On recycling in high power impulse magnetron sputtering discharges

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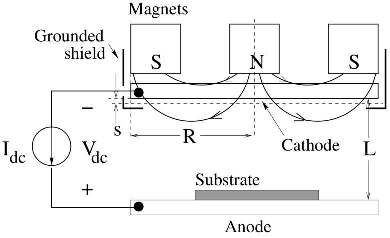
Introduction

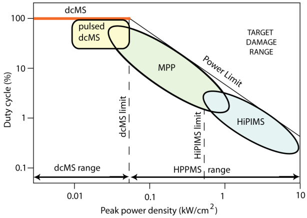
- Magnetron sputtering has been a highly sucessfull technique that is essential in a number of industrial applications

- A magnet is placed at the back of the cathode target with the pole pieces at the center and perimeter
- The magnetic field confines the energetic electrons near the cathode
- The electrons undergo numerous ionizing collisions before being lost to a grounded surface



High power impulse magnetron sputtering discharge

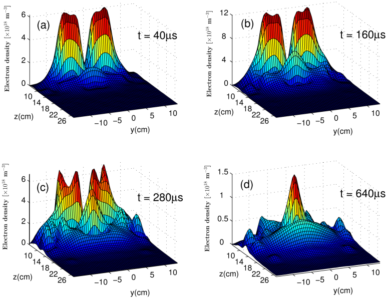
- High ionization of sputtered material requires very high density plasma
- In a conventional dc magnetron sputtering discharge the power density (plasma density) is limited by the thermal load on the target
- High power pulsed magnetron sputtering (HPPMS)
- In a HiPIMS discharge a high power pulse is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



Gudmundsson et al. (2012) JVSTA **30** 030801

- Power density limits
  $p_t = 0.05$ kW/cm^2 dcMS limit
  $p_t = 0.5$ kW/cm^2 HiPIMS limit

High power impulse magnetron sputtering discharge



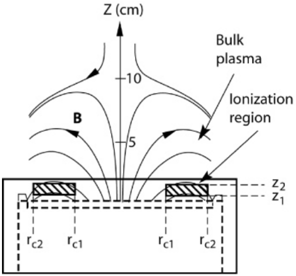
(After Bohlmark et al. (2005), IEEE Trans. Plasma Sci. **33** 346)

- Temporal and spatial variation of the electron density
- Ar discharge at 20 mTorr, Ti target, pulse length 100 μs
- The electron density in the substrate vicinity is of the order of $10^{18} - 10^{19}$ m^{-3} – ionization mean free path $\lambda_{\rm iz} \sim 1$ cm

Ionization region model studies of HiPIMS discharges

Ionization region model of HiPIMS

- 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

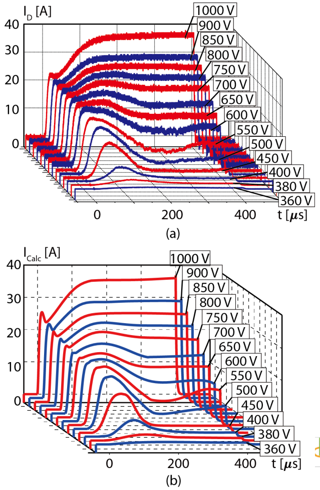
Ionization region model of HiPIMS

- 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^{C} (Maxwellian), hot electrons e^{H} (sheath acceleration)
  - argon atoms $Ar(3s^2 3p^6)$, warm argon atoms in the ground state Ar^{W}, hot argon atoms in the ground state Ar^{H}, Ar^{m} ($1s_5$ and $1s_3$) (11.6 eV), argon ions Ar^{+} (15.76 eV)
  - titanium atoms Ti(a ^{3}F), titanium ions Ti^{+} (6.83 eV), doubly ionized titanium ions Ti^{2+} (13.58 eV)
  - aluminium atoms $Al(^2P_{1/2})$, aluminium ions Al^{+} (5.99 eV), doubly ionized aluminium ions Al^{2+} (18.8 eV)

Detailed model description is given in Huo et al. (2017) JPD **50** 354003

Ionization region model of HiPIMS

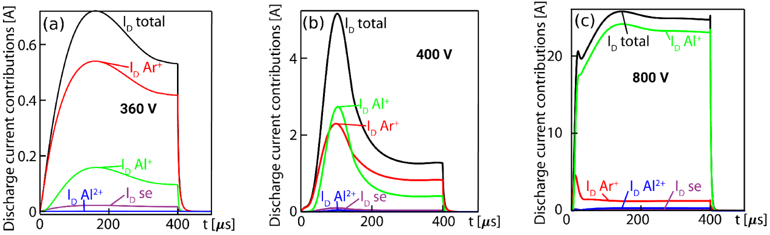
- The model is constrained by experimental data input and fitted to reproduce the measured discharge current and voltage curves, $I_D(t)$ and $V_D(t)$, respectively
- Two model fitting parameters were found to be sufficient for a discharge with Al target
  - V_{IR} accounts for the power transfer to 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

