Modeling of high power impulse magnetron sputtering discharges with graphite target

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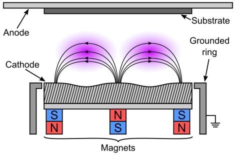
Introduction – Magnetron sputtering

- Magnetron sputtering is a highly sucessfull and widely used technique for thin film deposition

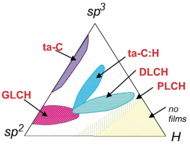
Gudmundsson (2020) PSST **29** 113001



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

- When operated as a dc magnetron sputtering (dcMS) discharge the film-forming material consists mainly of neutral atoms
- Most of the ions available in the discharge, and the ions that bombard the substrate, are ions of a noble working gas

Introduction – Diamond like carbon films

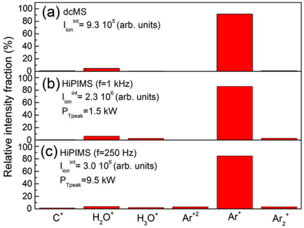


Casiraghi et al. (2007), *Materials Today* **10** 44

- For many applications, it is desired to have a high ionization fraction in the flux of the film-forming material
- When depositing amorphous carbon films bombardment by energetic ions increases the sp^3 content in the films
- Diamond-like carbon (DLC) refers to a metastable form of amorphous carbon that contains a significant fraction of tetrahedrally bonded sp^3
- These films exhibit high hardness, are resistant to scratching, have high dielectric constants, high index of refraction and excellent optical transparency

Introduction – HiPIMS

- In high power impulse magnetron sputtering (HiPIMS) pulses, that have a peak power density are delivered at low repetition frequency and low duty cycle and lead to ionized flux of the sputtered species – at least for metal targets



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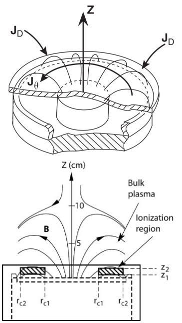
Sarakinos et al. (2012), *SCT* **206** 2706

- However, it has turned out to be a significant challenge to ionize carbon atoms, that are sputtered off of a graphite target in a magnetron sputtering discharge
- The measured ionized flux fraction in HiPIMS discharge with a graphite target is below 5 % DeKoven et al. (2003) SVC Proceedings

Ionization region model

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 in

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

Ionization region model non-reactive HiPIMS

- 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
- 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)
  - Carbon atoms $C(1s^2 2s^2 2p^2)$ $^3P_{0,1,2}$, three metastable carbon atom states (^1D, ^1S, $^5S^{\circ}$), carbon ions C^{+} (11.26 eV), doubly ionized carbon ions C^{2+} (24.38 eV)

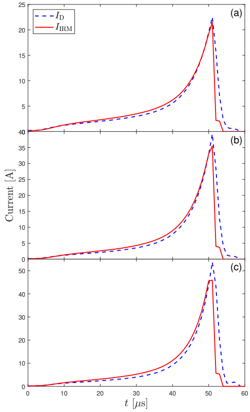
Detailed model description is given in Huo et al. (2017), JPD **50** 354003

The carbon discharge model is given by Eliasson et al. (2021), PSST **30** 115017

Model results

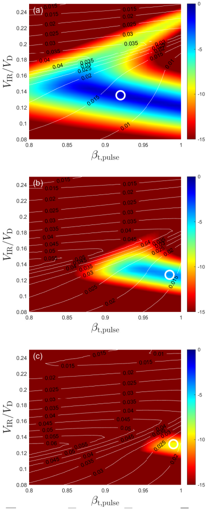
Discharge current waveform

- The discharge voltage and current waveforms were measured for a HiPIMS discharge in argon (1 Pa) with a graphite target
- The sputter target was a 2 inch graphite disk mounted on the magnetron assembly
- The average discharge power $\langle P_{\mathrm{D}} \rangle = 80$W maintained by varying the repetition frequency, as the pulse was kept at 50 μs
- Three peak discharge currents were investigated $I_{\mathrm{D,peak}}$ = 20, 40, and 60 A, leading to peak discharge current densities of 1, 2, and 3 A cm^{-2}, respectively



Fitting map

- The best fit to the experimentally determined discharge current is determined using a fitting map
- It shows the fraction of the discharge voltage that drops across the IR $f = V_{\rm IR}/V_{\rm D}$ versus the back-attraction probability of an ion of the sputtered species during the pulse $\beta_{\rm t,pulse}$
- The blue zones in the fitting map indicate the combinations where the root mean square deviation is the smallest
- The white circles show where a well fitted discharge current profile is observed



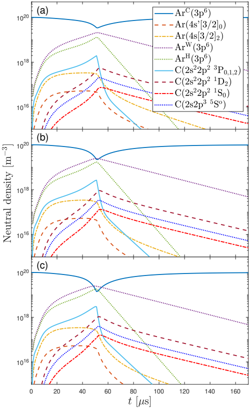
Internal discharge parameters

$I_{\mathrm{D,peak}}$	20 A	40 A	60 A
$J_{\mathrm{D,peak}}$	1 A/cm^2	2 A/cm^2	3 A/cm^2
$\langle \alpha_{\mathrm{t,pulse}} \rangle$	13 %	21 %	27 %
$\beta_{\mathrm{t,pulse}}$	92 %	99 %	99 %
$f = V_{\mathrm{IR}}/V_{\mathrm{D}}$	14 %	13 %	13 %
F_{flux}	1.3 %	1.6 %	2.2 %
rarefaction	66 %	78 %	86 %

