On the deposition rate and ionized flux fraction in high power impulse magnetron sputtering

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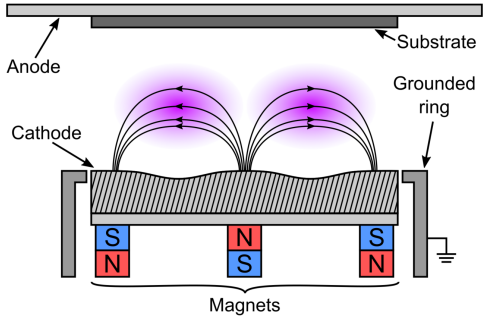
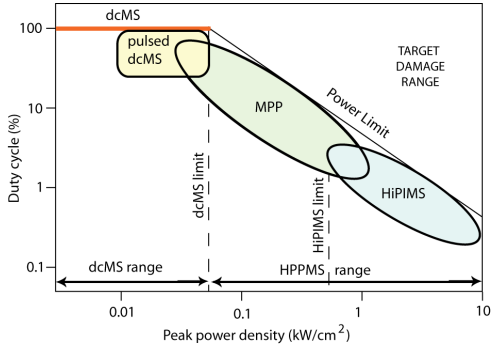
Introduction – Magnetron sputtering

- Magnetron sputtering is a highly successful and widely used technique for thin film deposition

Gudmundsson (2020) PSST **29** 113001



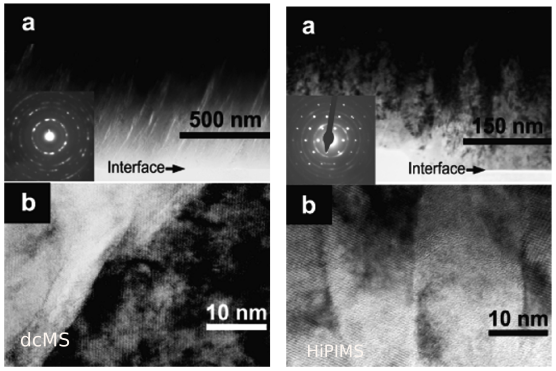


Gudmundsson and Lundin (2020) in High Power Impulse Magnetron Sputtering Discharge, Elsevier, 2020

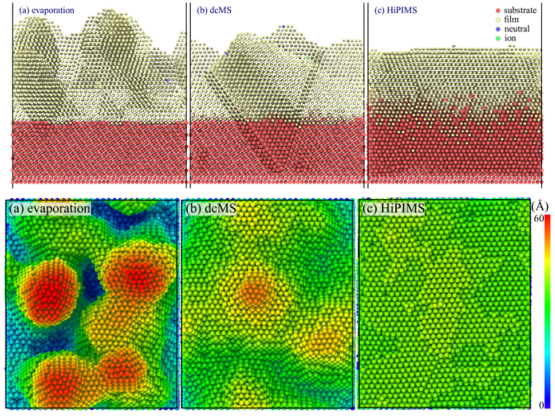
- High power impulse magnetron sputtering (HiPIMS)
  - pulses with high peak power
  - pulse length 50 – 400 μs
  - low repetition frequency (50 – 5000 Hz)
  - low duty cycle ($<$ 10 %)

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

Introduction – Thin film properties



Alami et al. (2005) JVSTA **23** 278



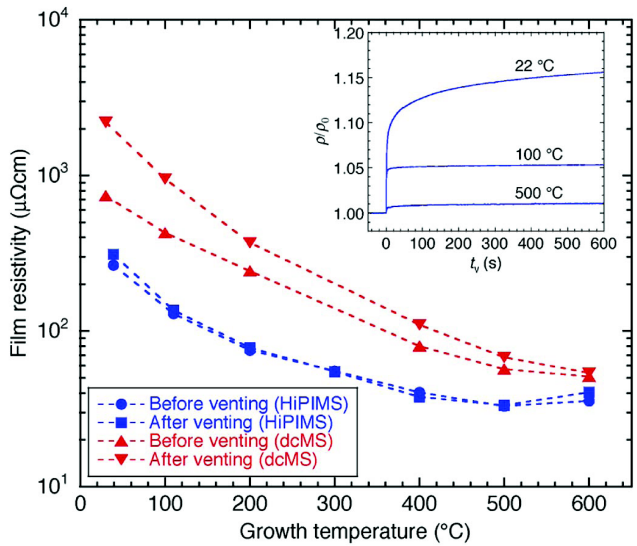
Kateb et al. (2019) JVSTA **37** 031306

- HiPIMS provides higher ionized flux fraction than dc magnetron sputtering (dcMS)
- Due to the higher fraction of ionization of the sputtered species
  - the films are smooth and dense
  - control over phase composition and microstructure is possible
  - enhanced mechanical, electrical and optical properties
  - improved film adhesion

Introduction – Thin film properties

- HiPIMS deposited TiN films have significantly lower resistivity than dcMS deposited films on SiO_2 at all growth temperatures
- Ultrathin continuous TiN films with
  - superior electrical characteristics
  - high resistance towards oxidation

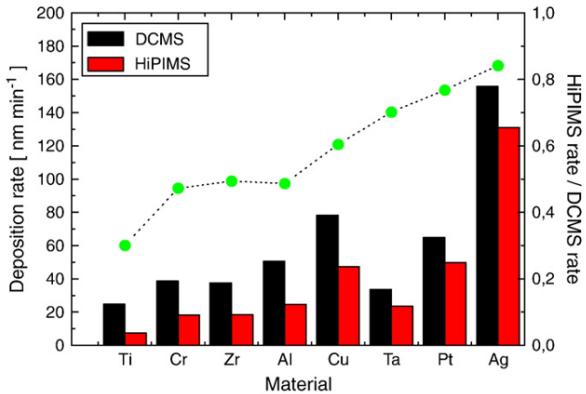
  can be obtained with HiPIMS at reduced temperatures



Magnus et al. (2012) IEEE EDL **33** 1045

Introduction – Deposition rate

- There is a drawback
- The deposition rate is lower for HiPIMS when compared to dcMS operated at the same average power
- The HiPIMS deposition rates are typically in the range of 30 – 85% of the dcMS rates depending on target material
- Many of the ions of the target material are attracted back to the target surface by the cathode potential