Ionization region model of HiPIMS



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

From Huo et al. (2017) JPD **50** 354003

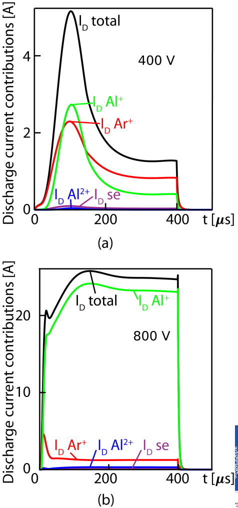
Experimental data from Anders et al. (2007) JAP **102** 113303

Ionization region model of HiPIMS

- When the discharge is operated at 400 V the contributions of Al^+ and Ar^+-ions to the discharge current are very similar
- At 800 V Al^+-ions dominate the discharge current (**self-sputtering**) while the contribution of Ar^+ 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



Ionization region model of HiPIMS

- A primary current $I_{\rm prim}$ is defined as ions of the working gas, here Ar^{+}, 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

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

- Discharge currents $I_{\rm D}$ above $I_{\rm crit}$ 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

Anders et al. (2012) JPD **45** 012003

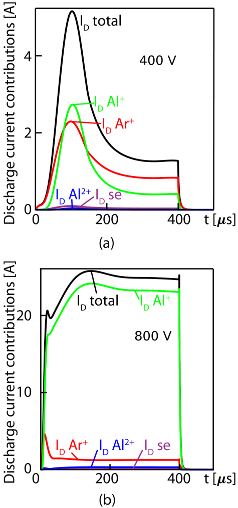
Huo et al. (2014) PSST **23** 025017

Ionization region model of HiPIMS

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

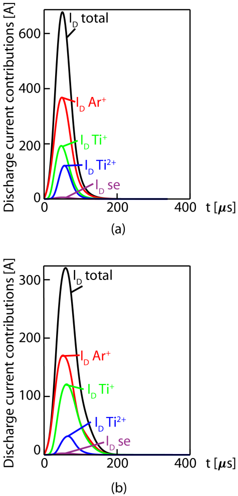
From Huo et al. (2017) JPD **50** 354003

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



Ionization region model of HiPIMS

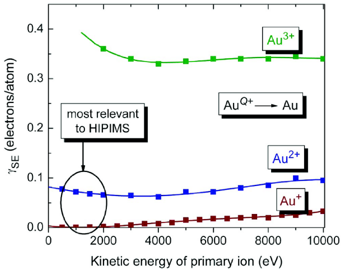
- For discharges with Ti target the peak current is far above the critical current (up to 650 A, while $I_{\rm crit} \approx 19$ A)
- However, this discharge shows close to a 50/50 combination of **self-sputter recycling** $I_{\rm SS-recycle}$ and **working gas-recycling** $I_{\rm gas-recycle}$
- Almost 2/3 of the current to the target is here carried by Ar^{+} and Ti^{2+}-ions, which both can emit secondary electrons upon target bombardment, and this gives a significant sheath energization



From Huo et al. (2017) JPD **50** 354003

Ionization region model of HiPIMS

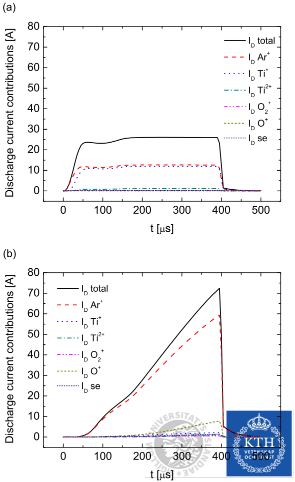
- Recall that singly charged metal ions cannot create the secondary electrons – for metal self-sputtering (γ_{SE} is practically zero)
- The first ionization energies of many metals are insufficient to overcome the workfunction of the target material
- For the discharge with Al target operated at high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



