Experiments and modelling of high power impulse magnetron sputtering discharges with metallic target

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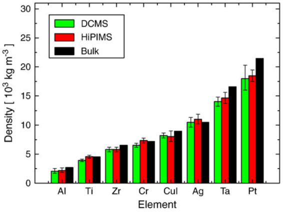
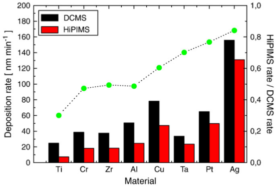
Introduction – Magnetron sputtering

- In a high power impulse magnetron sputtering (HiPIMS) the discharge is driven by high power pulses of low repetition frequency, and with low duty cycle
- The film mass density is higher, the films exhibit better crystallinity, and overall improved film properties, when deposited with HiPIMS
- There is a drawback: The deposition rate is lower for HiPIMS when compared to dcMS operated at the same average power
- Many of the ions of the target material are attracted back to the target surface by the cathode potential





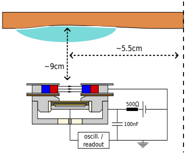
From Samuelsson et al. (2010) SCT **202** 591

Overview

- Ionized flux fraction – measurements
- The ionization region model (IRM)
- Deposition rate vs ionized flux fraction
- Working gas rarefaction
- Summary

Ionized flux fraction – measurements

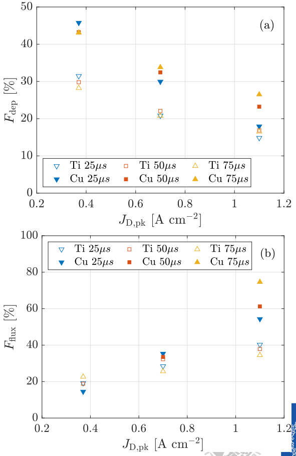
Ionized flux fraction – measurements



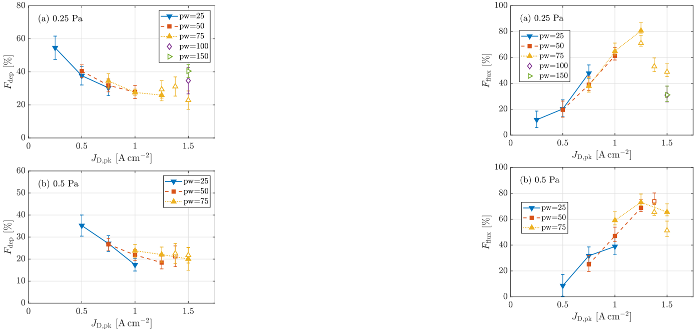
- The ionized flux fraction and the deposition rate fraction – measured by an ion meter in HiPIMS discharges with Cu and Ti targets and working gas pressure of 0.3 Pa

Cu: Fischer et al. (2023) PSST **32** 125006

Ti: Shimizu et al. (2021) PSST **30** 045006



Ionized flux fraction – measurements



From Fischer et al. (2023) PSST **32** 125006

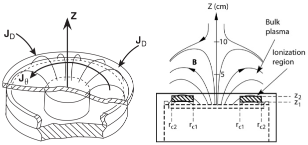
- The measured normalized deposition rate (left) and ionized flux fraction (right) as a function of the peak discharge current density $J_{D,peak}$ for working gas pressure of (a) 0.25 Pa and (b) 0.5 Pa

The ionization region model (IRM)

Ionization region model

- The ionization region model (IRM) is a time-dependent volume averaged plasma chemical model of the ionization region (IR) of the HiPIMS discharge
- The IRM gives the temporal evolution of the densities of ions, neutrals and electrons
- The IRM gives also two internal parameters that are of importance
  - α_t – ionization probability
  - β_t – back-attraction probability

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



The definition of the volume covered by the IRM

- The IR is defined as an annular cylinder of width $w_{rt} = r_{c2} - r_{c1}$ and thickness $L = z_2 - z_1$, extends from z_1 to z_2 axially away from the target

From Raadu et al. (2011) PSST **20** 065007

Ionization region model

- 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 (except electrons)
- The species assumed in the non-reactive-IRM are
  - cold electrons e^{C}, hot electrons e^{H}
  - 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), doubly ionized argon ions Ar^{2+} (27.63 eV)
  - Metal atoms, sometimes metastable states, metal ion M^{+}, and doubly ionized metal ions M^{2+}

