On the connection between the self-sputter yield and deposition rate in HiPIMS operation

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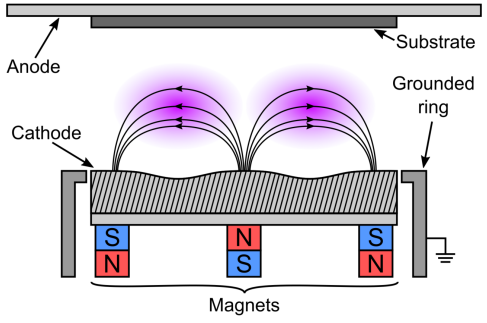
Introduction – Magnetron sputtering



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

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

  Gudmundsson (2020) PSST **29** 113001
- 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
- This results in high discharge current density, increased electron density, and increased ionization of the sputtered species

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

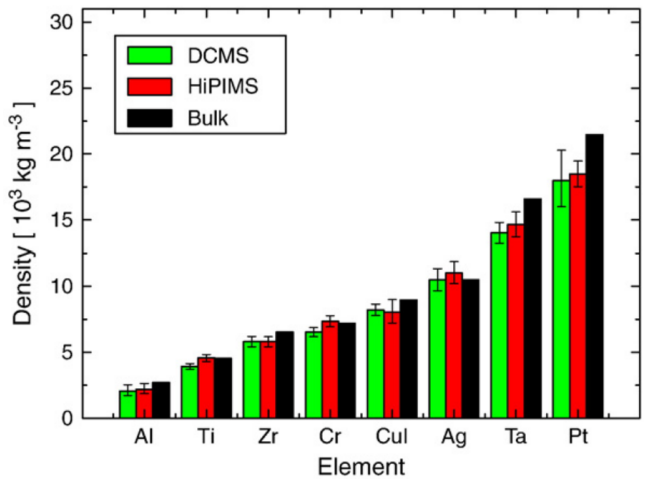
Overview

- Thin film deposition
- The ionization region model (IRM)
- Deposition rate vs ionized flux fraction
- Working gas rarefaction
- Summary

Thin film deposition

Thin film deposition

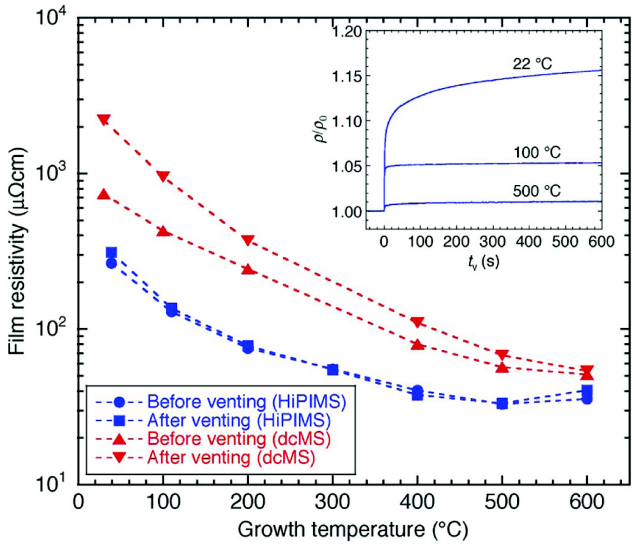
- The film mass density is always higher
- The surfaces are significantly smoother – when depositing with HiPIMS compared to dcMS at the same average power
- The films typically exhibit better crystallinity, and overall improved film properties
  - lower electrical resistivity
  - improved optical properties
  - improved mechanical properties
  - better oxidation resistance



