

# The role of recycling in pulsed sputtering magnetrons

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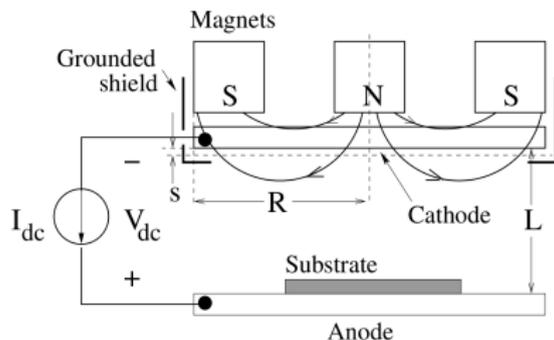
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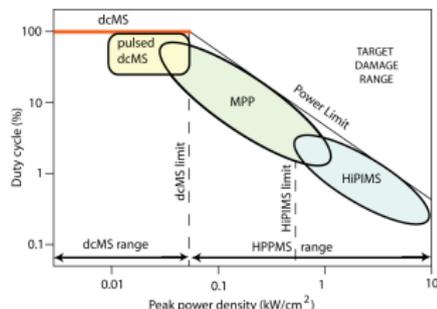
## Introduction

- Magnetron sputtering has been a highly successful 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

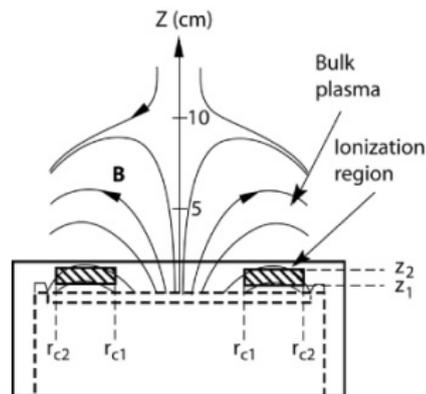
- Power density limits  
 $\rho_t = 0.05 \text{ kW/cm}^2$  dcMS limit  
 $\rho_t = 0.5 \text{ kW/cm}^2$  HiPIMS limit

# 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  $Ar(3s^23p^6)$ , 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  $Ti(a^3F)$ , 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)

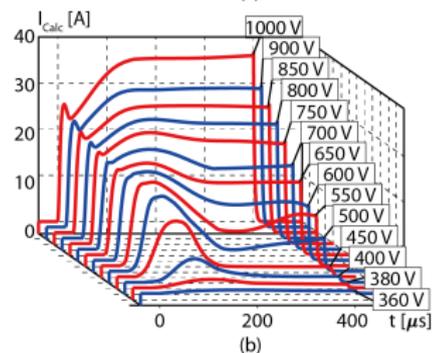
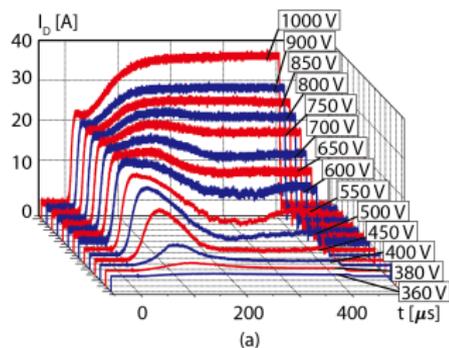


