of Al<sup>+</sup> and Ar<sup>+</sup>-ions to the discharge current are very similar
- At 800 V Al<sup>+</sup>-ions dominate the discharge current (self-sputtering) while the contribution of Ar<sup>+</sup> is below 10 % except at the initiation of the pulse

From Huo et al. (2017) JPD 50 354003

Experimental data from Anders et al. (2007) JAP 102 113303



- A primary current *I*<sub>prim</sub> is defined as ions of the working gas, here Ar<sup>+</sup>, that are ionized for the first time and then drawn to the target
- This is the dominating current in dc magnetron sputtering discharges
- This current has a critical upper limit

$$I_{\rm crit} = S_{
m RT} e p_{
m g} \sqrt{rac{1}{2\pi m_{
m g} k_{
m B} T_{
m g}}} = S_{
m RT} e n_{
m g} \sqrt{rac{k_{
m B} T_{
m g}}{2\pi m_{
m g}}}$$

 Discharge currents *I*<sub>D</sub> above *I*<sub>crit</sub> are only possible if there is some kind of recycling of atoms that leave the target, become subsequently ionized and then are drawn back to the target



- For the 50 mm diameter AI target the critical current is  $I_{\rm crit} \approx$  7 A
- The experiment is operated from far below *I*<sub>crit</sub> to high above it, up to 36 A.
- With increasing current *I*<sub>prim</sub> gradually becomes a very small fraction of the total discharge current *I*<sub>D</sub>
- The current becomes mainly carried by singly charged Al<sup>+</sup>-ions, meaning that self-sputter recycling or the current I<sub>SS-recycle</sub> dominates

From Huo et al. (2017) JPD 50 354003

Experimental data from Anders et al. (2007) JAP 102 113303



- Recall that singly charged metal ions cannot create the secondary electrons – for metal self-sputtering (γ<sub>SE</sub> is practically zero)
- The first ionization energies of many metals are insufficient to overcome the workfunction of the target material
- For the discharge with AI target operated at high voltage, self-sputter dominated, the effective secondary electron emission is essentially zero



From Anders (2008) APL 92 201501



#### Reactive HiPIMS

- Ar/O<sub>2</sub> discharge with Ti target
- In the metal mode Ar<sup>+</sup> and Ti<sup>+</sup>-ions contribute roughly equally to the current – combined self-sputter recycling and working gas recycling
- In the poisoned mode the current increaes and Ar<sup>+</sup>-ions dominate the current – working gas recycling

From Gudmundsson et al. (2016) PSST 25(6) 065004



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- For the AI target, Ohmic heating is in the range of 87 % (360 V) to 99 % (1000 V)
- The domination of Al<sup>+</sup>-ions, which have zero secondary electron emission yield, has the consequence that there is negligible sheath energization
- The ionization threshold for twice ionized Al<sup>2+</sup>, 18.8 eV, is so high that few such ions are produced



From Huo et al. (2017) JPD 50 354003



- For a Ti target Ohmic heating is about 92 %
  - Both Ar<sup>+</sup> and Ti<sup>2+</sup>-ions contribute to creation of secondary electrons
- For Ti target in Ar/O<sub>2</sub> mixture
  - In the metal mode Ohmic heating is found to be 90 % during the plateau phase of the discharge pulse
  - For the poisoned mode Ohmic heating is 70 % with a decreasing trend, at the end of the pulse



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#### The generalized recycling model



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# Generalized recycling

A working gas-sputtering parameter

 $\pi_{\rm g} = \alpha_{\rm g} \beta_{\rm g} \xi_{\rm pulse}$ 

#### where

- $\alpha_{\rm g}$  is ionization probability
- $\beta_{g}$  is back attraction probability
- $\xi_{\text{pulse}} = 1$  is return fraction in a pulse
- The total current carried by working gas ions

$$I_{g} = I_{prim} + I_{gas-recycle} = I_{prim} \left( 1 + \frac{\pi_{g}}{1 - \pi_{g}} \right)$$



From Brenning et al. (2017) PSS

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• The total self-sputter current is