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

Thin film deposition

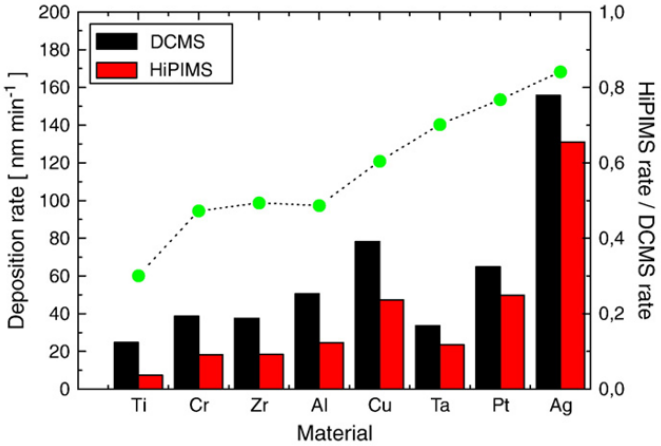
- TiN as diffusion barriers for interconnects
- HiPIMS deposited films have significantly lower resistivity than dcMS deposited films on SiO_2 at all growth temperatures due to reduced grain boundary scattering
- Thus, ultrathin continuous TiN films with superior electrical characteristics and high resistance towards oxidation can be obtained with HiPIMS at reduced temperatures



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

Thin film deposition

- 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



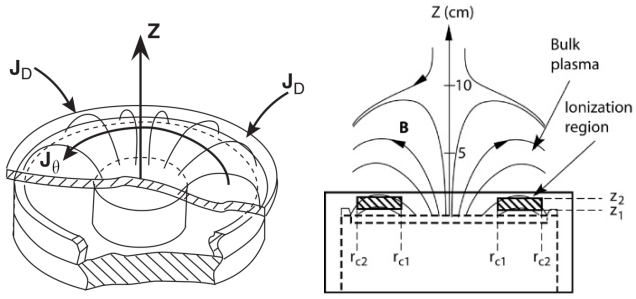
From Samuelsson et al. (2010) SCT **202** 591

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

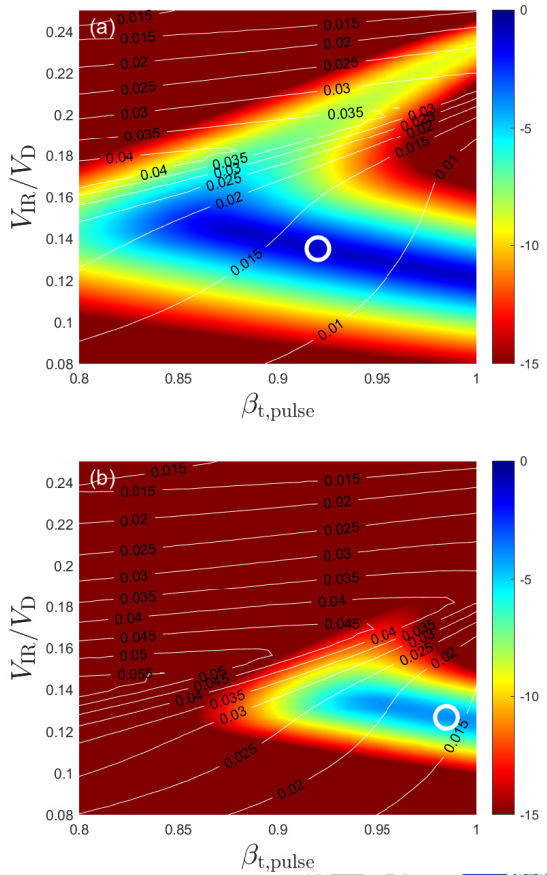
$$\frac{\mathrm{d}n_{\mathrm{M}^+}}{\mathrm{d}t} = \underbrace{k^{\mathrm{c}}_{\mathrm{iz,M}} n_{\mathrm{e}} n_{\mathrm{M}} + k^{\mathrm{h}}_{\mathrm{iz,M}} n_{\mathrm{e}} n_{\mathrm{M}}}_{\text{electron impact ionization}} + \underbrace{k_{\mathrm{P,iz}} n_{\mathrm{Ar^m}} n_{\mathrm{M}}}_{\text{Penning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