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

Ionization region model

- As an example the particle balance equation for the metal ion M^+ is

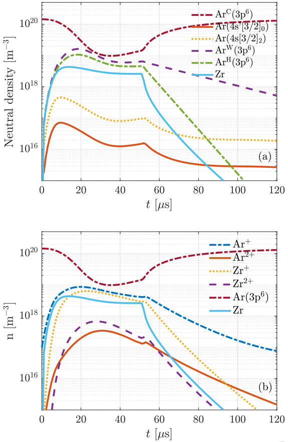
$$\frac{dn_{M^+}}{dt} = \underbrace{k_{iz,M}^{c} n_{e,c} n_{M} + k_{iz,M}^{h} n_{e,h} n_{M}}_{\text{electron impact ionization}} + \underbrace{k_{P,iz} n_{Ar^m} n_{M}}_{\text{Penning ionization}}$$

$$+ \underbrace{k_{chexc,1} n_{M} n_{Ar^+} + k_{chexc,2} n_{M^{2+}} n_{Ar}}_{\text{charge exchange}} - \underbrace{k_{iz,M^+}^{c} n_{e,c} n_{M^+} - k_{iz,M^+}^{h} n_{e,h} n_{M^+}}_{\text{electron impact ionization to create } M^{2+}}$$

$$- \underbrace{\frac{\Gamma_{M^+}^{RT} + \Gamma_{M^+}^{BP}(S_{IR} - S_{RT})}{V_{IR}}}_{\text{ion flux out of the ionization region}}$$

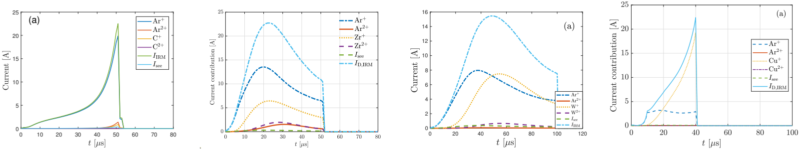
Ionization region model studies of HiPIMS

- The temporal evolution of the neutral and ion densities in a discharge with zirconium target
- Ar^{+} ions dominate the discharge – but Zr^{+} ions are not far off
- Ar^{2+} and Zr^{2+} ions have much lower densities
- Working gas rarefaction is very apparent



From Suresh Babu et al. (2024) JVSTA **42** 043007

Ionization region model



C: PSST (2021) **30** 115017 Zr: JVSTA (2024) **42** 043007 W: PSST (2022) **31** 065009 Cu: SCT (2022) **442** 128189

- The temporal evolution of the discharge current composition at the target surface for four different targets
- With Cu target Cu^+ ions dominate, with graphite target Ar^+ ions dominate
- For Zr and W targets there is a mix of Ar^+ and metal ions

Deposition rate vs ionized flux fraction

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

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

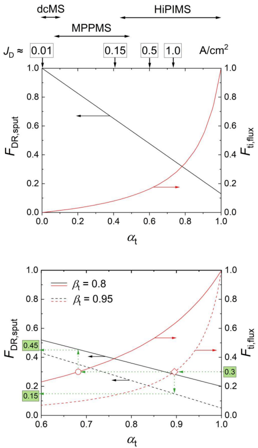
and ionization probability α_t

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

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

Deposition rate – Optimization

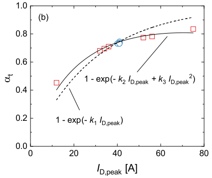
- There are two measures of how good a HiPIMS discharge is:
  - the fraction $F_{\mathrm{DR,sput}}$ of all the sputtered material that reaches the diffusion region (DR)
  - the fraction $F_{\mathrm{ti,flux}}$ of ionized species in that flux
- There is a trade off between the goals of higher $F_{\mathrm{DR,sput}}$ and higher $F_{\mathrm{ti,flux}}$
- The question that remains:
  - How can we vary the ionization probability α_{t} and maybe more importantly the back-attraction probability β_{t} ?