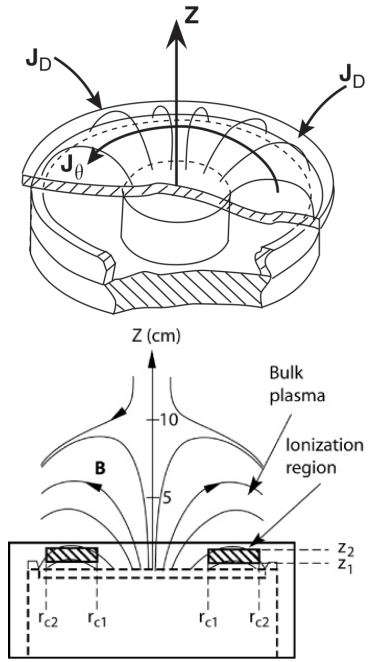


From Samuelsson et al. (2010) SCT **202** 591

Christie (2005) JVSTA **23** 330

Ionization region model of HiPIMS

- The ionization region model (IRM) is a time-dependent volume averaged plasma chemical model of the ionization region (IR) of the HiPIMS discharge
- It gives the temporal evolution of the densities of ions, neutrals and electrons
- 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

Detailed model description is given in

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

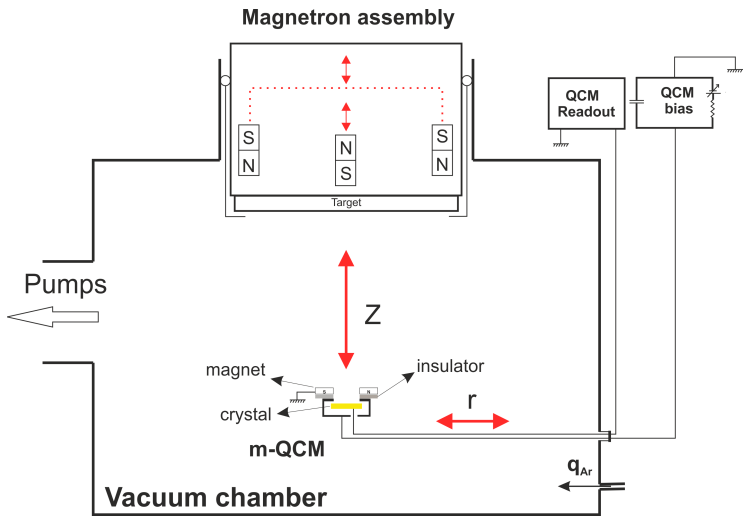
Experiment: Deposition rate and ionized flux fraction

Deposition rate

- For a titanium target the deposition rate and the ionized flux fraction are measured using a gridless ion meter (m-QCM)

Kubart et al. (2014) SCT **238** 152

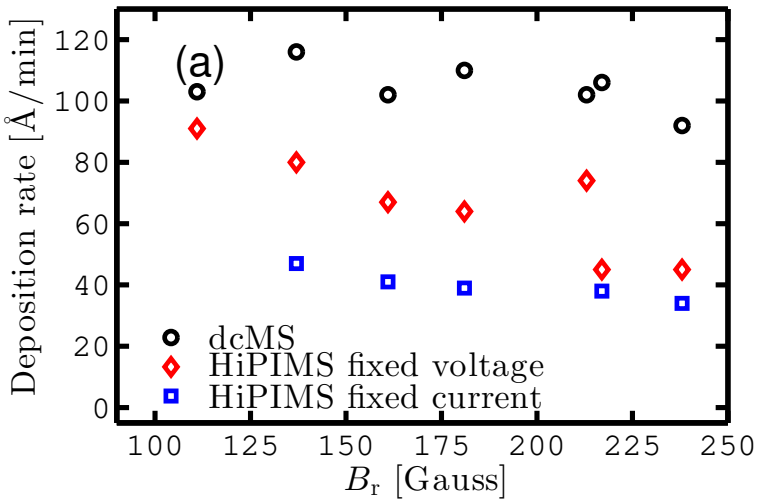
- The ion meter is mounted on a probe holder which can be moved around within the chamber
- The Ar working gas pressure was set to 1 Pa, the pulse width 100 μs and the average power 300 W
- The confining magnetic field is varied by moving the magnets



From Hajihoseini et al. (2019) Plasma **2** 201

Deposition rate

- The Ti deposition rate recorded at substrate position using a gridless ion meter (m-QCM)
  - **dcMS**
    +10% with decreasing |**B**|
    (but no obvious trend)
  - **HiPIMS fixed voltage**
    +110% with decreasing |**B**|
  - **HiPIMS fixed peak current**
    +40% with decreasing |**B**|
- In HiPIMS operation the deposition rate increases with decreasing |**B**|
- The recorded $|B_{r,\mathrm{rt}}|$ value above the race track is used as a measure of |**B**|

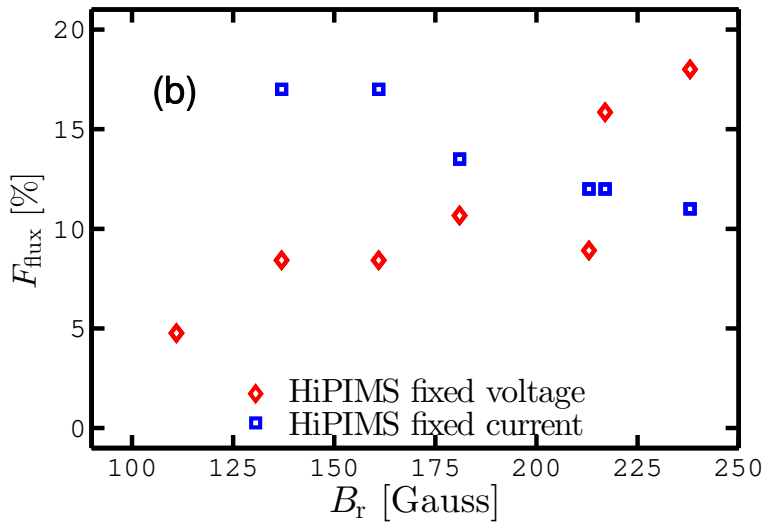


From Gudmundsson (2020) PSST **29**(11) 113001

based on Hajihoseini et al. (2019) Plasma **2** 201

Ionized flux fraction

- Ionized flux fraction recorded
  - **dcMS**
    Always around 0 %
    (Kubart et al., 2014)
  - **HiPIMS fixed voltage**
    -75% with decreasing $|\mathbf{B}|$
  - **HiPIMS fixed peak current**
    $+50\%$ with decreasing $|\mathbf{B}|$
- The ionized flux fraction decreases with decreasing $|\mathbf{B}|$ when the HiPIMS discharge is operated in fixed voltage mode but increases in fixed peak current mode
- Opposing trends