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

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

Ionization region model

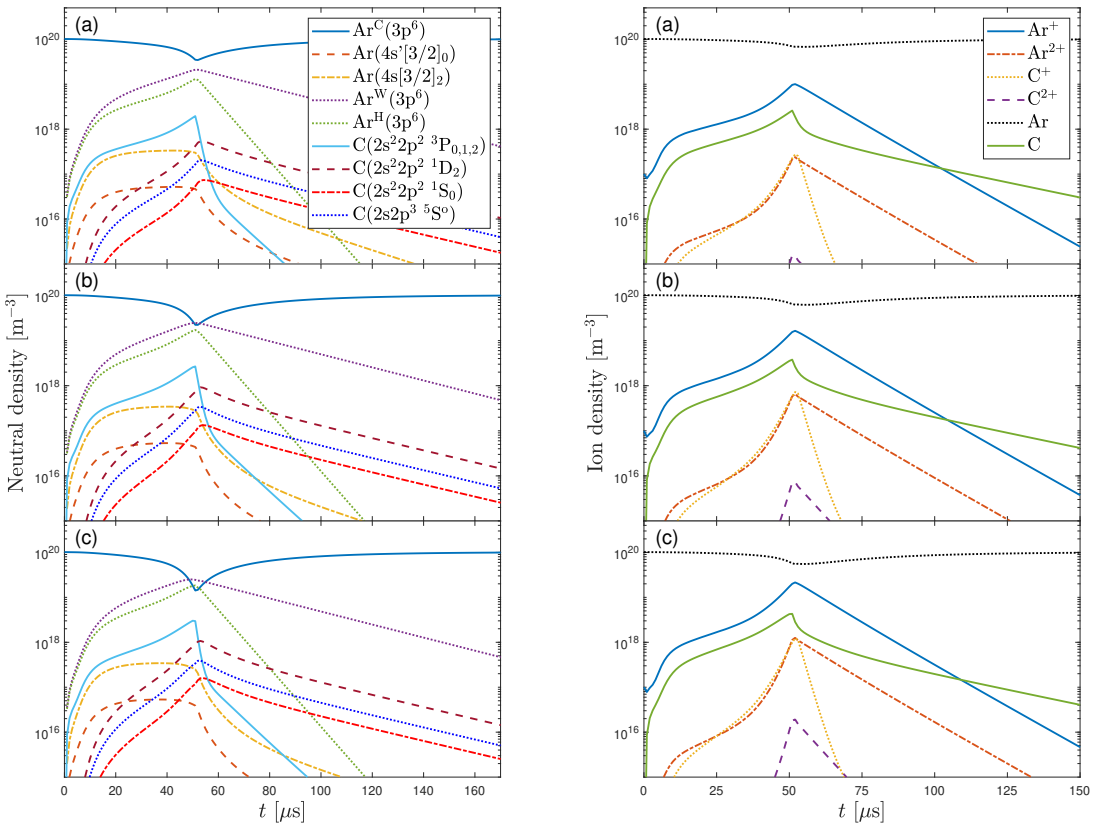
- The IRM is a semi-empirical discharge model and requires the measured discharge current and voltage waveforms
- The IRM has three unknown fitting parameters
  - the ion back-attraction probability for the metal ions $\beta_{\mathrm{t,pulse}}$ and gas ions $\beta_{\mathrm{g,pulse}}$
  - the potential drop across the IR $f = V_{\mathrm{IR}}/V_{\mathrm{D}}$
  - the electron recapture probability $r = 0.7$
- This leaves the ($\beta_{\mathrm{t,pulse}}$, f) parameter space to be explored through the model fitting procedure – the blue zones in the fitting map indicate the smallest mean square error