From Anders (2008) APL **92** 201501

Ionization region model of HiPIMS

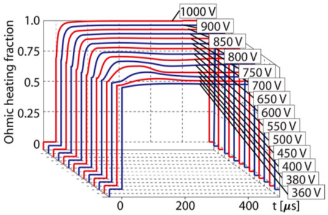
- **Reactive** HiPIMS
- Ar/O_2 discharge with Ti target
- For this system $I_{crit} \approx 5$ A
- In the metal mode Ar^+ and Ti^+-ions contribute roughly equally to the current – combined **self-sputter recycling** and **working gas recycling**
- In the poisoned mode the current increaes and Ar^+-ions dominate the current – **working gas recycling**



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

Ionization region model of HiPIMS

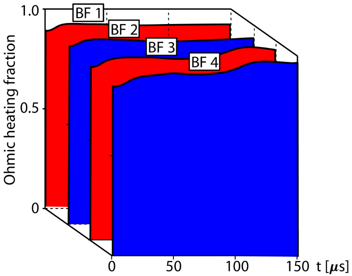
- For the Al target, Ohmic heating is in the range of 87 % (360 V) to 99 % (1000 V)
- The domination of Al^{+}-ions, which have zero secondary electron emission yield, has the consequence that there is negligible sheath energization
- The ionization threshold for twice ionized Al^{2+}, 18.8 eV, is so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



From Huo et al. (2017) JPD **50** 354003

Ionization region model of HiPIMS

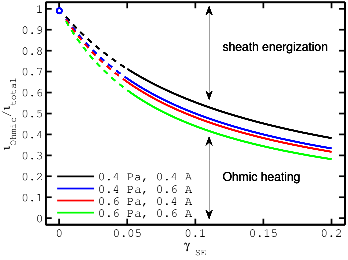
- For a Ti target Ohmic heating is about 92 %
  - Both Ar^{+} and Ti^{2+}-ions contribute to creation of secondary electrons
- For Ti target in Ar/O_2 mixture
  - In the metal mode Ohmic heating is found to be 90 % during the plateau phase of the discharge pulse
  - For the poisoned mode Ohmic heating is 70 % with a decreasing trend, at the end of the pulse



From Huo et al. (2017) JPD **50** 354003

Ionization region model of HiPIMS

- Ohmic heating is also very significant in dc magnetron sputtering discharges
- The relative contributions to the total ionization ι_{total} due to Ohmic heating, ι_{Ohmic}, and sheath energization, ι_{sheath}
- A blue circle marks the HiPIMS study modelled by Huo et al. (2013)
- Note that this HiPIMS case $\gamma_{\text{SE,eff}}$ is consistent with the dcMS cases



From Brenning et al. (2016) PSST **25** 065024

The generalized recycling model

Generalized recycling

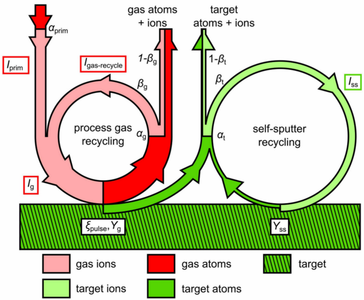
- A working gas-sputtering parameter

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

where

  - α_g is ionization probability
  - β_g is back attraction probability
  - $\xi_{pulse} = 1$ 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) PSST **26** 125003

Generalized recycling

- The total self-sputter current is

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

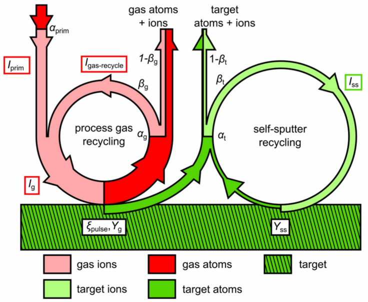
where the self-sputter parameter is

$$\pi_{\rm SS} = \alpha_{\rm t}\beta_{\rm t}Y_{\rm SS}$$

- The total discharge current is

$$I_{\rm D} = I_{\rm prim} + I_{\rm gas-recycle} + I_{\rm SS}$$

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



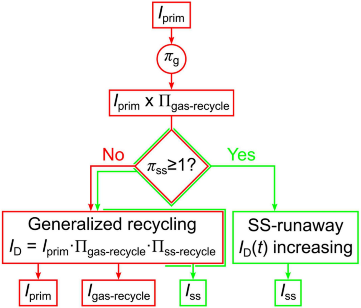
From Brenning et al. (2017) PSST **26** 125003.

