

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

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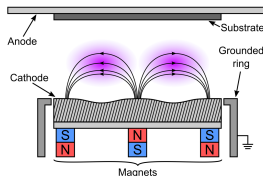
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Introduction – Magnetron sputtering

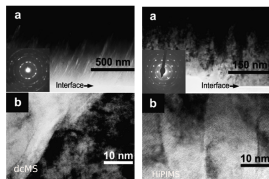
- Magnetron sputtering discharges are widely used in thin film deposition
 - It is a highly successful technique that is essential in a number of industrial applications



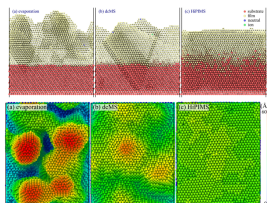
Gudmundsson and Lundin (2020) in High Power Impulse Magnetron Sputtering Discharge, Elsevier, 2020

- 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

Introduction – Fraction of ionization



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



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

- High power impulse magnetron sputtering (HiPIMS) provides higher ionized flux fraction than dc magnetron sputtering (dcMS)

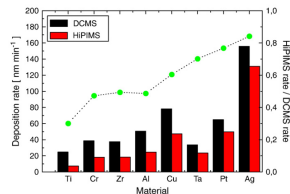
Gudmundsson (2020) PSST **29** 113001

- 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 – 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

Introduction – Fraction of ionization

- We distinguish between three approaches to describe the degree (or fraction) of ionization
 - the ionized flux fraction

$$F_{\text{flux}} = \frac{\Gamma_i}{\Gamma_i + \Gamma_n}$$

- the ionized density fraction

$$F_{\text{density}} = \frac{n_i}{n_i + n_n}$$

- the fraction α_t of the sputtered metal atoms that become ionized in the discharge (probability of ionization)



HiPIMS discharge with titanium target



HiPIMS discharge with titanium target

- A primary current I_{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

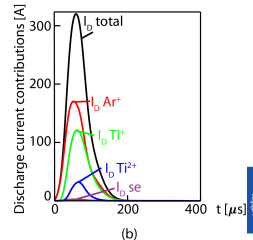
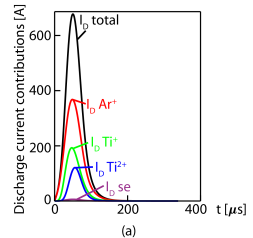
$$I_{\text{crit}} = S_{\text{RT}} e n_g \sqrt{\frac{1}{2\pi m_g k_B T_g}} = S_{\text{RT}} e n_g \sqrt{\frac{k_B T_g}{2\pi m_g}}$$

- Discharge currents I_D above I_{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

HiPIMS discharge with titanium target

- For discharges with Ti target the peak current is far above the critical current (up to 650 A, while $I_{\text{crit}} \approx 19$ A)
- However, this discharge shows close to a 50/50 combination of **self-sputter recycling** $I_{\text{SS-recycle}}$ and **working gas-recycling** $I_{\text{gas-recycle}}$
- 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



Influence of magnetic field

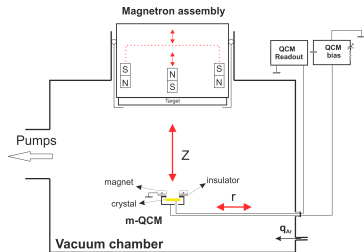


Influence of magnetic field – Deposition rate

- The Ti deposition rate and the ionized flux fraction are measured using a gridless ion meter (m-QCM)

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

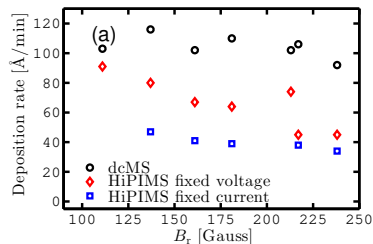
- The ion meter is mounted on a probe holder which can be moved around within the chamber
- The Ar working gas pressure was set to 1 Pa
- In all cases the pulse width was $100\ \mu\text{s}$ at an average power of 300 W



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

Influence of magnetic field – Deposition rate

- The Ti deposition rate recorded at substrate position using a gridless ion meter (m-QCM)
 - **dcMS**
+10% with decreasing $|\mathbf{B}|$
(but no obvious trend)
 - **HiPIMS fixed voltage**
+110% with decreasing $|\mathbf{B}|$
 - **HiPIMS fixed peak current**
+40% with decreasing $|\mathbf{B}|$
- In HiPIMS operation the deposition rate increases with decreasing $|\mathbf{B}|$