Graphite target from Eliasson et al. (2021) PSST **29** 115017

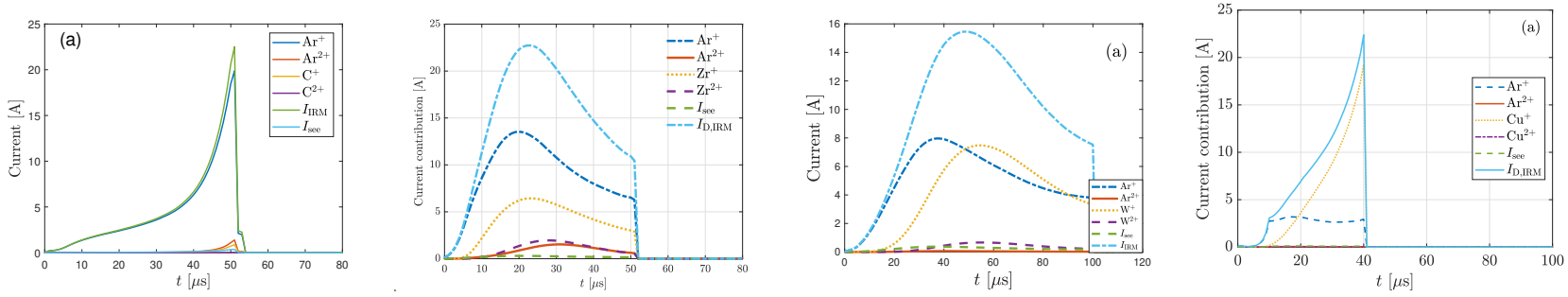
Ionization region model

- The temporal evolution of the neutral and ion densities in a discharge with graphite target
- Ar^+ ions dominate the discharge – constitute over 90% of the discharge current
- Working gas rarefaction is apparent
- The back-attraction probability is high $\beta_{\mathrm{t,pulse}} > 0.83$



From Eliasson et al. (2021) PSST **29** 115017

Ionization region model



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

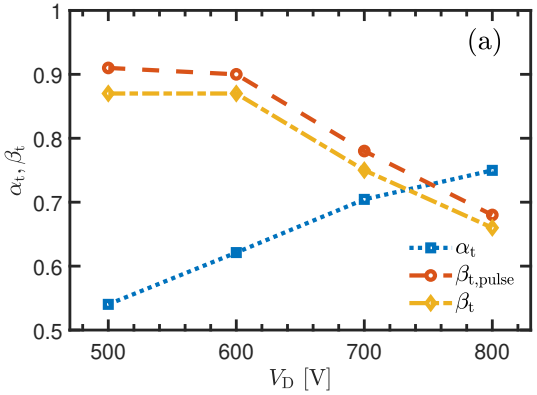
- The temporal evolution of the discharge current composition at the target surface for three different targets
- With Cu target Cu^+ ions dominate, with graphite target Ar^+ ions dominate
- For Zr and W targets there is a mix of Ar^+ and metal ions

Ionization region model

- For tungsten target the ionization probability α_t increases with increased discharge voltage
- The peak discharge current increases with increased discharge voltage
- Earlier we have argued that the ionization probability depends only on the peak discharge current and increases with increased peak discharge current

Rudolph et al. (2022) JPD **55** 015202

- The back-attraction probability $\beta_{t,pulse}$ decreases with increased discharge voltage

