On three different ways to quantify the degree of ionization in sputtering magnetrons

J. T. Guðmundsson[1,2], H. Hajihoseini[2,3], Alexandre Butler[3], Martin Čada[4], Zdenek Hubička[4], Nils Brenning[1,3,5], Michael A Raadu[1], Tiberiu Minea[3], and Daniel Lundin[3,5]

[1] Department of Space and Plasma Physics, School of Electrical Engineering, KTH Royal Institute of Technology, Stockholm, Sweden

[2] Science Institute, University of Iceland, Reykjavik, Iceland

[3] Laboratoire de Physique des Gaz et Plasmas - LPGP, UMR 8578 CNRS, Université Paris-Sud, 91405 Orsay Cedex, France

[4] Institute of Physics v. v. i., Academy of Sciences of the Czech Republic, Na Slovance 2, 182 21 Prague 8, Czech Republic

[5] Plasma and Coatings Physics, IFM-Materials Physics, Linköping University, Sweden

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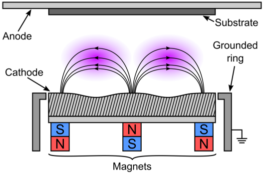
Introduction – Magnetron sputtering

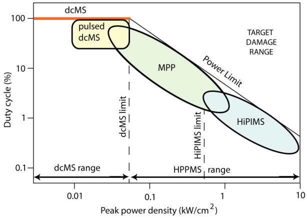
- Magnetron sputtering has been a highly sucessfull technique that is essential in a number of industrial applications



- Conventional 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 – HiPIMS

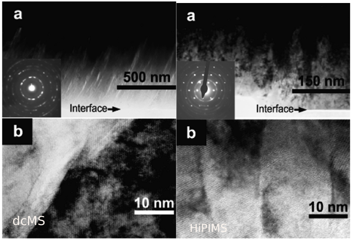
- High ionization of sputtered material requires very high density plasma
- In a conventional dc magnetron sputtering discharge the power density (plasma density) is limited by the thermal load on the target
- High power pulsed magnetron sputtering (HPPMS)
- In a HiPIMS discharge a high power pulse is supplied for a short period
  - low frequency
  - low duty cycle
  - low average power



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

- Power density limits
  $p_t = 0.05$ kW/cm^2 dcMS limit
  $p_t = 0.5$ kW/cm^2 HiPIMS limit

Introduction – fraction of ionization



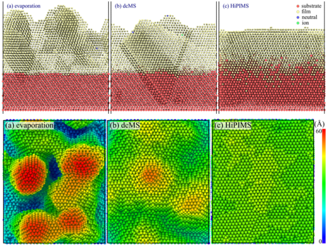
dc magnetron HiPIMS

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

- In HiPIMS deposition, the high fraction of ionization of the sputtered species has been shown to lead to
  - the growth of smooth and dense films
  - enable control over their phase composition and microstructure
  - enhance mechanical and optical properties
  - improving film adhesion
  - enabling deposition of uniform films on complex-shaped substrates
- For optimization of HiPIMS thin film deposition processes, quantification and control of the fraction of ionization of the sputtered species are for obvious reasons key requirements

Introduction – fraction of ionization

- The effect of ionization fraction on the epitaxial growth of Cu film on Cu(111) substrate explored using Molecular Dynamics simulation
- Three deposition methods
  - thermal evaporation, fully neutral
  - dcMS, 50 % ionized
  - HiPIMS, 100 % ionized
- Higher ionization fraction of the deposition flux leads to smoother surfaces by two major mechanisms
  - decreasing clustering in the vapor phase
  - bicollision of high energy ions at the film surface that prevents island growth to become dominant



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

Fraction of ionization

Fraction of ionization

- Quantification and control of the fraction of ionization of the sputtered species are crucial in magnetron sputtering
- We distinguish between three approaches to describe the degree (or fraction) of ionization
  - the ionized flux fraction

$$F_{\mathrm{flux}} = \frac{\Gamma_{\mathrm{i}}}{\Gamma_{\mathrm{i}} + \Gamma_{\mathrm{n}}}$$