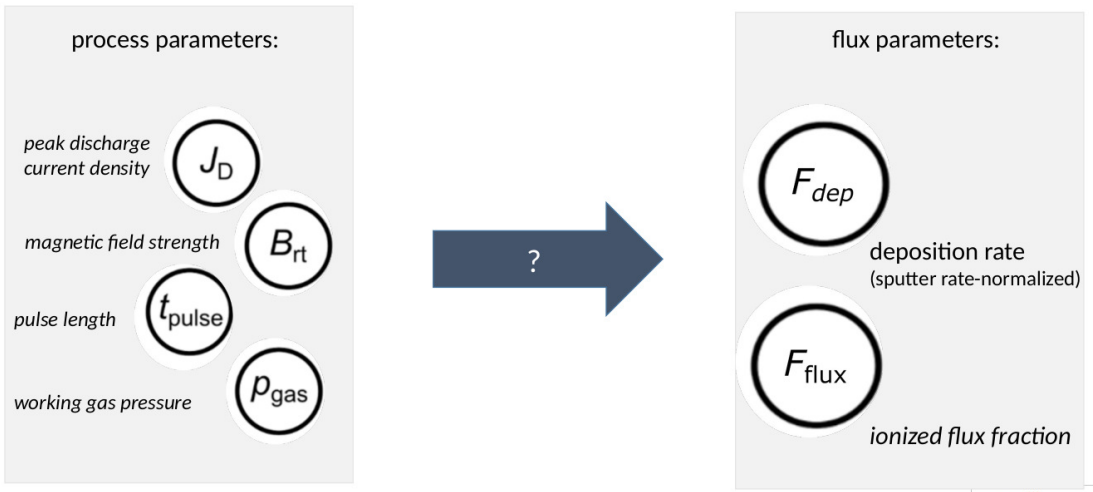
From Gudmundsson (2020) PSST **29**(11) 113001

based on Hajihoseini et al. (2019) *Plasma* **2** 201

Internal parameters and optimization

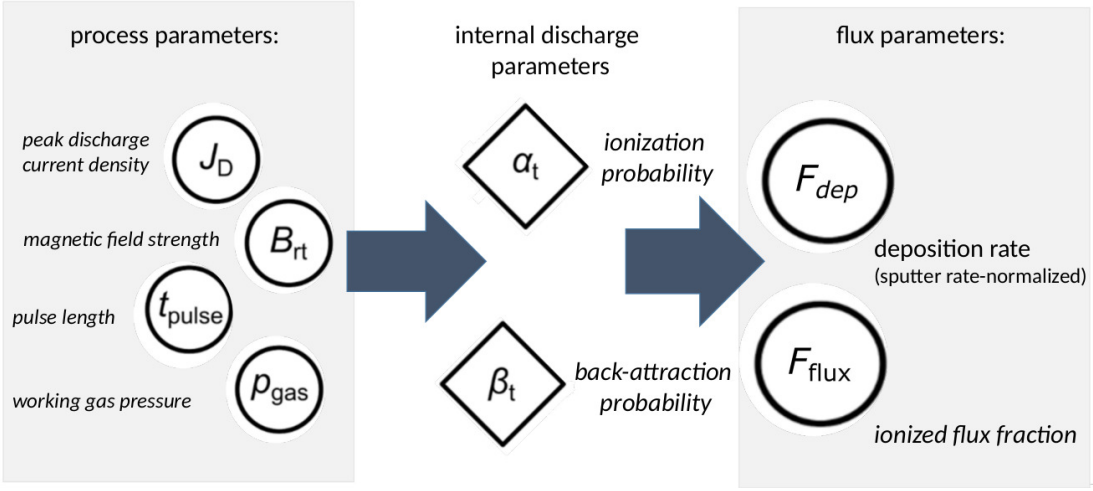
Deposition rate – α_t and β_t

- Low deposition rate is the main drawback of this sputter technology and hampers its use for industrial applications



- We want to relate the process parameters to the flux prameters – deposition rate and ionized flux fraction

Deposition rate – α_t and β_t



- Two internal parameters are of importance
  - α_t – ionization probability
  - β_t – back-attraction probability

Deposition rate – α_t *and* β_t

- We can relate the measured quantities normalized deposition rate $F_{\mathrm{DR,sput}}$ and the ionized flux fraction $F_{\mathrm{ti,flux}}$

$$F_{\mathrm{DR,sput}} = \frac{\Gamma_{\mathrm{DR}}}{\Gamma_0} = (1 - \alpha_t \beta_t)$$

$$F_{\mathrm{ti,flux}} = \frac{\Gamma_{\mathrm{DR,ions}}}{\Gamma_{\mathrm{DR,sput}}} = \frac{\Gamma_0 \alpha_t (1 - \beta_t)}{\Gamma_0 (1 - \alpha_t \beta_t)} = \frac{\alpha_t (1 - \beta_t)}{(1 - \alpha_t \beta_t)}$$

to the internal parameters back attraction probability β_t

$$\beta_t = \frac{1 - F_{\mathrm{DR,sput}}}{1 - F_{\mathrm{DR,sput}}(1 - F_{\mathrm{ti,flux}})}$$

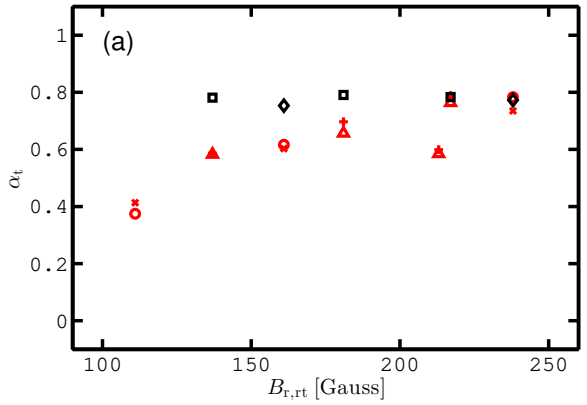
and ionization probability α_t

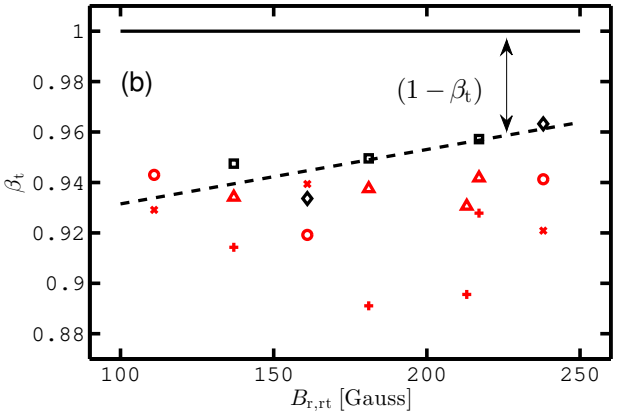
$$\alpha_t = 1 - F_{\mathrm{DR,sput}}(1 - F_{\mathrm{ti,flux}})$$