## ***Ionization region model of HiPIMS***

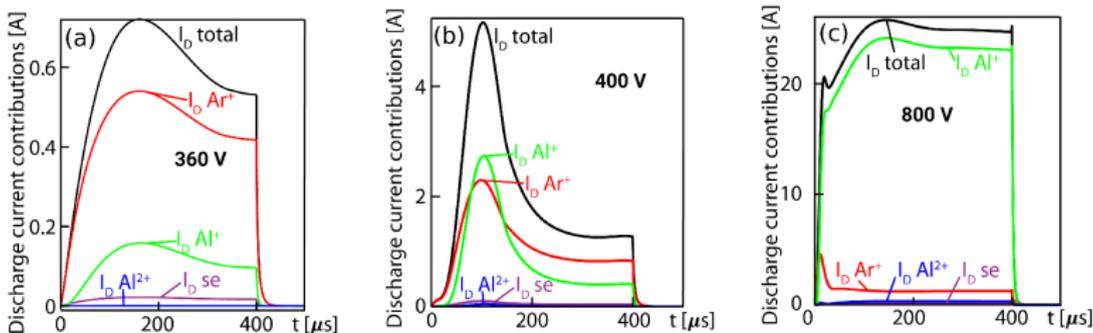
- The model is constrained by experimental data input and fitted to reproduce the measured discharge current and voltage curves,  $I_D(t)$  and  $V_D(t)$ , respectively
- Two model fitting parameters were found to be sufficient for a discharge with Al target
  - $V_{IR}$  accounts for the power transfer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

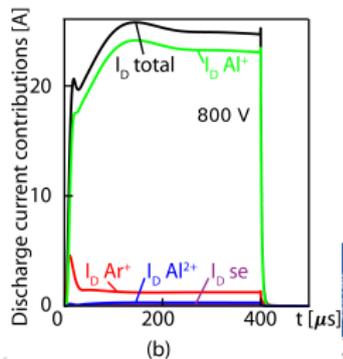
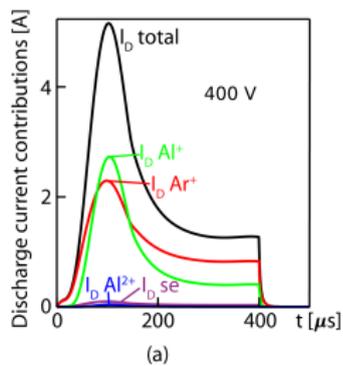


## ***Ionization region model of HiPIMS***

- When the discharge is operated at 400 V the contributions of  $\text{Al}^+$  and  $\text{Ar}^+$ -ions to the discharge current are very similar
- At 800 V  $\text{Al}^+$ -ions dominate the discharge current (**self-sputtering**) while the contribution of  $\text{Ar}^+$  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  $I_{\text{prim}}$  is defined as ions of the working gas, here  $\text{Ar}^+$ , 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

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

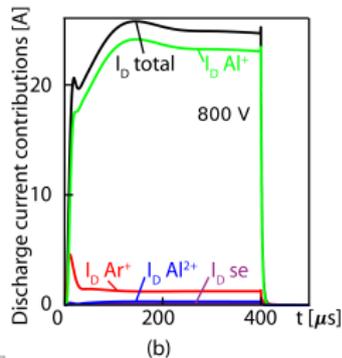
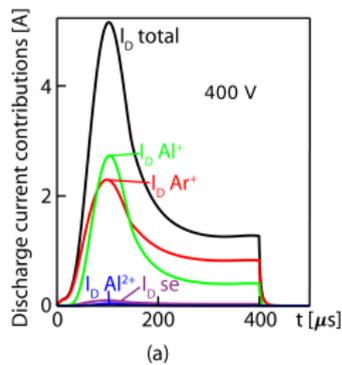
- Discharge currents  $I_{\text{D}}$  above  $I_{\text{crit}}$  are only possible if there is some kind of recycling of atoms that leave the target, become subsequently ionized and then are drawn back to the target

## ***Ionization region model of HiPIMS***

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

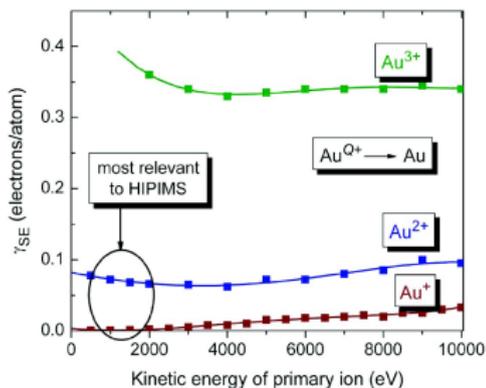
From Huo et al. (2017) JPD **50** 354003