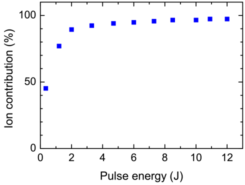
  - the ionized density fraction

$$F_{\mathrm{density}} = \frac{n_{\mathrm{i}}}{n_{\mathrm{i}} + n_{\mathrm{n}}}$$

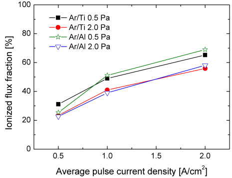
  - the fraction α of the sputtered metal atoms that become ionized in the plasma (probability of ionization)

Fraction of ionization

- There have been conflicting reports on the ionized flux fraction F_{flux}
  - 70 % for Cu (Kouznetsov et al., 1999)
  - 40 % for $Ti_{0.5}Al_{0.5}$ (Macak et al., 2000)
  - 9.5 % for Al (DeKoven et al., 2003)
  - 4.5 % for C (DeKoven et al., 2003)
  - 20 – 60 % for Ti (Kubart et al., 2014)
  - 20 – 68 % for Ti (Lundin et al., 2015)
- The degree of ionization F_{density}
  - 90 % for Ti (Bohlmark et al., 2005)
- The ionization flux fraction depends on applied power, discharge current density, pulse frequency and pulse length and the magnetic field strength



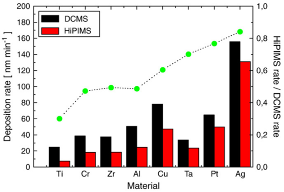
From Bohlmark et al. (2005) JVSTA **23** 18



From Lundin et al. (2015) PSST **24** 035018

Fraction of ionization

- There have been a number of reports demonstrating the lower deposition rate in HiPIMS when compared to dcMS operated at the same average power (Helmersson et al., 2006; Anders, 2010).
- Samuelsson et al. (2010) compared the deposition rates from eight metal targets (Ti, Cr, Zr, Al, Cu, Ta, Pt, Ag) in pure Ar for both dcMS and HiPIMS discharges applying the same average power
- They observed that the HiPIMS deposition rates were in the range of 30 – 85% of the dcMS rates depending on target material.

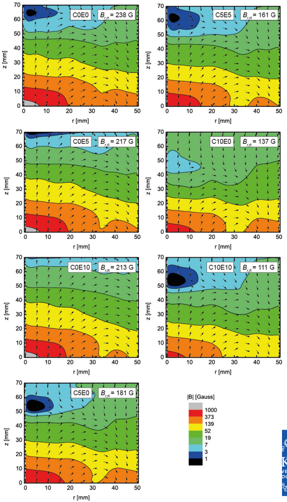


From Samuelsson et al. (2010) SCT **202** 591

Influence of magnetic field

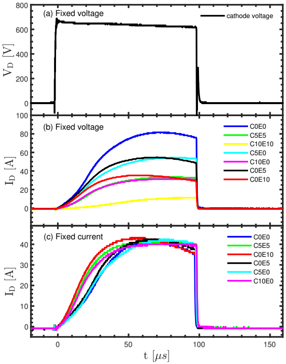
Influence of magnetic field – Deposition rate

- The magnetic field distribution above the target for seven different magnet configurations: C0E0, C5E5 and C10E10, C0E5, C0E10, C5E0, and C10E
- For the configurations investigated, it was found that a magnetic null point was always present, which means that all configurations ware categorized as unbalanced type II
- The magnetic null was used as a measure of the degree of balancing and is in the range 43–74 mm from the target surface above the target center