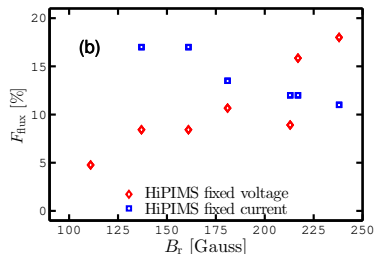


From Gudmundsson (2020) *PSST* **29**(11) 113001

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

Influence of magnetic field – 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



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Internal parameters and optimization



Influence of magnetic field – α_t and β_t

- Low deposition rate is the main drawback of this sputter technology and hampers its use for industrial applications
- The main reason for the low deposition rate of the HiPIMS discharge is suggested to be due to the back-attraction of the ions of the sputtered species to the cathode target
- Increased deposition rate in HiPIMS often comes at the cost of a lower ionized flux fraction of the sputtered material
- Two internal parameters are of importance
 - α_t – ionization probability
 - β_t – back-attraction probability



Influence of magnetic field – α_t and β_t

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

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

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

to the internal parameters back attraction probability β_t

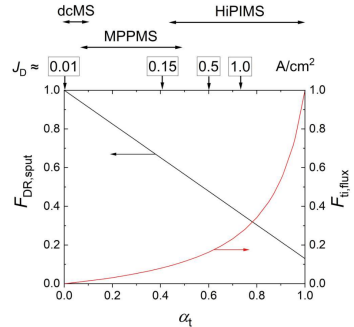
$$\beta_t = \frac{1 - F_{\text{DR,sput}}}{1 - F_{\text{DR,sput}}(1 - F_{\text{ti,flux}})}$$

and ionization probability α_t

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

Influence of magnetic field – Optimization

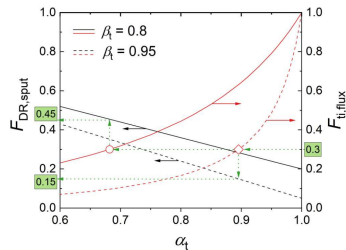
- There are two measures of how good a HiPIMS discharge is:
 - the fraction $F_{\text{DR,sput}}$ of all the sputtered material that reaches the diffusion region (DR)
 - the fraction $F_{\text{ti,flux}}$ of ionized species in that flux
- There is a trade off between the goals of higher $F_{\text{DR,sput}}$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



From Brenning et al. (2020) JVSTA 38 033001

Influence of magnetic field – Optimization

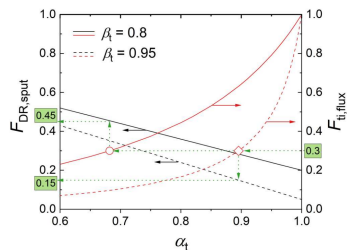
- For a particular application an ionized flux fraction of 30 % is suitable but $0.8 \leq \beta_t \leq 0.95$
- For $\beta_t = 0.95$ following the green dotted line from the value $F_{ti,flux} = 0.30$ to the red dashed curve gives $\alpha_t = 0.9$ (red square)
- The black dashed line then shows α_t only 15 % of the total sputtered flux enters the diffusion region ($F_{DR,sput} = 0.15$)



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

Influence of magnetic field – Optimization

- If the back-attraction can be reduced to $\beta_t = 0.8$ the deposition rate is increased
- The solid lines show that reducing the back-attraction to $\beta_t = 0.8$ where $\alpha_t = 0.69$ is sufficient to maintain $F_{ti,flux} = 0.30$ (red circle) $F_{DR,sput} = 0.45$ or a factor of three increase in the deposition rate
- The question that remains:
 - How can we vary the ionization probability α_t and maybe more importantly the back-attraction probability β_t ?

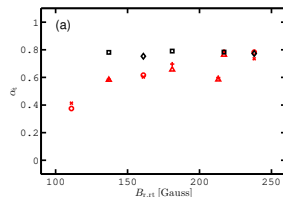


From Brenning et al. (2020) *JVSTA* **38** 033008



Influence of magnetic field – α_t and β_t

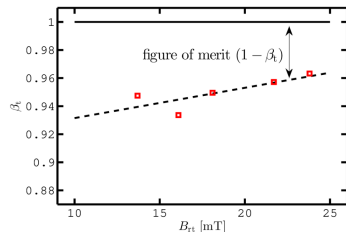
- When operating in the fixed peak current mode (**black**) the ionization probability α_t is roughly constant independent of the magnetic field strength
- When operating in the fixed voltage mode (**red**) the ionization probability α_t increases with increased magnetic field strength – which is essentially due to the increased discharge current
- α_t can be varied in the range $0 \leq \alpha_t \leq 1$ by the discharge current amplitude J_D