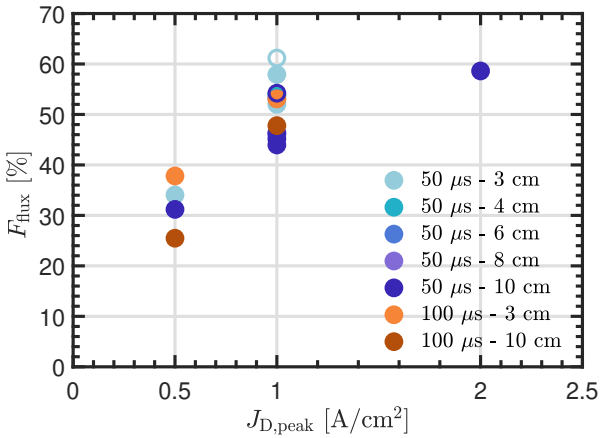


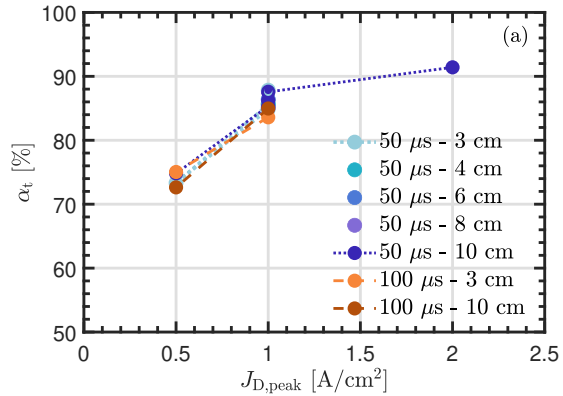
A discharge with a tungsten target

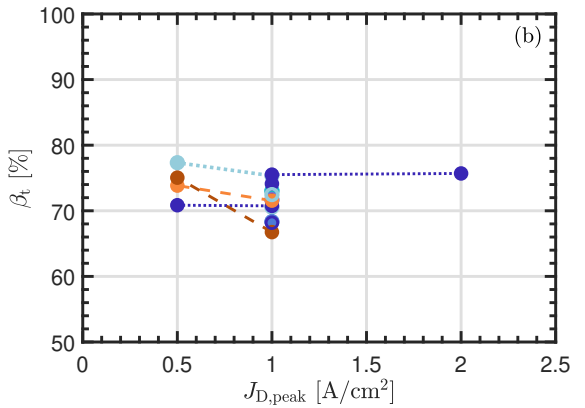
From Suresh Babu et al. (2022) PSST **31** 065009

Ionization region model

- For zirconium target the ionization probability α_t increases with increased current density
- The back-attraction probability $\beta_{t,pulse}$ does not show any trend







- The measured ionized flux fraction is used to lock the model

discharge with a zirconium target

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

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

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

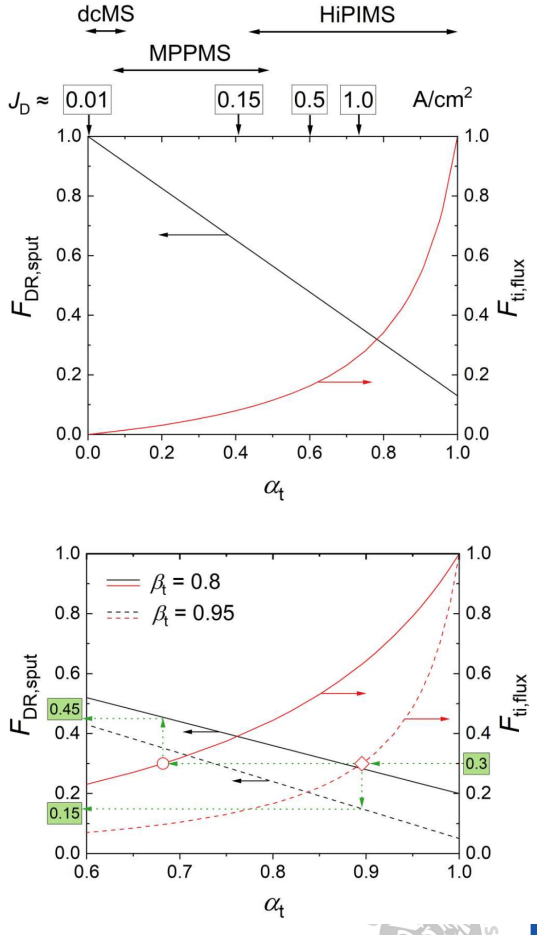
and ionization probability α_t

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

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 ?