Influence of magnetic field – Deposition rate

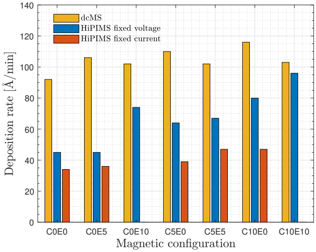
- The HiPIMS discharge current and voltage waveforms recorded for various magnetic field configurations
  - (a) the discharge voltage in fixed voltage mode
  - (b) the discharge current in fixed voltage mode
  - (c) discharge current in fixed peak current mode
- The Ar pressure was set to 1 Pa
- In all cases the pulse width was 100 μs 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 from both dcMS and HiPIMS discharges operated in fixed voltage mode and fixed current mode using various magnetic field configurations measured at 70 mm axial distance over center of cathode
- The magnet configurations on the $x-$axis are ordered from high $|\mathbf{B}|$ at the left to low $|\mathbf{B}|$ on the right
- The recorded $|B_{r,\mathrm{rt}}|$ value above the race track is used as a measure of $|\mathbf{B}|$



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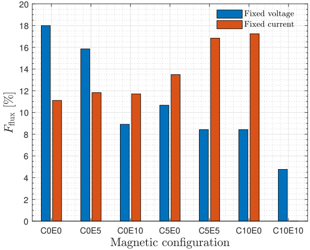
Influence of magnetic field – Deposition rate

- The Ti ionized flux fraction in a HiPIMS discharge using various magnet configurations measured at 70 mm axial distance over the center of the cathode
- The magnet configurations on the $x-$axis are ordered from high $|\mathbf{B}|$ at the left to low $|\mathbf{B}|$ on the right
- The recorded $|B_{r,\mathrm{rt}}|$ value above the race track is used as a measure of $|\mathbf{B}|$



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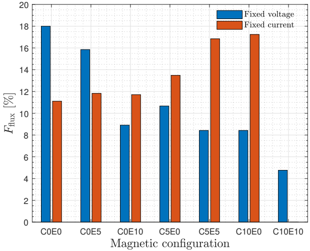
Influence of magnetic field – Deposition rate

- The ionized flux fraction decreases with decreasing $|\mathbf{B}|$ when the HiPIMS discharge is operated in fixed voltage mode
- When operating in fixed peak current mode the ionized flux fraction F_{flux} increases slightly with decreasing $|\mathbf{B}|$
- In this case ionized flux fraction increases from 11% to 16.8% when comparing cases C0E0 and C5E5 (no data from C10E10), i.e. by a factor 1.5 when decreasing $|\mathbf{B}|$



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Influence of magnetic field – Deposition rate

- We derive a few general equations that relate the measured quantities deposition rate and the ionized flux fraction to the parameters α_t and β_t
- Let us call the total flux (atoms/s) of atoms sputtered from the target Γ_0 and the flux of sputtered species (ions and neutrals) that leave the ionization region (IR) towards the diffusion region (DR) Γ_DR
- The useful fraction of the sputtered species becomes

$$F_\mathrm{DR} = \frac{\Gamma_\mathrm{DR}}{\Gamma_0} = (1 - \alpha_\mathrm{t}\beta_\mathrm{t})$$

- This equation indicates a reduced fraction of the sputtered species reaching the substrate when the ionization of the sputtered material increases

Influence of magnetic field – Deposition rate

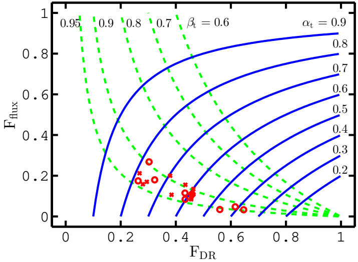
- Recall that the main drawback using HiPIMS is the low deposition rate
- A relationship between the ionization flux fraction F_{flux} and the parameters α_{t} and β_{t} has been derived from the pathway model (Vlček and Burcalová, 2010; Butler et al., 2018)

$$F_{\text{flux}} = \frac{\Gamma_{\text{DR,ions}}}{\Gamma_{\text{DR}}} = \frac{\Gamma_0 \alpha_{\text{t}}(1-\beta_{\text{t}})}{\Gamma_0(1-\alpha_{\text{t}}\beta_{\text{t}})} = \frac{\alpha_{\text{t}}(1-\beta_{\text{t}})}{(1-\alpha_{\text{t}}\beta_{\text{t}})}$$

  where no additional ionization of the sputtered material in the diffusion region is assumed
- Our goal is to assess how much $|\mathbf{B}|$ and the magnetic field structure influence α_{t} and β_{t}, repectively