- The back-attraction probability is very high > 0.9
- The ionization probability is low
- The ionized flux fraction is low ~ 2 %
- Rarefaction is significant (> 50%)

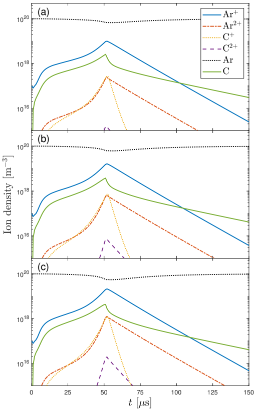
Neutral densities

- The temporal evolution of the neutral particle densities are similar for all the three peak discharge currents
- The ground state working gas argon atoms dominate the discharge but their density decreases with increased discharge current during the pulse, exhibiting a minimum at the end of the pulse – working gas rarefaction
- All carbon species densities increase as more and more carbon is sputtered off the target during the pulse



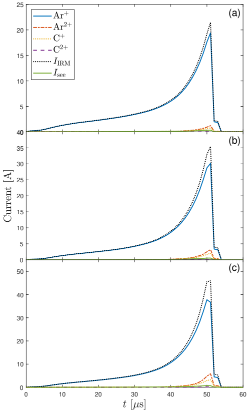
Ion densities

- The Ar^+ ion is the dominating positively charged species in the discharge and its density peaks at the end of the pulse
- The density of the C^+ ions is more than one order of magnitude smaller, and comparable to the Ar^{2+} density
- The C^{2+} ion density is more than three orders of magnitude smaller than the Ar^+ ion density



Current composition

- At the target surface, the discharge current is mainly due to Ar^+ ions, with a small contribution from Ar^{2+} and C^+ ions, while secondary electrons, and C^{2+} ions, have a negligible contribution
- Less than 5 % of the total discharge current at the target surface, is carried by carbon ions
- This agrees with that Ar^+ ions dominate the discharge, while C^+ ions constitute only about 1 % of the total ionic contribution in the substrate vicinity Sarakinos et al. (2012), *SCT* **206** 2706



Recycling in HiPIMS discharges

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

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

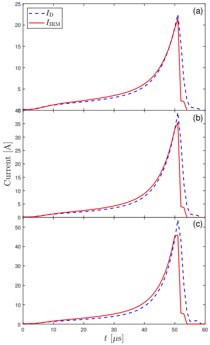
- Discharge currents I_{D} above I_{crit} are only possible if there is some kind of recycling of atoms that leave the target, become subsequently ionized and then are drawn back to the target

Anders et al. (2012) JPD **45** 012003

Huo et al. (2014) PSST **23** 025017

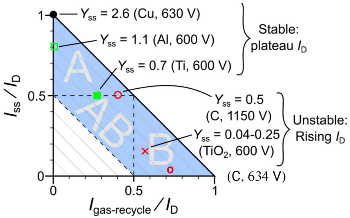
Recycling in HiPIMS discharges

- For the 2 inch diameter graphite target the critical current is $I_{\rm crit} \approx 7.6$ A
- The experiment is operated from far below $I_{\rm crit}$ to high above it, up to 40 A
- With increasing discharge current $I_{\rm prim}$ gradually becomes a very small fraction of the total discharge current $I_{\rm D}$
- The current becomes mainly carried by singly charged Ar^{+}-ions, meaning that **working gas recycling** dominates
- Less than 5 % of the total discharge current is carried by C^{+} ions



Recycling in HiPIMS discharges

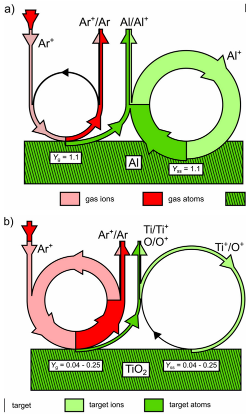
- Recycling map for five different targets with varying self-sputter yield
  - Cu – $Y_{SS} = 2.6$
  - Al – $Y_{SS} = 1.1$
  - Ti – $Y_{SS} = 0.7$
  - C – $Y_{SS} = 0.5$ (1150 V)
  - C – $Y_{SS} = 0.24$ (634 V)
  - TiO_2 – $Y_{SS} = 0.04 - 0.25$
- For very high self-sputter yields $Y_{SS} > 1$, the discharges above I_{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

- Recycling loops
- Discharge with Al or Cu target – SS recycling dominates
  - high self sputter yield
- Reactive discharge with TiO_2 target and discharge with graphite target at low voltage – working gas recycling dominates
  - low self sputter yield



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

Summary

Summary

- An ionization region model (IRM) of a HiPIMS discharge in argon with a graphite target has been developed
- The ionized flux fraction for carbon is low or about 2% in the discharge, lower than typically observed for a HiPIMS discharge with a metallic target
- This is due to
  - low ionization probability of carbon caused by the low electron impact ionization cross section of the carbon atom
  - the high ionization potential
  - high cohesive energy
  - low mass of the carbon atom, causes the sputtered carbon atoms to pass the IR at high velocity
- The discharge is governed by working gas recycling and the Ar^+ ion dominates the discharge

Thank you for your attention

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The slides can be downloaded at

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

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