Hajihoseini et al. (2019) *Plasma* **2** 201 and later refined by Rudolph et al. (2021) JAP **129** 033303

Depostion rate – α_t and β_t

- In the fixed peak current mode (**black**) α_t is almost constant (range 0.75 – 0.79)
- In fixed voltage mode (**red**) α_t increases with increased $|\mathbf{B}|$
- In the fixed peak current mode (**black**) β_t increases slightly with increased $|\mathbf{B}|$ in the range 0.93 – 0.96
- If we assume a linear increase in β_t with $|\mathbf{B}|$ the fraction $(1 - \beta_t)$ is roughly 30% higher at the highest $|\mathbf{B}|$ than at the lowest $|\mathbf{B}|$
- In fixed voltage mode (**red**) β_t is rather scattered

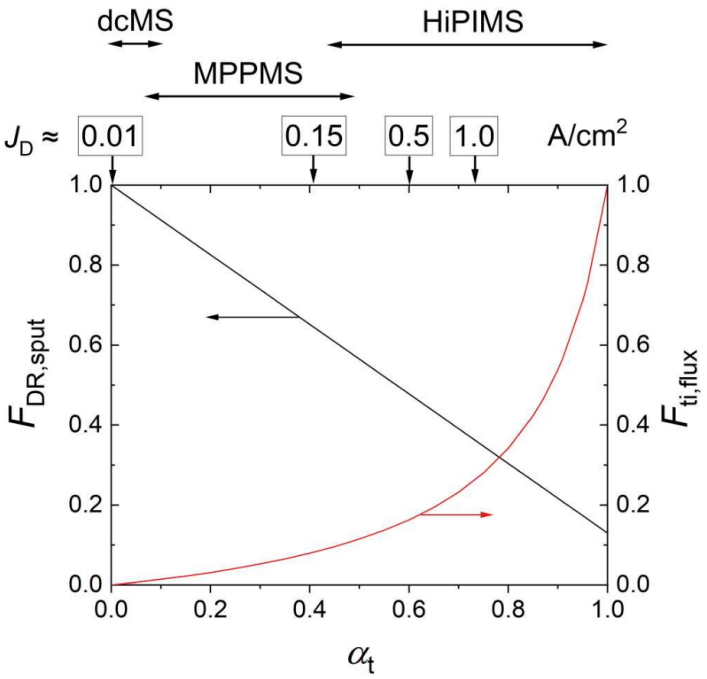




From Hajihoseini et al. (2019) *Plasma* **2** 201

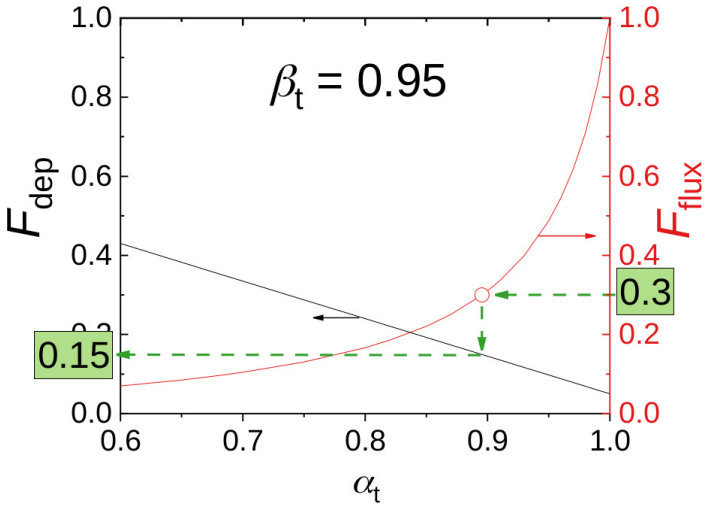
Deposition rate – α_t and β_t

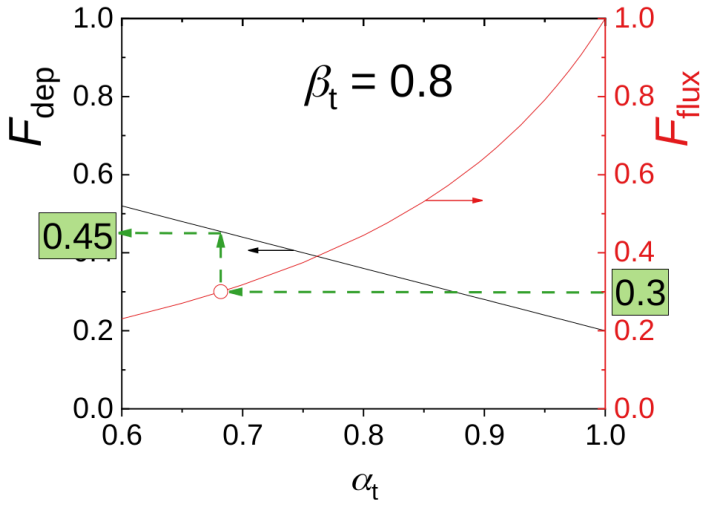
- There are two measures of how good a HiPIMS discharge is:
  - the fraction of all the sputtered material that reaches the diffusion region (DR) $F_{\mathrm{DR,sput}}$
  - the fraction of ionized species in that flux $F_{\mathrm{ti,flux}}$
- There is a trade off between the goals of higher $F_{\mathrm{DR,sput}}$ and higher $F_{\mathrm{ti,flux}}$
- The figure shows $F_{\mathrm{DR,sput}}$ and $F_{\mathrm{ti,flux}}$ as functions of α_t at assumed fixed value of $\beta_t = 0.87$