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

Influence of magnetic field – Optimization

- The figure shows β_t as a function of the magnetic field strength (measured 11 mm above the racetrack center) for a fixed peak discharge current
- There is a clear trend that β_t is lowered when the magnetic field strength is reduced
- Our proposed figure of merit $(1 - \beta_t)$ changes by a factor of $(1 - 0.93)/(1 - 0.96) = 1.8$



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

and/or Hajihoseini et al. (2019) *Plasma* **2** 201



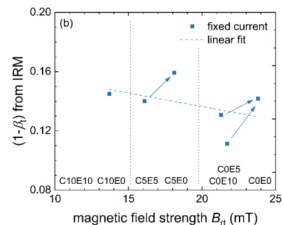
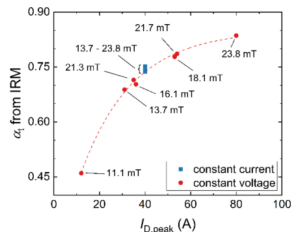
Influence of magnetic field – α_t and β_t

- The internal discharge parameters α_t and β_t from the ionization region model (IRM)

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

- The ionization probability α_t versus the discharge current
- The ion escape fraction $(1 - \beta_t)$ versus the magnetic field strength

From Rudolph et al. (2021a) manuscript in preparation

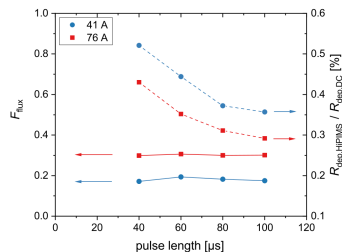


Influence of pulse length



Influence of pulse length

- For the same average power, shorter pulses give higher deposition rate than longer pulses
- To maintain the same average power the 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



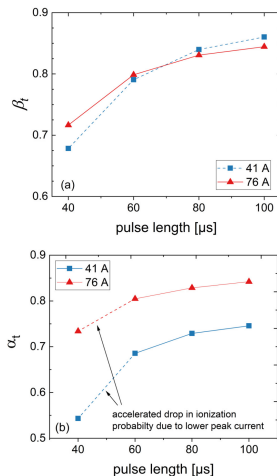
From Rudolph et al. (2020) *PSST* 29 05LT01



Influence of 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

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

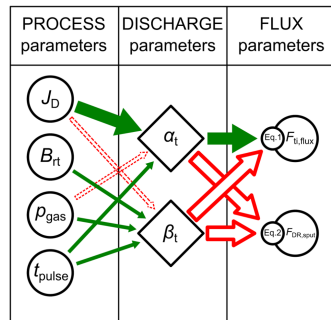


HiPIMS discharge optimization



HiPIMS discharge optimization

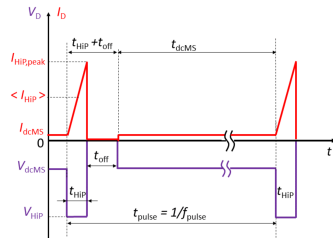
- HiPIMS can be optimized by selecting
 - pulse power
 - pulse length
 - working gas pressure
 - magnetic field strength
- The HiPIMS compromise – a fully ionized material flux is not required to achieve significant improvement of the thin film properties
- A sufficiently high peak discharge current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



From 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

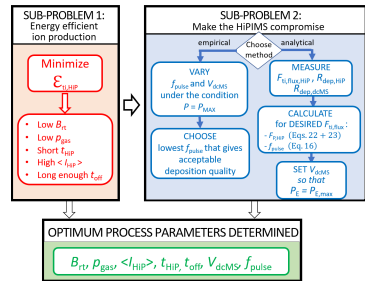


Brenning et al. (2021) *PSST* **30** 015015



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

- With varying magnetic field:
 - For HiPIMS in the fixed voltage mode: A trade-off between the deposition rate (increases by more than a factor of two) and the ionized flux fraction (decreases by a factor 4 to 5) with decreasing $|\mathbf{B}|$
 - For HiPIMS in the fixed peak current mode: Decreasing $|\mathbf{B}|$ improves both the deposition rate (by 40%) and the ionized flux fraction (by 50%)
- There is an inescapable conflict between the goals of higher deposition rate and higher fraction of ionized species in the sputtered material flux
- 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

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.
- 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. *Journal of Physics D: Applied Physics* 45(1), 012003.
- 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.
- Butler, A., N. Brenning, M. A. Raadu, J. T. Gudmundsson, T. Minea, and D. Lundin (2018). On three different ways to quantify the degree of ionization in sputtering magnetrons. *Plasma Sources Science and Technology* 27(10), 105005.
- 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. Ůnaldi, 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.



References

- 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.
- 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 Science and Technology* 23(2), 025017.
- 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.
- 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.