Influence of magnetic field – Deposition rate

- A graph that shows F_{DR} on the horizontal axis, and F_{flux} on the vertical axis
- We have also plotted two sets of lines
  - lines of constant β_{t} with α_{t} varied from 0 to 1 (green dashed lines)
  - lines of constant α_{t}, with β_{t} varied from 0 to 1 (blue solid lines)
- Plotting the experimentally determined combinations of F_{DR} and F_{flux} in this plane gives us estimates of the corresponding values of α_{t} and β_{t}



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Influence of magnetic field – Deposition rate

- We can derive an equation that gives the back attraction probability $\beta_{\rm t}$ as a function of the measured quantities $F_{\rm flux}$ and $F_{\rm DR}$

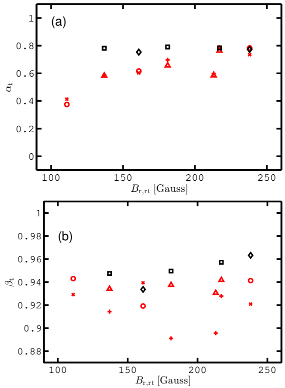
$$\beta_{\rm t} = \frac{1 - F_{\rm DR}}{1 - F_{\rm DR}(1 - F_{\rm flux})}$$

and similarly we can derive an equation that gives $\alpha_{\rm t}$ as a function of the measured quantities

$$\alpha_{\rm t} = 1 - F_{\rm DR}(1 - F_{\rm flux}).$$

Influence of magnetic field – Deposition rate

- When operating in the fixed voltage mode (red) the ionization probability α_{t} increases with increased magnetic field strength
- When operating in the fixed peak current mode the ionization probability α_{t} is roughly constant independent of the magnetic field strength
- The back attraction probability is always high in the range 0.89 – 0.96 over the entire range of $B_{r,\mathrm{rt}}$



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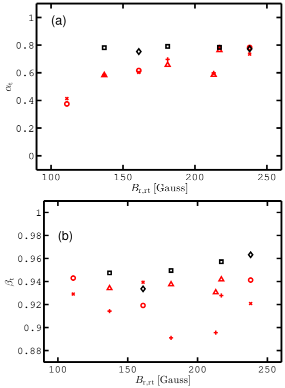
Influence of magnetic field – Deposition rate

- In the fixed peak current mode (**black**) β_{t} increases slightly with increased $|\mathbf{B}|$ in the range 0.93 – 0.96 while α_{t} is almost constant in a narrow range 0.75 – 0.79
- If we assume a linear increase in β_{t} with $|\mathbf{B}|$ the fraction $(1-\beta_{\mathrm{t}})$ is roughly 30% higher at the highest $|\mathbf{B}|$ than at the lowest $|\mathbf{B}|$
- Recall that the total flux of ions of the sputtered material away from the target toward the substrate is $\Gamma_{\mathrm{DR,ions}} = \alpha_{\mathrm{t}}(1-\beta_{\mathrm{t}})\Gamma_0$



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Summary

Summary

- For HiPIMS in the fixed voltage mode: A trade-off between the deposition rate (decreases by more than a factor of two) and the ionized flux fraction (increases by a factor 4 to 5) with increasing $|\mathbf{B}|$
- For HiPIMS in the fixed peak current mode: Decreasing $|\mathbf{B}|$ improves both the deposition rate (by 40%) and the ionized flux fraction (by 60%)
- When operating in the fixed peak current mode the ionization probability of the sputtered species is roughly constant while the parameter $(1-\beta_{\mathrm{t}})$ increases roughly 30% with decreasing $|\mathbf{B}|$
- When operating a HiPIMS discharge in fixed voltage mode the ionization probability α_{t} is varied by $|\mathbf{B}|$ and β_{t} remains roughly constant, while in the fixed peak current mode β_{t} varies with $|\mathbf{B}|$ and α_{t} remains roughly constant

Thank you for your attention

The slides can be downloaded at

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

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