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

where the self-sputter parameter is

$$\pi_{\rm SS} = \alpha_{\rm t} \beta_{\rm t} \, \mathbf{Y}_{\rm SS}$$

The total discharge current is

$$I_{\rm D} = I_{\rm prim} + I_{\rm gas-recycle} + I_{\rm SS}$$
$$= I_{\rm prim} \left(1 + \frac{\pi_{\rm g}}{1 - \pi_{\rm g}}\right) \left(1 + \frac{Y_{\rm g}}{Y_{\rm SS}} \frac{\pi_{\rm SS}}{1 - \pi_{\rm S}}\right)$$





The discharge current

 $\textit{I}_{D} = \textit{I}_{prim}\Pi_{gas-recycle}\Pi_{SS-recycl}$ 

- *I*<sub>prim</sub> is the seed current acts as a seed to the whole discharge current and has an upper limit *I*<sub>crit</sub>
- *I*<sub>prim</sub>Π<sub>gas-recycle</sub> is the seed current for the self-sputter process
- If  $\pi_{SS} > 1$  the discharge goes into SS-runaway



- Recycling map
- A graph in which the ion current mix of *I*<sub>prim</sub>, *I*<sub>gas-recycle</sub>, and *I*<sub>SS</sub> to the target in a magnetron discharge is defined by a point
- The value of  $I_{\rm prim}/I_{\rm D}=39$  %, can be read on the diagonal lines ( $Y_{\rm SS}=0.5$ )
- $I_{\rm prim}/I_{\rm D}=0.85$  defines the dcMS regime
- For  $I_{\rm SS}/I_{\rm D} > 0.5$  we have the SS-recycle dominated range A
- For  $I_{\text{gas-recycle}}/I_{\text{D}} > 0.5$  we have the gas-recycle dominated range B



- The discharge with AI target moves from the dcMS regime to the HiPIMS discharge regime with increased discharge voltage – type A
- A discharge with carbon target jumps from the dcMS regime to the HiPIMS regime – both SS recycling and working gas recycling play a role – intermediate type AB



 Recycling map for five different targets with varying self-sputter yield

• 
$$Cu - Y_{SS} = 2.6$$

• 
$$A_{I} - Y_{SS} = 1.1$$
  
•  $T_{I} - Y_{SS} = 0.7$ 

• 
$$\Pi = V_{SS} = 0.7$$

• 
$$TiO_2 - Y_{SS} = 0.04 - 0.25$$

- For very high self-sputter yields Y<sub>SS</sub> > 1, the discharges above *l*<sub>crit</sub> are of type A with dominating SS-recycling
- For very low self-sputter yields Y<sub>SS</sub> < 0.2, the discharges above I<sub>crit</sub> are of type B with dominating working gas recycling





PSST 26 125003



- Recycling loops
- Discharge with AI target SS recycling dominates
  - high self sputter yield
- Reactive discharge with TiO<sub>2</sub> target working gas recycling dominates
  - low self sputter yield



#### HiPIMS - Voltage - Current - time

- For Ar/O<sub>2</sub> discharge with Ti target
- At high frequencies, oxide is not able to form between pulses, and self-sputtering recycling by Ti<sup>+</sup>-ions is the dominant process
- At low frequency, the long off-time results in an oxide layer being formed (TiO<sub>2</sub>) on the target surface and working gas recycling dominates – triangular current waveform



Magnus et al. (2012), JVSTA 30 05060

# Summary

- For high currents the discharge with AI target develops almost pure **self-sputter recycling**, while the discharge with Ti target exhibits close to a 50/50 combination of **self-sputter recycling** and **working gas-recycling**
- For very high self-sputter yields, above approximately  $Y_{\rm SS} \approx 1$ , the discharges above  $I_{\rm crit}$  are of type A with
  - dominating SS-recycling
  - very little secondary electron emission
  - little sheath energization of electrons
- For very low self-sputter yields, below approximately
  - $Y_{SS} \approx$  0.2, the discharges above  $\textit{I}_{crit}$  are of type B with
    - dominating working gas recycling
    - significant secondary electron emission
    - significant sheath energization of electrons.
- The fraction of the total electron heating that is attributed to Ohmic heating is over 90 % in the HiPIMS discharge



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# Thank you for your attention



The slides can be downloaded at

http://langmuir.raunvis.hi.is/~tumi/ranns.html
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