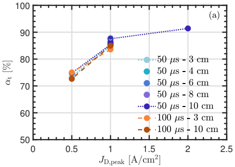
From Brenning et al. (2020) JVSTA **38** 033008

Deposition rate vs ionized flux fraction – α_t and β_t

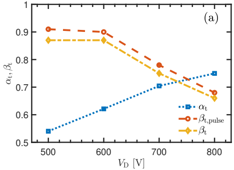
- The internal discharge parameters α_t and β_t from the ionization region model (IRM)



Ti: JPD **55** 015202



Zr: JVSTA **42** 043007

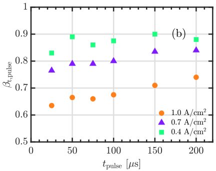


W: PSST **31** 065009

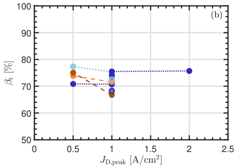
- The ionization probability α_t increases with increased peak increased discharge current density
- The peak discharge current increases with increased discharge voltage

Deposition rate vs ionized flux fraction – α_t and β_t

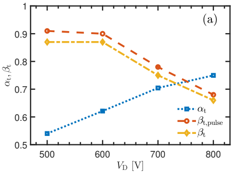
- The internal discharge parameters α_t and β_t from the ionization region model (IRM)



Cr: unpublished



Zr: JVSTA **42** 043007

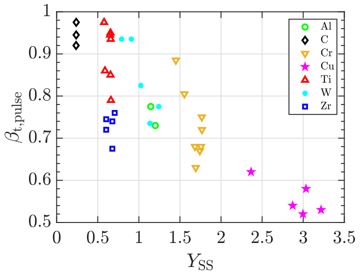


W: PSST **31** 065009

- The back-attraction probability β_t has less clear dependence on the peak discharge current density – decreases with increased peak discharge current density for Cr and W – no clear trend for Zr

Deposition rate vs ionized flux fraction – α_t *and* β_t

- What determines the back-attraction probability ?
- How can one influence the back-attraction probability ?
- The back-attraction probability $\beta_{t,pulse}$, determined by IRM, versus the self-sputter yield for various target materials
- The data indicate that the back-attraction probability decreases roughly linearly with increased self-sputter yield

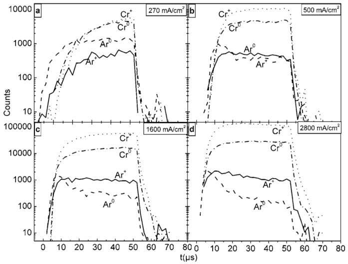


From Barynova et al. (2025) PSST **34** 06LT01

Working gas rarefaction

Working gas rarefaction

- The sputtered species enter the discharge at considerable energy, which is determined by the cohesive energy of the solid target
- The interaction between the energetic sputtered particles and the working gas atoms can lead to a reduction in the working gas density – as has been observed experimentally in the HiPIMS discharge
- Working gas rarefaction has been observed in the HiPIMS discharge



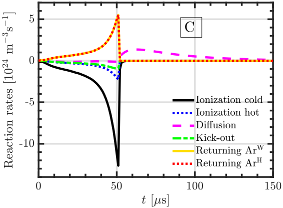
From Alami et al. (2006) APL **89**(15) 154104

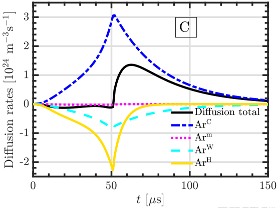
Working gas rarefaction

- HiPIMS discharge with graphite target and $J_{\mathrm{D,peak}} = 1$ A cm^{-2}

  Eliasson et al. (2021) PSST **30** 115017

- Argon atoms are lost mainly through electron impact ionization by primary and secondary electrons
- Contributions of kick-out and charge-exchange are negligible
- Diffusion contributes to a net loss of argon atoms during the pulse, but to a flow into the ionization region after the pulse is off





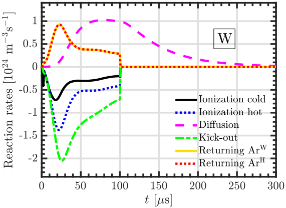
From Barynova et al. PSST **33**(6) 065010

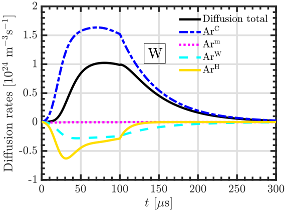
Working gas rarefaction

- HiPIMS discharge with tungsten target and $J_{\mathrm{D,peak}} = 0.54$ A cm^{-3}

  Suresh Babu et al. (2022) PSST **31** 065009

- The main contributor to the loss of argon atoms from the IR is kick-out by tungsten atoms sputtered from the target (39 – 48 % contribution)
- The second most important loss process is electron impact ionization by secondary electrons followed by electron impact ionization by the primary electrons