From Brenning et al. (2020) JVSTA **38** 033008

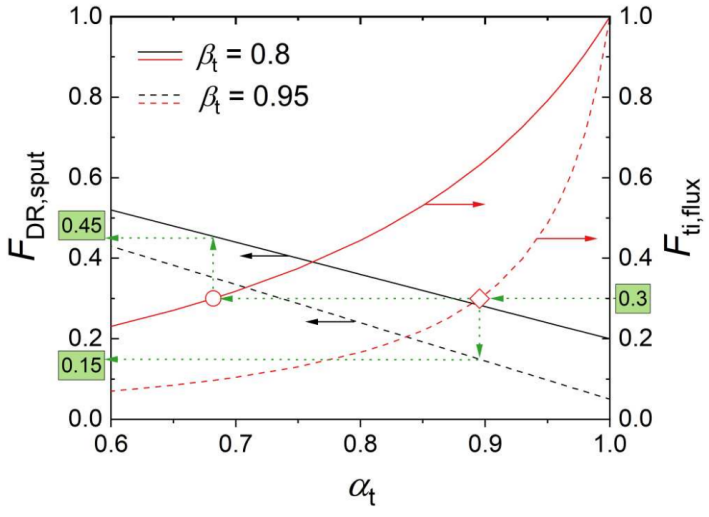
Deposition rate – α_t and β_t





- Lets say that ionized flux fraction of 30 % is desired
- For $\beta_t = 0.95$ following the green dashed line $F_{\mathrm{ti,flux}} = 0.30$ to the red curve gives $\alpha_t = 0.9$ and then $F_{\mathrm{DR,sput}} = 0.15$
- Reducing the back-attraction to $\beta_t = 0.8$ then $\alpha_t = 0.69$ is sufficient to maintain $F_{\mathrm{ti,flux}} = 0.30$ (red circle) and $F_{\mathrm{DR,sput}} = 0.45$ or a factor of three increase

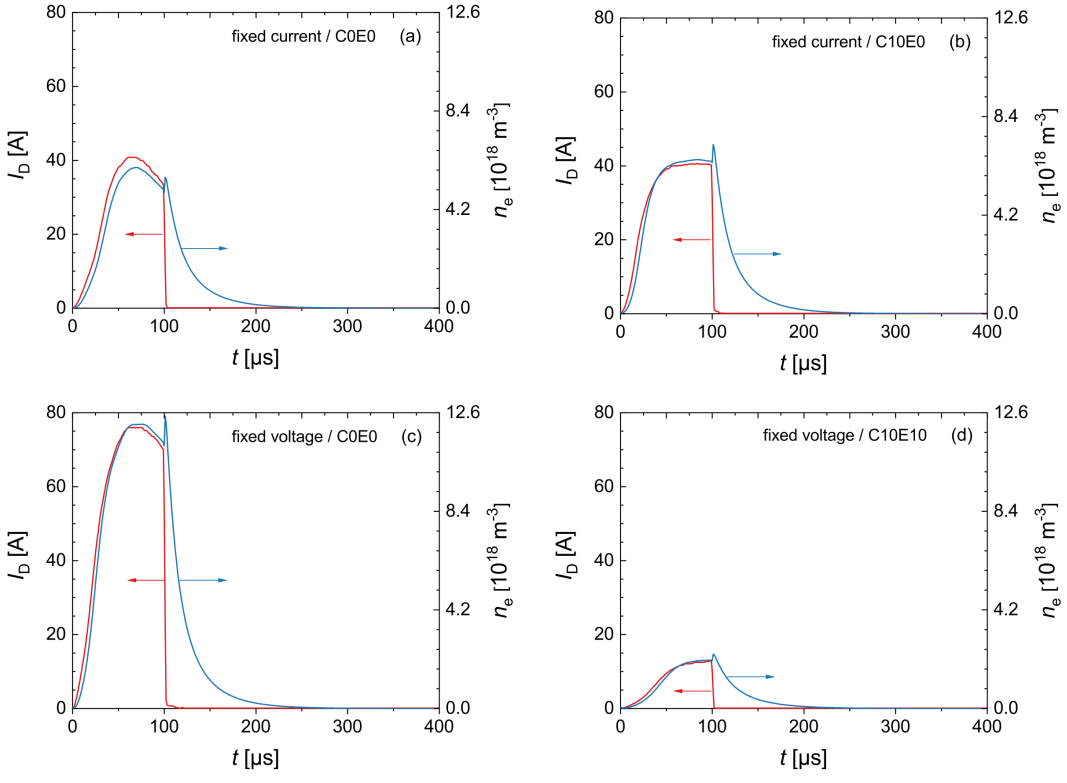
Deposition rate – α_t and β_t



From Brenning et al. (2020) JVSTA **38** 033008

- The question that remains:
  - How can we vary the ionization probability α_t and maybe more importantly the back-attraction probability β_t ?

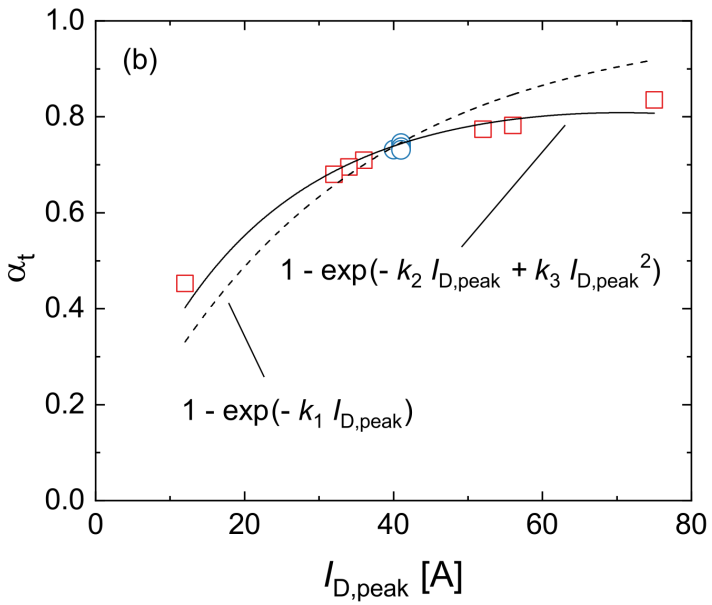
Deposition rate – α_t and β_t



From Rudolph et al. (2022) *J. Phys. D: Appl. Phys.* **55** 015202

- The electron density calculated by the IRM follows the discharge current waveform – despite varying magnetic field configurations – $I_D \propto n_e$ during the pulse