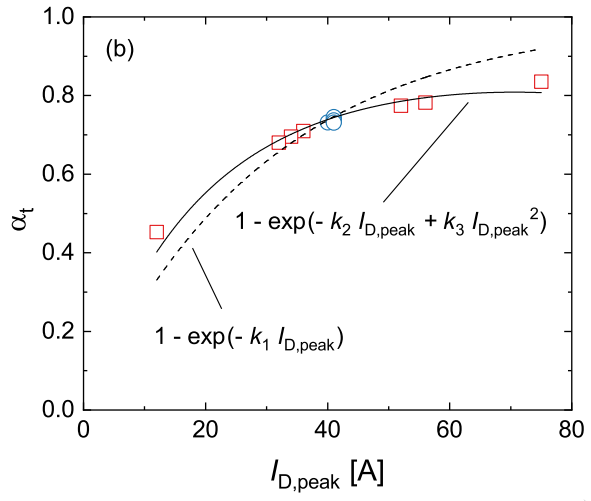


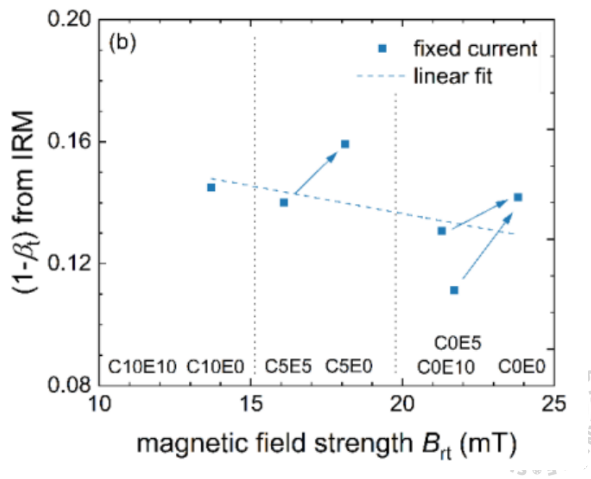
From Brenning et al. (2020) *JVSTA* **38** 033008

Depostion rate – Optimization

- The internal discharge parameters α_t and β_t from the ionization region model (IRM)
- The ionization probability α_t increases with increased discharge current
- The ion escape fraction $(1 - \beta_t)$ versus the magnetic field strength

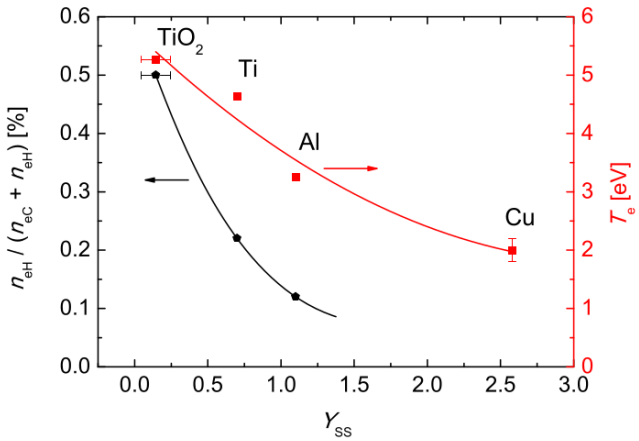
From Rudolph et al. (2022) JPD **55** 015202





Depostion rate – Optimization

- We know that the electron temperature and the hot electron density fall with increased sputter yield
- Held *et al.* observed that titanium atoms are ionized within 0.5 mm from the target surface (high $\beta_{\mathrm{t,pulse}}$), while aluminum and chromium atoms can travel further before being ionized (lower $\beta_{\mathrm{t,pulse}}$)
- The measured electron temperature is 4.5 eV for titanium target compared to 2.6 eV (aluminum) and 1.5 eV (chromium)

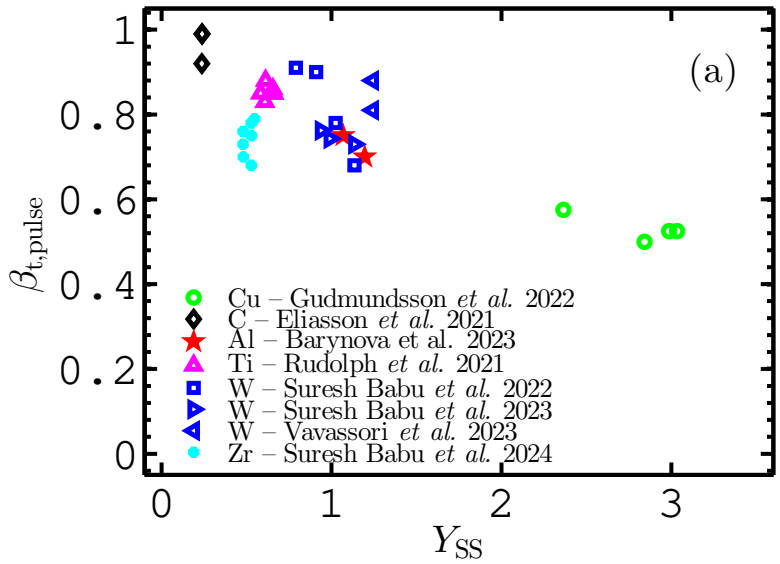


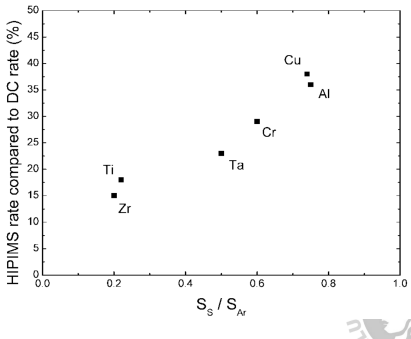
From Brenning et al. (2017) PSST **26** 125003