Generalized recycling

- The discharge current

$$I_{\mathrm{D}} = I_{\mathrm{prim}} \Pi_{\mathrm{gas-recycle}} \Pi_{\mathrm{SS-recycl}}$$

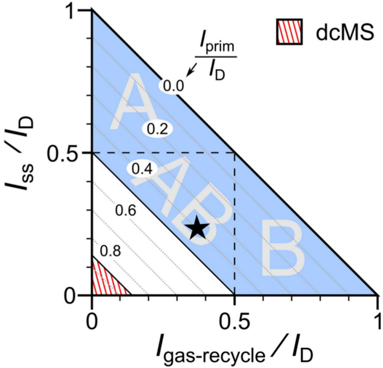
- I_{prim} is the seed current acts as a seed to the whole discharge current and has an upper limit I_{crit}
- $I_{\mathrm{prim}} \Pi_{\mathrm{gas-recycle}}$ is the seed current for the self-sputter process
- If $\pi_{\mathrm{SS}} > 1$ the discharge goes into SS-runaway



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

Generalized recycling

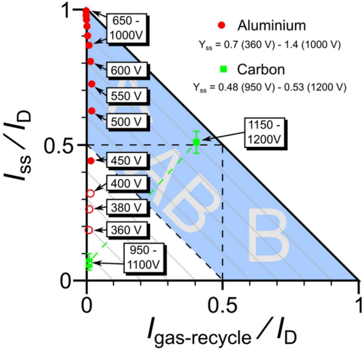
- Recycling map
- A graph in which the ion current mix of $I_{\rm prim}$, $I_{\rm gas-recycle}$, and $I_{\rm SS}$ to the target in a magnetron discharge is defined by a point
- The value of $I_{\rm prim}/I_{\rm D} = 39$ %, can be read on the diagonal lines ($Y_{\rm SS} = 0.5$)
- $I_{\rm prim}/I_{\rm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



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

Generalized recycling

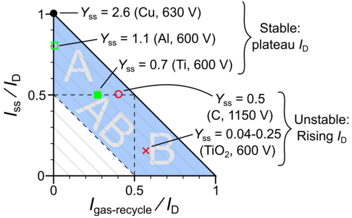
- The discharge with Al target moves from the dcMS regime to the HiPIMS discharge regime with increased discharge voltage – **type A**
- A discharge with carbon target jumps from the dcMS regime to the HiPIMS regime – both SS recycling and working gas recycling play a role – intermediate **type AB**



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

Generalized recycling

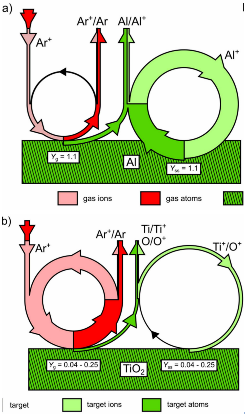
- Recycling map for five different targets with varying self-sputter yield
  - Cu – $Y_{SS} = 2.6$
  - Al – $Y_{SS} = 1.1$
  - Ti – $Y_{SS} = 0.7$
  - C – $Y_{SS} = 0.5$
  - TiO_2 – $Y_{SS} = 0.04 - 0.25$
- For very high self-sputter yields $Y_{SS} > 1$, the discharges above I_{crit} are of **type A** with dominating **SS-recycling**
- For very low self-sputter yields $Y_{SS} < 0.2$, the discharges above I_{crit} are of **type B** with dominating **working gas recycling**



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

Generalized recycling

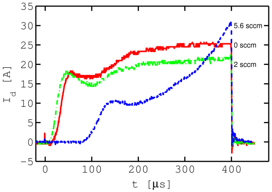
- Recycling loops
- Discharge with Al target – SS recycling dominates
  - high self sputter yield
- Reactive discharge with TiO_2 target – working gas recycling dominates
  - low self sputter yield

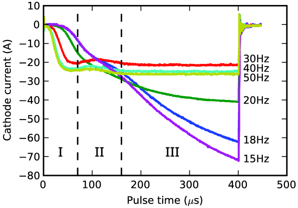


From Brenning et al. (2017) PSST **26** 125003

HiPIMS - Voltage - Current - time

- For Ar/O_2 discharge with Ti target
- At high frequencies, oxide is not able to form between pulses, and **self-sputtering recycling** by Ti^+-ions is the dominant process
- At low frequency, the long off-time results in an oxide layer being formed (TiO_2) on the target surface and **working gas recycling dominates** – triangular current waveform





From Gudmundsson (2016), PPCF **58** 014002

Magnus et al. (2012), JVSTA **30** 050601

Summary

- For high currents the discharge with Al 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 $Y_{\rm SS} \approx 0.2$, the discharges above $I_{\rm crit}$ 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 attributable to Ohmic heating is over 90 % in the HiPIMS discharge

Thank you for your attention

The slides can be downloaded at

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

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