Deposition rate – α_t and β_t



- Electron impact ionization dominates ionization of the sputtered species and the ionization probability α_t scales with n_e
- The ionization probability α_t depends on the peak discharge current

$$\alpha_t(I_{D,peak}) = 1 - \exp(-k_1 I_{D,peak})$$

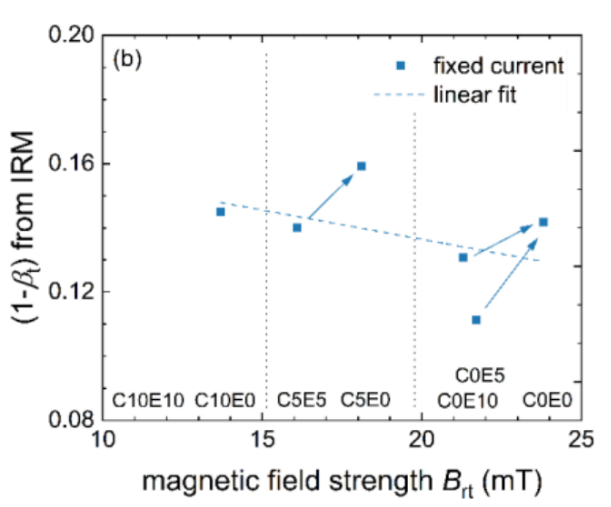
- Corrected for rarefaction

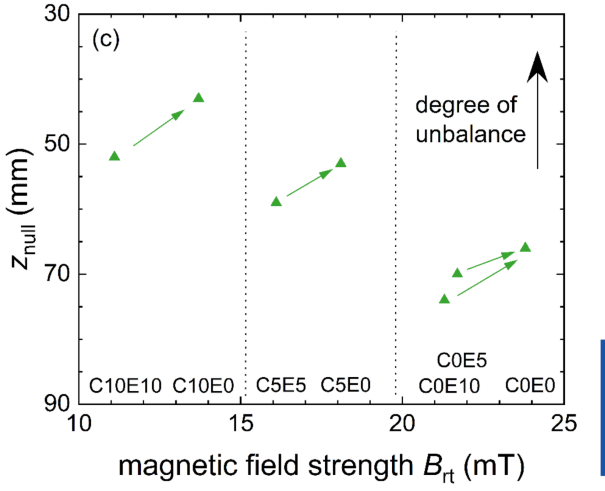
$$\alpha_t(I_{D,peak}) = 1 - \exp(-k_2 I_{D,peak} + k_3 I_{D,peak}^2)$$

From Rudolph et al. (2022) *J. Phys. D: Appl. Phys.* **55** 015202

Depostion rate – α_t and β_t

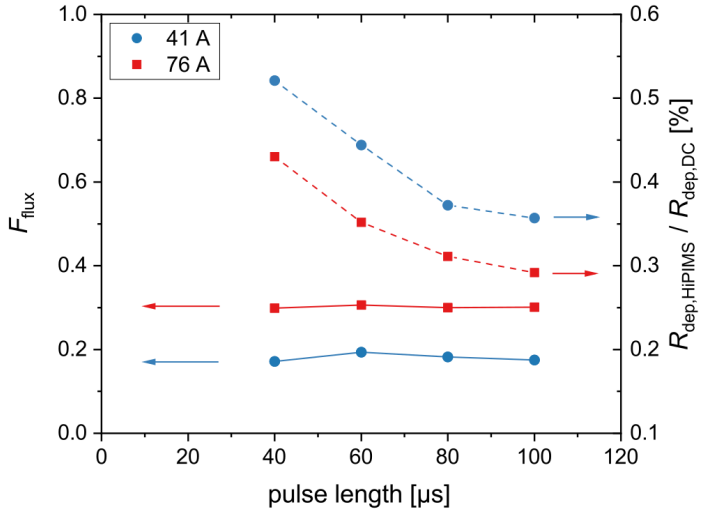
- The ion escape fraction $(1-\beta_t)$ versus the magnetic field strength
- Stronger B_{rt} leads to larger back attraction β_t
- How about magnetic unbalance ?
- The physical mechanism is still unclear
  - Stronger B_{rt} allows for larger potential drops V_{IR} over the IR
  - Does higher V_{IR} give higher back-attraction ?
  - What is the role of spokes ?

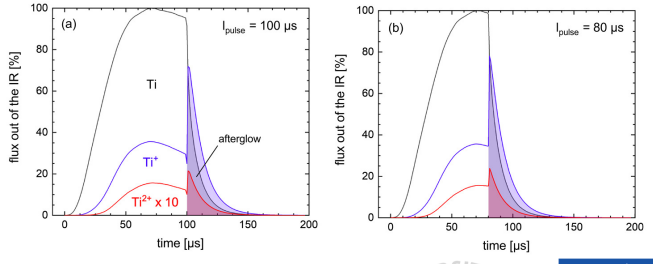




Deposition rate – Pulse length

- For the same average power, shorter pulses give higher deposition rate than longer pulses
- To maintain the same average power the repetition frequency is varied
- Shortening the pulses does not affect the ionized flux fraction, which remains essentially constant
  - with shorter pulses, the afterglow contributes increasingly more to the total deposition rate
  - the ionized flux fraction from the afterglow is typically higher compared to that during the pulse due to absent back-attracting electric field



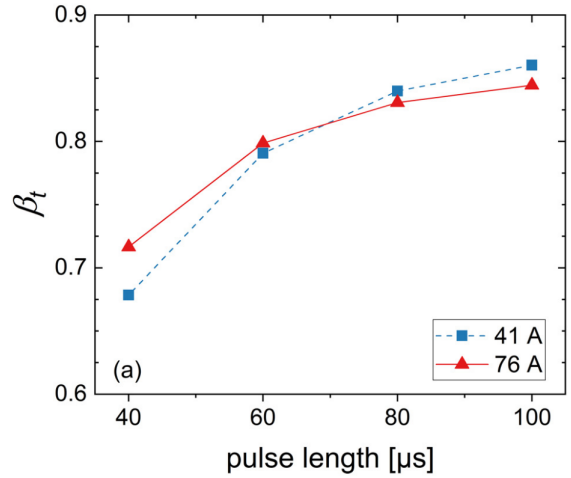


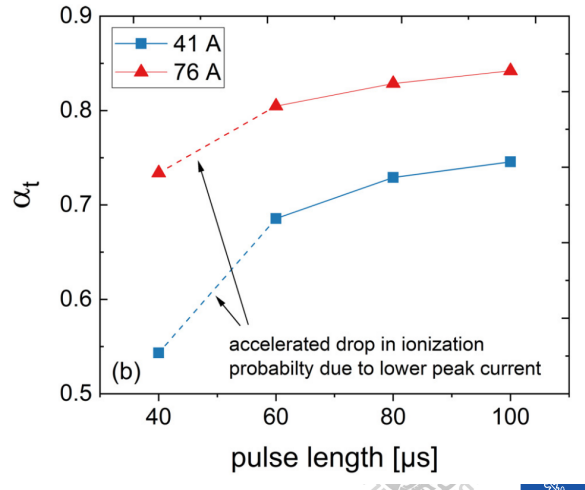
Rudolph et al. (2020) PSST **29** 05LT01