Held et al. (2023) PSST **32** 065002

Deposition rate – Optimization

- What determines the back-attraction probability ?
- How can one influence the back-attraction probability ?
- The back-attraction probability $\beta_{\mathrm{t,pulse}}$, 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



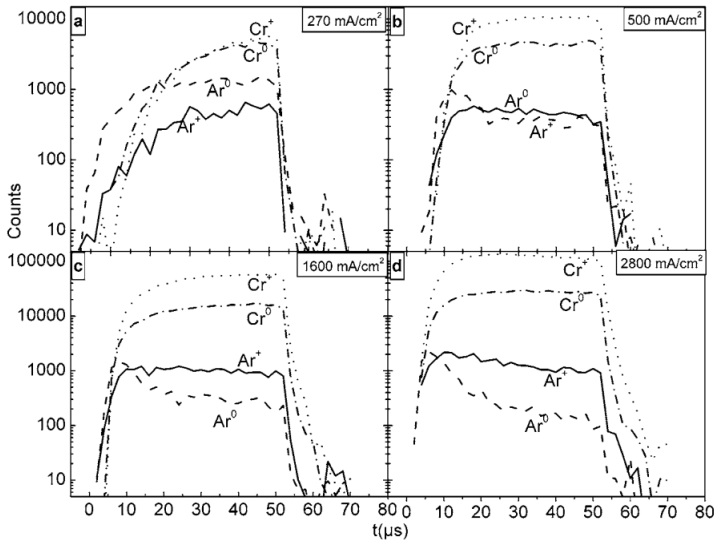


Helmersson et al. (2005) SVC Conf. Proc. 2005, p. 458

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



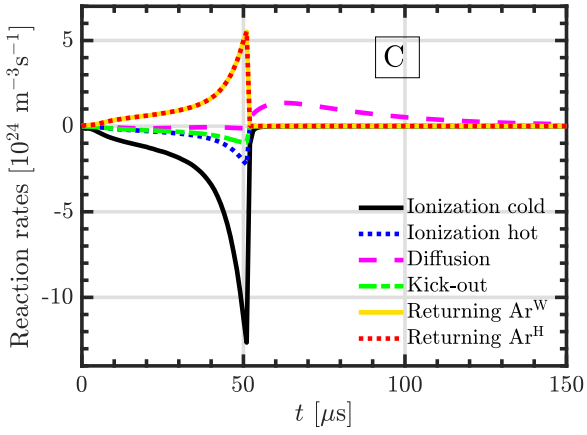
From Alami et al. (2006) APL **89**(15) 154104

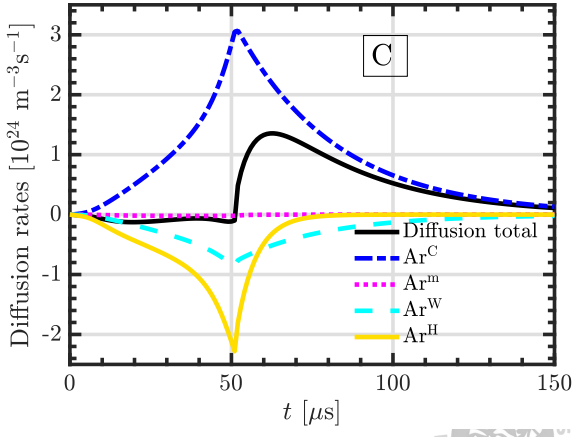
Working gas rarefaction

- 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





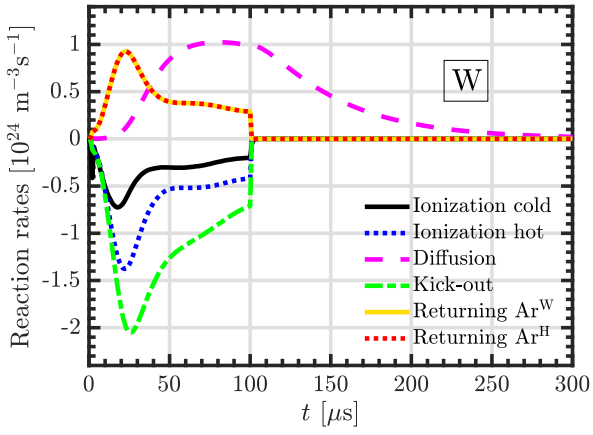
From Barynova et al. PSST **33**(6) 065010