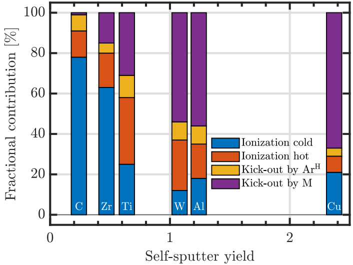




From Barynova et al. PSST **33**(6) 065010

Working gas rarefaction

- The relative contributions of the various processes to working gas rarefaction varies greatly depending on the target material
- The various contributions versus the atomic mass of the target material for $J_{\mathrm{D,peak}} \sim 1$ A/cm^2 and $p_{\mathrm{g}} \sim 1$ Pa



From Barynova et al. PSST **33**(6) 065010

Summary

Summary

- The discharge current composition at the target surface depends on the target material
- There is an inescapable conflict between the goals of higher deposition rate and higher fraction of ionized species in the sputtered material flux
- The back-attraction probability appears to depend on the self-sputter yield – it is lower for higher self-sputter yield
- The main contributor to working gas rarefaction for low sputter yield target is electron impact ionization, while for targets with high sputter yield kick-out by the sputtered species is the main contributor

Thank you for your attention

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The slides can be downloaded at

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

Further reading

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


Topical Review

Physics and technology of magnetron sputtering discharges

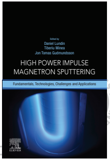
J T Gudmundsson

- J. T. Gudmundsson, André Anders, and Achim von Keudell, Foundations of physical vapor deposition with plasma assistance, Plasma Sources Science and Technology, **31**(8) (2022) 083001


Topical Review

Foundations of physical vapor deposition with plasma assistance

- Daniel Lundin, Tiberiu Minea and Jon Tomas Gudmundsson (eds.), High Power Impulse Magnetron Sputtering: Fundamentals, Technologies, Challenges and Applications, Elsevier, 2020



References

Alami, J., K. Sarakinos, G. Mark, and M. Wuttig (2006). On the deposition rate in a high power pulsed magnetron sputtering discharge. *Applied Physics Letters 89*(15), 154104.

Barynova, K., N. Brenning, S. Suresh Babu, J. Fischer, D. Lundin, M. A. Raadu, J. T. Gudmundsson, and M. Rudolph (2025). Self-regulating electron temperature in high-power impulse magnetron sputtering discharges and its effect on the metal ion escape. *Plasma Sources Science and Technology 34*(6), 06LT01.

Barynova, K., S. Suresh Babu, M. Rudolph, J. Fischer, D. Lundin, M. A. Raadu, N. Brenning, and J. T. Gudmundsson (2024). On working gas rarefaction in high power impulse magnetron sputtering. *Plasma Sources Science and Technology 33*(6), 065010.

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., 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 Science and Technology 26*(12), 125003.

Eliasson, H., M. Rudolph, N. Brenning, H. Hajihoseini, M. Zanáška, M. J. Adriaans, M. A. Raadu, T. M. Minea, J. T. Gudmundsson, and D. Lundin (2021). Modeling of high power impulse magnetron sputtering discharges with graphite target. *Plasma Sources Science and Technology 30*(11), 115017.

Fischer, J., M. Renner, J. T. Gudmundsson, M. Rudolph, H. Hajihoseini, N. Brenning, and D. Lundin (2023). Insights into the copper HiPIMS discharge: Deposition rate and ionised flux fraction. *Plasma Sources Science and Technology 32*(12), 125006.

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

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. 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.

References

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.

Held, J., V. Schulz-von der Gathen, and A. von Keudell (2023). Ionization of sputtered material in high power impulse magnetron sputtering plasmas – comparison of titanium, chromium and aluminum. *Plasma Sources Science and Technology 32*(6), 065006.

Helmersson, U., M. Lattemann, J. Alami, J. Bohlmark, A. P. Ehiasarian, and J. T. Gudmundsson (2005). High power impulse magnetron sputtering discharges and thin film growth: A brief review. In *Proceedings of the 48th Society of Vacuum Coaters Annual Technical Conferenece*, pp. 458 – 464.

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.

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., 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.

References

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.

Suresh Babu, S., M. Rudolph, D. Lundin, T. Shimizu, J. Fischer, M. A. Raadu, N. Brenning, and J. T. Gudmundsson (2022). Ionization region model of a high power impulse magnetron sputtering of tungsten. *Plasma Sources Science and Technology 31*(6), 065009.

Suresh Babu, S., J. Fischer, M. Rudolph, D. Lundin, and J. T. Gudmundsson (2024). Modeling of high power impulse magnetron sputtering discharges with a zirconium target. *Journal of Vacuum Science and Technology A 42*(4), 043007.