Deposition rate – Pulse length

- By switching-off the cathode potential during the afterglow decreases the effective β_t
- β_t decreases with decreasing pulse length
- The relative contribution of the afterglow ions to the flux toward the DR increases steadily for shorter pulses
- The ionization probability α_t also decreases with a shorter pulse length
- The useful fraction of the sputtered species therefore increases

$$F_{DR,sput} = \frac{\Gamma_{DR}}{\Gamma_0} = (1 - \alpha_t \beta_t)$$

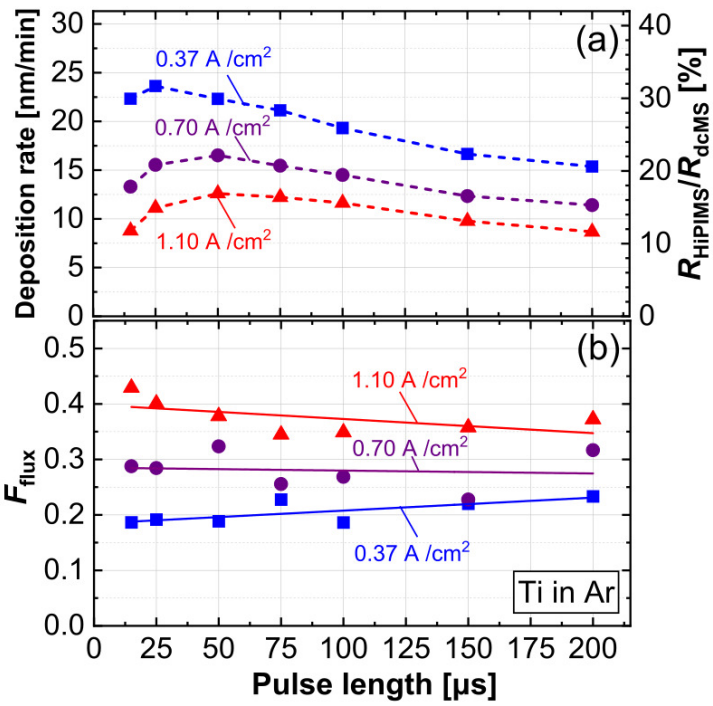




Brenning et al. (2020) JVSTA **38** 033008

Deposition rate – Pulse length

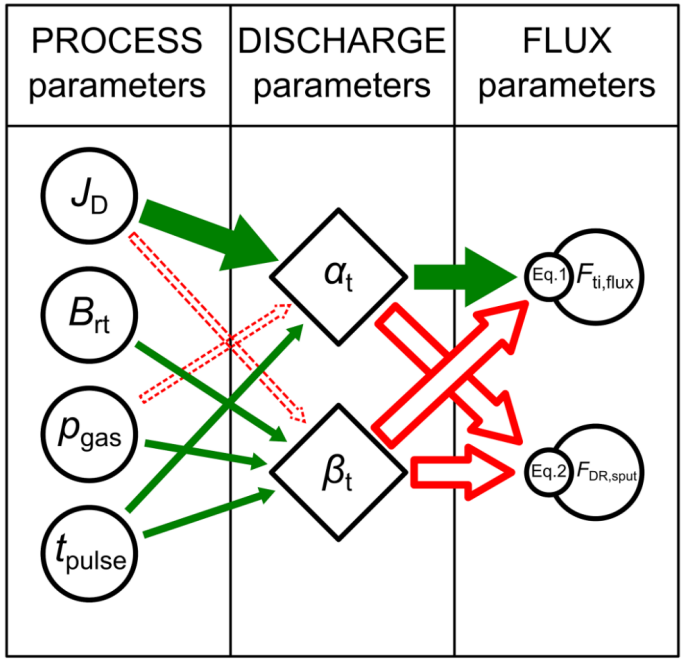
- These findings have been confirmed experimentally
- 6“ circular target with Ti target
- The pulse length is in the range of 15 – 200 μs, and the peak discharge current density $J_{\mathrm{D,peak}} = 0.37, 0.70, 1.10$ A/cm^2 ajusted the the discharge voltage
- The average sputtering power delivered to the target was kept at 1 kW by adjusting the pulse repetition frequency in the range 85 – 980 Hz



Shimizu et al. (2021) PSST **29** 045006

Influence of magnetic field – Pulse length

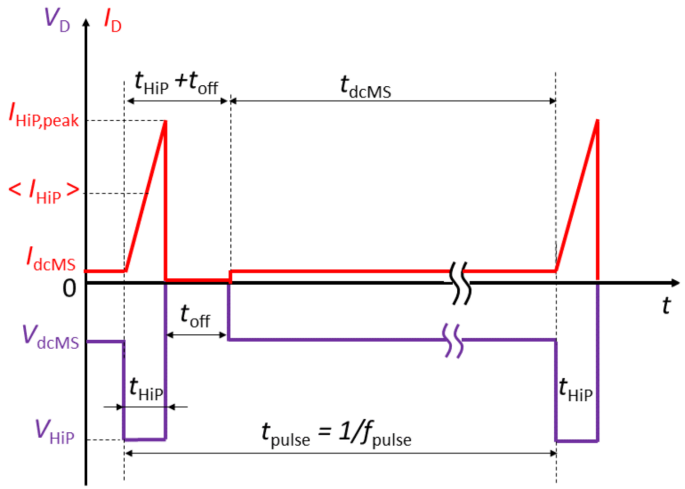
- HiPIMS can be optimized by selecting
  - pulse power
  - pulse length
  - working gas pressure
  - magnetic field strength
- The HiPIMS compromise – a fully ionized material flux is not required to achieve significant improvement of the thin film properties
- A sufficiently high peak discharge current is required to reach the desired ionized flux fraction
- Further increase would lead to unnecessarily low deposition rates



Brenning et al. (2020) JVSTA **38** 033008

Mixed high power and low power pulsing

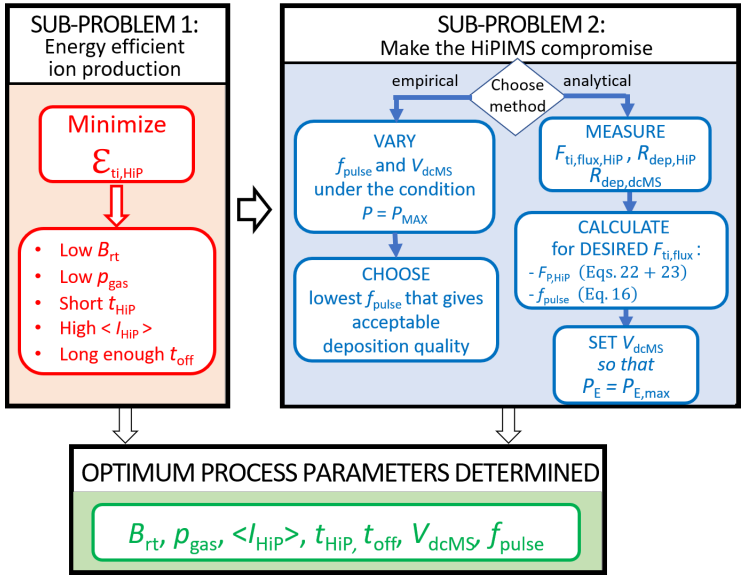
- The HiPIMS discharge can also be optimized by mixing two different power levels in the pulse pattern
  - Standard HiPIMS pulses create the ions of the film-forming material
  - An off-time follows, during which no voltage (or a reversed voltage) to let ions escape towards the substrate
  - Then long second pulse, in the dc magnetron sputtering range, is applied, to create neutrals of the film-forming material