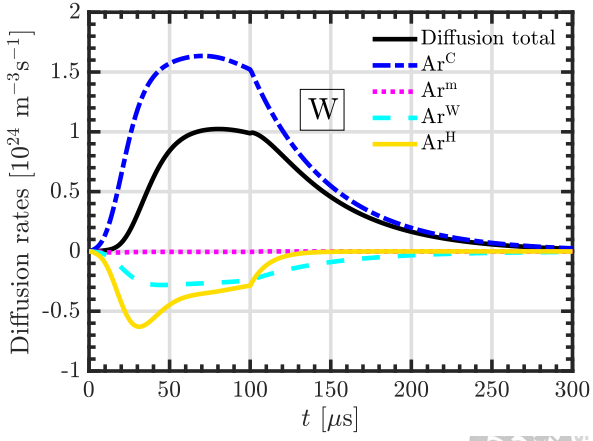
Working gas rarefaction

- 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

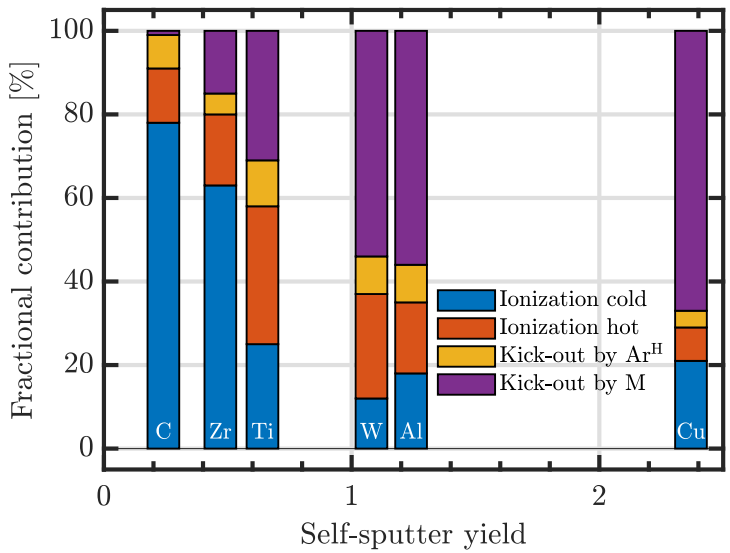




From Barynova et al. PSST **33**(6) 065010

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

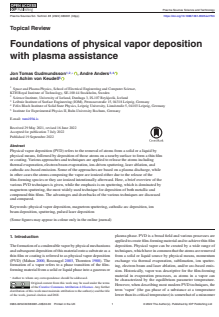
http://langmuir.raunvis.hi.is/~tumi/ranns.html

Further reading

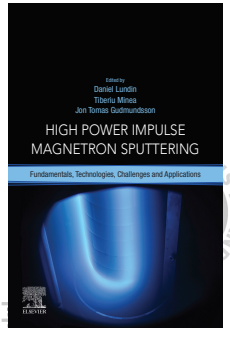
- J. T. Gudmundsson, Physics and technology of magnetron sputtering discharges, Plasma Sources Science and Technology, **29**(11) (2020) 113001
- J. T. Gudmundsson, André Anders, and Achim von Keudell, Foundations of physical vapor deposition with plasma assistance, Plasma Sources Science and Technology, **31**(8) (2022) 083001
- Daniel Lundin, Tiberiu Minea and Jon Tomas Gudmundsson (eds.), High Power Impulse Magnetron Sputtering: Fundamentals, Technologies, Challenges and Applications, Elsevier, 2020


Topical Review

Physics and technology of magnetron sputtering discharges


Topical Review

Foundations of physical vapor deposition with plasma assistance



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