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Introduction – Magnetron sputtering

Magnetron sputtering is a highly successful and widely ۲ used technique for thin film deposition

> . Substrate Grounded Magnets

Gudmundsson (2020) PSST 29 113001

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

 If the cathode plate is circular, the magnetic confinement is seen as a torus shaped plasma that hovers in front of the target



Introduction



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)
- 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





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 KTH Huo et al. (2017) JPD **50** 354009

Recycling in HiPIMS discharges



- 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

$$I_{\mathrm{crit}} = S_{\mathrm{RT}} e p_{\mathrm{g}} \sqrt{rac{1}{2\pi m_{\mathrm{g}} k_{\mathrm{B}} T_{\mathrm{g}}}} = S_{\mathrm{RT}} e n_{\mathrm{g}} \sqrt{rac{k_{\mathrm{B}} T_{\mathrm{g}}}{2\pi m_{\mathrm{g}}}}$$

 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

> Anders et al. (2012) JPD **45** 012003 Huo et al. (2014) PSST **23** 025017



- For a 50 mm diameter AI target the critical current is $I_{\rm crit} \approx$ 7 A
- The experiment is operated from far below *I*_{crit} to high above it, up to 36 A.
- With increasing discharge current *I*_{prim} gradually becomes a very small fraction of the total discharge current *I*_D
- The current becomes mainly carried by singly charged Al⁺-ions, meaning that self-sputter recycling or the current I_{SS-recycle} dominates

From Huo et al. (2017) JPD 50 354003

Experimental data from Anders et al. (2007) JAP 102 113303



- With increased discharge voltage the discharge with AI target moves from the dcMS regime to the HiPIMS discharge regime – type A
- For reactive sputtering of Ti target in poisoned mode working gas recycling dominates – type B



 Recycling map for five different targets with varying self-sputter yield

•
$$Cu - Y_{SS} = 2.6$$

• Al –
$$Y_{\rm SS} = 1.1$$

•
$$Ti - Y_{SS} = 0.7$$

•
$$C - Y_{SS} = 0.5$$

•
$$TiO_2 - Y_{SS} = 0.04 - 0.25$$

- For very high self-sputter yields Y_{SS} > 1, the discharges above *l*_{crit} are of type A with dominating SS-recycling
- For very low self-sputter yields Y_{SS} < 0.2, the discharges above I_{crit} are of type B with dominating working gas recycling



From Brenning et al. (2017) PSST 26 125003



Recycling in HiPIMS discharges – copper

- The temporal evolution of the discharge current composition at a Cu target surface for a peak discharge current density 2 A/cm²
- A discharge with 2 inch copper target $I_{\rm crit} \approx 3.8$ A
- The Cu⁺ ion is the dominating positively charged species in the discharge
- The ionized flux fraction of copper is roughly 15 %



From Gudmundsson et al. (2021) manuscript in preparation



Recycling in HiPIMS discharges – carbon

- The temporal evolution of the discharge current composition at a graphite target surface for a peak discharge current density 2 A/cm²
- A discharge with 2 inch graphite target − *I*_{crit} ≈ 7.6 A
- The Ar⁺ ion is the dominating positively charged species in the discharge
- Less than 5 % of the total discharge current is carried by C⁺ ions
- The ionized flux fraction of carbon is roughly 2 %



From Eliasson et al. (2021) manuscript in preparation



- Recycling loops
- Discharge with Al or Cu target SS recycling dominates
 - high self sputter yield
- Reactive discharge with graphite or TiO₂ target – working gas recycling dominates
 - low self sputter yield



Electron power absorption



Electron power absorption



T. J. Petty, LPGP, Université Paris Sud

Gudmundsson and Hecimovic (2017) PSST 26 123001

- A dc discharge with a cold cathode is sustained by secondary electron emission from the cathode due to ion bombardment
- The discharge current at the target consists of electron current *I*_e and ion current *I*_i or

$$I_{\rm D} = I_{\rm e} + I_{\rm i} = I_{\rm i}(1 + \gamma_{\rm see})$$

where γ_{see} is the secondary electron emission coefficient

 Note that γ_{see} ~ 0.05 – 0.2 for most metals, so at the target ion current dominates



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Electron power absorption

 In magnetron sputtering effective secondary electron emission coefficient

 $\gamma_{\text{see,eff}} = m\epsilon_{\text{e}}(1-r)\gamma_{\text{see}}$

where r is the recapture probability

• To sustain the discharge the condition

$$\gamma_{\text{see,eff}} \mathcal{N} = \mathbf{1}$$

with $\ensuremath{\mathcal{N}}$ the number of electron-ion pairs defines the minimum voltage (Thornton equation)

$$V_{\mathrm{D,min}} = rac{\mathcal{E}_{\mathrm{c}}}{\beta \gamma_{\mathrm{see,eff}}}$$

Magnetron sputtering: basic physics and application to cylindrical magnetrons

John A. Thornton

ida Carporation, 2017 Colorado Avenue, Sante Monice, California 9040 Received 22 Beptember 1977; accepted 7 December 1977)

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PACS numbers: 81.15.-z, 52.75.-d