Brenning et al. (2021) PSST **30** 015015

Mixed high power and low power pulsing

- The optimum power split is decided by the lowest ionized flux fraction that gives the desired film properties for a specific application
  - The low-power pulse is a much more efficient way of creating neutral atoms of the sputtered species
  - The high-power pulse should be applied to create mostly ions



Brenning et al. (2021) PSST **30** 015015

Summary

Summary

- There is an inescapable conflict between the goals of higher deposition rate and higher fraction of ionized species in the sputtered material flux
- The peak discharge current dictates the ionization probability of the sputtered species α_t
  - A sufficiently high peak discharge current is required to reach the desired ionized flux fraction
  - Further increase would lead to unnecessarily low deposition rates
- The HiPIMS discharge can be optimized by adjusting the pulse power, pulse length, working gas pressure and the magnetic field strength

Thank you for your attention

tumi@hi.is

The slides can be downloaded at
http://langmuir.raunvis.hi.is/~tumi/ranns.html
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References

Alami, J., P. O. A. Petersson, D. Music, J. T. Gudmundsson, J. Bohlmark, and U. Helmersson (2005). Ion-assisted physical vapor deposition for enhanced film deposition on non-flat surfaces. *Journal of Vacuum Science and Technology A 23*(2), 278–280.

Brenning, N., A. Butler, H. Hajihoseini, M. Rudolph, M. A. Raadu, J. T. Gudmundsson, T. Minea, and D. Lundin (2020). Optimization of HiPIMS discharges: The selection of pulse power, pulse length, gas pressure, and magnetic field strength. *Journal of Vacuum Science and Technology A 38*(3), 033008.

Brenning, N., H. Hajihoseini, M. Rudolph, M. A. Raadu, J. T. Gudmundsson, T. M. Minea, and D. Lundin (2021). HiPIMS optimization by using mixed high-power and low-power pulsing. *Plasma Sources Science and Technology 30*(1), 015015.

Christie, D. J. (2005). Target material pathways model for high power pulsed magnetron sputtering. *Journal of Vacuum Science and Technology A 23*(2), 330–335.

Gudmundsson, J. T., N. Brenning, D. Lundin, and U. Helmersson (2012). The high power impulse magnetron sputtering discharge. *Journal of Vacuum Science and Technology A 30*(3), 030801.

Gudmundsson, J. T. (2020). Physics and technology of magnetron sputtering discharges. *Plasma Sources Science and Technology 29*(11), 113001.

Gudmundsson, J. T. and D. Lundin (2020). Introduction to magnetron sputtering. In D. Lundin, T. Minea, and J. T. Gudmundsson (Eds.), *High Power Impulse Magnetron Sputtering: Fundamentals, Technologies, Challenges and Applications*, pp. 1–48. Amsterdam, The Netherlands: Elsevier.

Hajihoseini, H., M. Čada, Z. Hubička, S. Ünaldi, M. A. Raadu, N. Brenning, J. T. Gudmundsson, and D. Lundin (2019). The effect of magnetic field strength and geometry on the deposition rate and ionized flux fraction in the HiPIMS discharge. *Plasma 2*(2), 201–221.

Huo, C., D. Lundin, J. T. Gudmundsson, M. A. Raadu, J. W. Bradley, and N. Brenning (2017). Particle-balance models for pulsed sputtering magnetrons. *Journal of Physics D: Applied Physics 50*(35), 354003.

References

Kateb, M., H. Hajihoseini, J. T. Gudmundsson, and S. Ingvarsson (2019). Role of ionization fraction on the surface roughness, density, and interface mixing of the films deposited by thermal evaporation, dc magnetron sputtering, and HiPIMS: An atomistic simulation. *Journal of Vacuum Science and Technology A 37*(3), 031306.

Kubart, T., M. Čada, D. Lundin, and Z. Hubička (2014). Investigation of ionized metal flux fraction in HiPIMS discharges with Ti and Ni targets. *Surface and Coatings Technology 238*, 152–157.

Magnus, F., A. S. Ingason, S. Olafsson, and J. T. Gudmundsson (2012). Nucleation and resistivity of ultrathin TiN films grown by high power impulse magnetron sputtering. *IEEE Electron Device Letters 33*(7), 1045 – 1047.

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 Science and Technology 20*(6), 065007.

Rudolph, M., N. Brenning, H. Hajihoseini, M. A. Raadu, T. M. Minea, A. Anders, D. Lundin, and J. T. Gudmundsson (2022). Influence of the magnetic field on the discharge physics of a high power impulse magnetron sputtering discharge. *Journal of Physics D: Applied Physics 55*(1), 015202.

Rudolph, M., N. Brenning, M. A. Raadu, H. Hajihoseini, J. T. Gudmundsson, A. Anders, and D. Lundin (2020). Optimizing the deposition rate and ionized flux fraction by tuning the pulse length in high power impulse magnetron sputtering. *Plasma Sources Science and Technology 29*(5), 05LT01.

Rudolph, M., H. Hajihoseini, M. A. Raadu, J. T. Gudmundsson, N. Brenning, T. M. Minea, A. Anders, and D. Lundin (2021). On how to measure the probabilities of target atom ionization and target ion back-attraction in high-power impulse magnetron sputtering. *Journal of Applied Physics 129*(3), 033303.

Samuelsson, M., D. Lundin, J. Jensen, M. A. Raadu, J. T. Gudmundsson, and U. Helmersson (2010). On the film density using high power impulse magnetron sputtering. *Surface and Coatings Technology 202*(2), 591–596.

Shimizu, T., M. Zanáška, R. P. Villoan, N. Brenning, U. Helmersson, and D. Lundin (2021). Experimental verification of deposition rate increase, with maintained high ionized flux fraction, by shortening the HiPIMS pulse. *Plasma Sources Science and Technology 30*(4), 045006.