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



## *Ionization region model of HiPIMS*

- Recall that singly charged metal ions cannot create the secondary electrons – for metal self-sputtering ( $\gamma_{SE}$  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



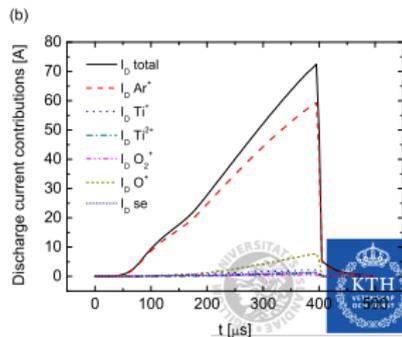
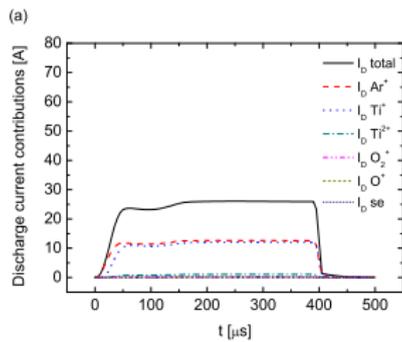
From Anders (2008) APL 92 201501



# *Ionization region model of HiPIMS*

- **Reactive HiPIMS**
- Ar/O<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 increases and Ar<sup>+</sup>-ions dominate the current – **working gas recycling**