Thornton (1978) JVST 15(2) 171



Electron power absorption

- However, the presence of a transverse magnetic field in magnetron sputtering enables a potential drop to exist outside the cathode sheath
- A potential $V_{\rm SH}$ falls over the sheath, and the rest of the applied voltage, $V_{\rm IR} = V_{\rm D} - V_{\rm SH}$, falls across the extended pre-sheath, the ionization region (IR), $\delta_{\rm IR} = V_{\rm IR}/V_{\rm D}$
- Ohmic heating, the dissipation of locally deposited electric energy J_e · E to the electrons in the plasma volume outside the sheath



From Brenning et al. (2016) PSST 25 065024



Electron power absorption

 The fraction of the total ionization that is due to Ohmic heating can be obtained directly from the line fit parameters *a* and *b* of the 1/V_D vs γ_{see} curves for dcMS

$$\frac{\iota_{\rm Ohmic}}{\iota_{\rm total}} = \frac{b}{a\gamma_{\rm see} + b}$$

 The fraction of the discharge voltage that falls over the ionization region

$$\delta_{\mathrm{IR}} = \frac{V_{\mathrm{IR}}}{V_{\mathrm{D}}} = 0.15 - 0.19$$



Electron power absorption

- Applying the ionization region model (IRM) to a HiPIMS discharge
- For the AI target, Ohmic heating is in the range of 87 % (360 V) to 99 % (1000 V)
- The domination of Al⁺-ions, which have zero secondary electron emission yield, has the consequence that there is negligible sheath energization
- For a Ti target Ohmic heating is about 92 %



Electron power absorption

- Discharges with Ti target with adjustable confining magnetic field Hajihoseini et al. (2019) Plasma 2 201
- $P_{\text{Ohm}}/(P_{\text{Ohm}} + P_{\text{SH}})$ versus the magnetic field parameter z_{gap} (magnetic field strength)
- For increasing z_{gap} (lower magnetic field), the fraction $P_{Ohm}/(P_{Ohm} + P_{SH})$ decreases
- P_{Ohm}/(P_{Ohm} + P_{SH}) can be regarded as a measure for energy efficiency of a discharge



From Rudolph et al. (2021) JPD submitted for publication



Electron power absorption



From Rudolph et al. (2021) JPD submitted for publication

- The use of the pulse power for different values of z_{gap}
 - ion acceleration (P_{ion})
 - Ohmic heating (P_{Ohm})
 - sheath energization ($P_{\rm SH}$).
- Most of the pulse power (*P*_{pulse}) is used to accelerate ions and this power is finally dissipated in the target as heat
- The fraction of the pulse power that is absorbed by the electrons decreases for higher values of z_{gap} and more energy is spent on heating up the target



Summary



Summary

- In HiPIMS discharge operation there is always recycling:
 - For high currents the discharge with Al or Cu target develops almost pure **self-sputter recycling**, while the discharge with Ti target exhibits close to a 50/50 combination of **self-sputter recycling** and **working gas-recycling**
 - For a poisoned Ti, or a graphite target the sputter yield is low and working gas-recycling necessary at high currents
- Ohmic heating of the electrons can play a significant role in both dc magnetron sputtering discharge and in particular HiPIMS
- The fraction of the total electron heating that is attributable to Ohmic heating is over 90 % in the HiPIMS discharge



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Thank you for your attention

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 - Prof. Nils Brenning, KTH Royal Institute of Technology, Stockholm, Sweden
 - Dr. Michael A. Raadu, KTH Royal Institite of Technology, Stockholm, Sweden
 - Dr. Martin Rudolph, Leibniz Institute of Surface Engineering (IOM), Leipzig, Germany
 - Prof. Tiberu Minea, Université Paris-Sud, Orsay, France
 - Dr. Hamidreza Hajihoseini, now at University of Twente, The Netherlands

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. Andersson, and A. Ehiasarian (2007). High power impulse magnetron sputtering: Current-voltage-time characteristics indicate the onset of sustained self-sputtering. *Journal of Applied Physics 102*(11), 113303.
- 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., J. T. Gudmundsson, D. Lundin, T. Minea, M. A. Raadu, and U. Helmersson (2016). The role of Ohmic heating in dc magnetron sputtering. *Plasma Sources Science and Technology 25*(6), 065024.
- 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.
- Depla, D., S. Mahieu, and R. De Gryse (2009). Magnetron sputter deposition: Linking discharge voltage with target properties. *Thin Solid Films* 517(9), 2825–2839.
- Gudmundsson, J. T. (2020). Physics and technology of magnetron sputtering discharges. Plasma Sources Science and Technology 29(11), 113001.
- Gudmundsson, J. T. and A. Hecimovic (2017). Foundations of dc plasma sources. Plasma Sources Science and Technology 26(12), 123001.
- 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.
- Gudmundsson, J. T., D. Lundin, N. Brenning, M. A. Raadu, C. Huo, and T. M. Minea (2016). An ionization region model of the reactive Ar/O₂ high power impulse magnetron sputtering discharge. *Plasma Sources Science and Technology 25*(6), 065004.



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.
- 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.
- Rudolph, M., N. Brenning, H. Hajihoseini, M. A. Raadu, T. M. Minea, A. Anders, D. Lundin, and J. T. Gudmundsson (2021). Influence of the magnetic field on the discharge physics of a high power impulse magnetron sputtering discharge. *Journal of Physics D: Applied Physics*, submitted for publication.
- 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.
- 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., 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.