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



# The generalized recycling model



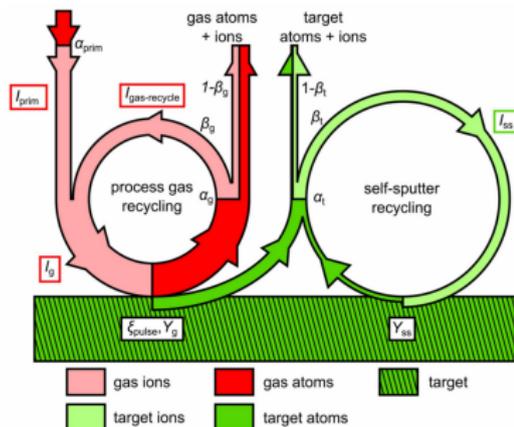
# Generalized recycling

- A working gas-sputtering parameter

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

where

- $\alpha_g$  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



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

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



## Generalized recycling

- The total self-sputter current is

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

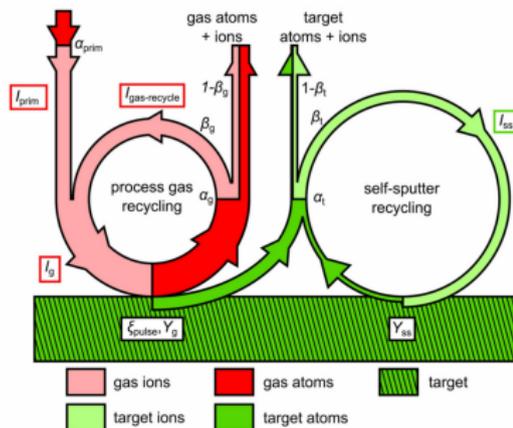
where the self-sputter parameter is

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

- The total discharge current is

$$I_D = I_{\text{prim}} + I_{\text{gas-recycle}} + I_{SS}$$

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



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

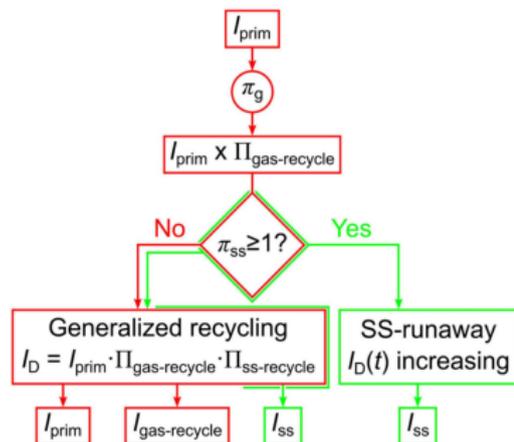


# Generalized recycling

- The discharge current

$$I_D = I_{\text{prim}} \Pi_{\text{gas-recycle}} \Pi_{\text{SS-recycle}}$$

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

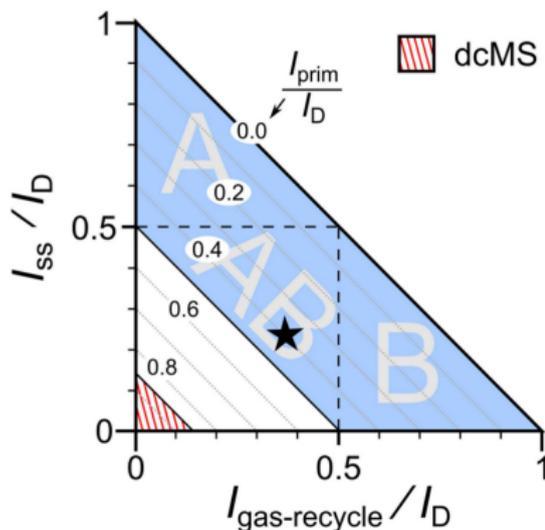


From Brenning et al. (2017) PSS-T 26 125003



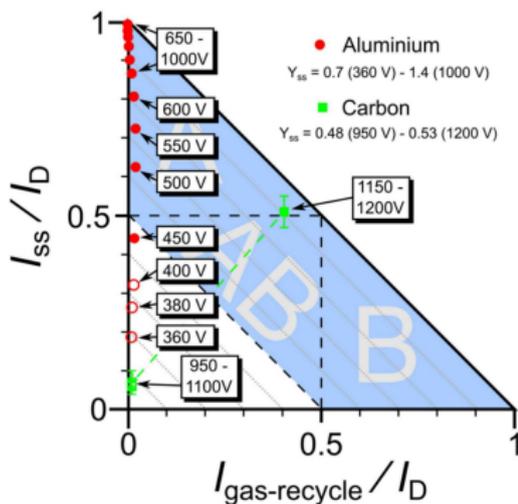
## Generalized recycling

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



## 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**

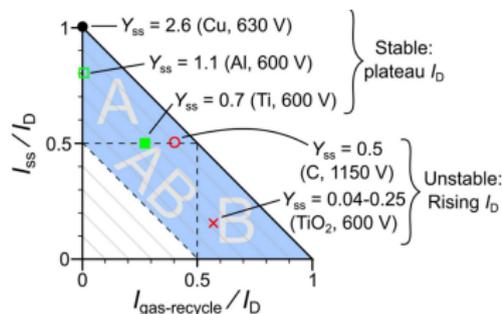


# Generalized recycling

- 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<sub>2</sub> –  $Y_{SS} = 0.04 - 0.25$

- 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**



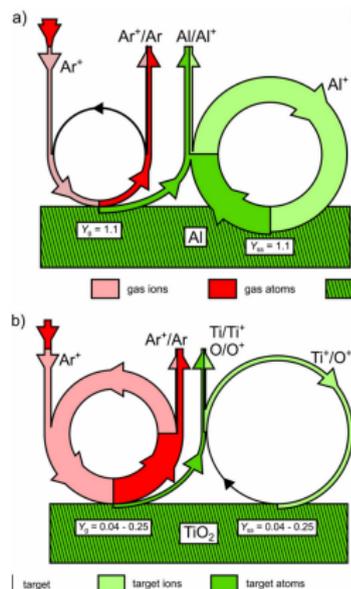
From Brenning et al. (2017),

PSST 26 125003



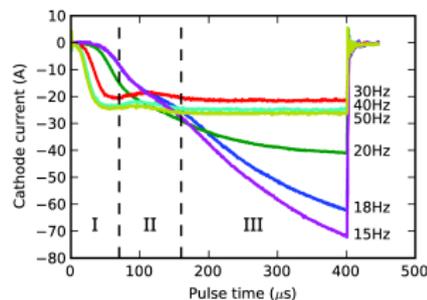
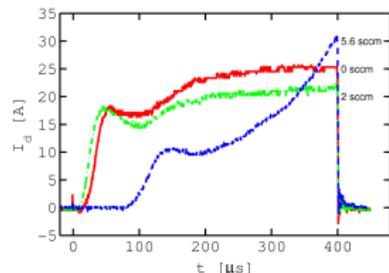
## Generalized recycling

- Recycling loops
- Discharge with Al target – SS recycling dominates
  - high self sputter yield
- Reactive discharge with  $\text{TiO}_2$  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



## 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  $Y_{SS} \approx 1$ , the discharges above  $I_{crit}$  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  $I_{crit}$  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 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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## References

- Anders, A., J. Andersson, and A. Ehasarian (2007). High power impulse magnetron sputtering: Current-voltage-time characteristics indicate the onset of sustained self-sputtering. *J. Appl. Phys.* **102**(11), 113303.
- Anders, A. (2008). Self-sputtering runaway in high power impulse magnetron sputtering: The role of secondary electrons and multiply charged metal ions. *Appl. Phys. Lett.* **92**(20), 201501.
- Anders, A., J. Čapek, M. Hála, and L. Martinu (2012). The 'recycling trap': a generalized explanation of discharge runaway in high-power impulse magnetron sputtering. *J. Phys D: Appl. Phys.* **45**(1), 012003.
- Bohlmark, J., J. T. Gudmundsson, J. Alami, M. Lattemann, and U. Helmersson (2005). Spatial electron density distribution in a high-power pulsed magnetron discharge. *IEEE Trans. Plasma Sci.* **33**(2), 346–347.
- Bradley, J. W., A. Mishra, and P. J. Kelly (2015). The effect of changing the magnetic field strength on HiPIMS deposition rates. *J. Phys. D: Appl. Phys.* **48**(21), 215202.
- Brenning, N., J. T. Gudmundsson, M. A. Raadu, T. J. Petty, T. Minea, and D. Lundin, (2017). A unified treatment of self-sputtering, process gas recycling, and runaway for high power impulse sputtering magnetrons. *Plasma Sources Sci. Technol.* **26**(12), 125003.
- Gudmundsson, J. T., N. Brenning, D. Lundin, and U. Helmersson (2012). The high power impulse magnetron sputtering discharge. *J. Vac. Sci. Technol. A* **30**(3), 030801.
- Gudmundsson, J. T., D. Lundin, N. Brenning, M. A. Raadu, C. Huo, and T. M. Minea (2016). An ionization region model of the reactive Ar/O<sub>2</sub> high power impulse magnetron sputtering discharge. *Plasma Sources Science and Technology* **25**(6), 065004.
- Gudmundsson, J. T. (2016). On reactive high power impulse magnetron sputtering. *Plasma Physics and Controlled Fusion* **58**(1), 014002.
- Huo, C., D. Lundin, J. T. Gudmundsson, M. A. Raadu, J. W. Bradley, and N. Brenning (2017). Particle-balance models for pulsed sputtering magnetrons. *J. Phys. D: Appl. Phys.* **50**(35), 354003.
- Huo, C., D. Lundin, M. A. Raadu, A. Anders, J. T. Gudmundsson, and N. Brenning (2014). On the road to self-sputtering in high power impulse magnetron sputtering: particle balance and discharge characteristics. *Plasma Sources Sci. Technol.* **23**(2), 025017.
- Magnus, F., T. K. Tryggvason, S. Olafsson, and J. T. Gudmundsson (2012). Current-voltage-time characteristics of the reactive Ar/O<sub>2</sub> high power impulse magnetron sputtering discharge. *Journal of Vacuum Science and Technology A* **30**(5), 050601.
- Raadu, M. A., I. Axnäs, J. T. Gudmundsson, C. Huo, and N. Brenning (2011). An ionization region model for high power impulse magnetron sputtering discharges. *Plasma Sources Sci. Technol.* **20**(6), 065007.

