

On electron heating in magnetron sputtering discharges

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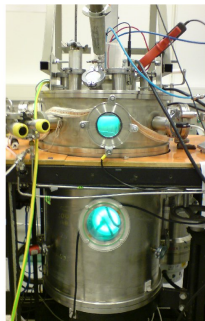
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Introduction

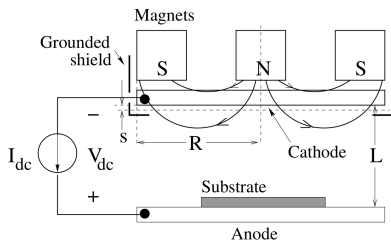
- Magnetron sputtering has been the workhorse of plasma based sputtering methods for almost five decades
- Magnetron sputtering discharges are widely used in thin film processing

- Applications include
 - thin films in integrated circuits
 - magnetic material
 - hard, protective, and wear resistant coatings
 - optical coatings
 - decorative coatings
 - low friction films



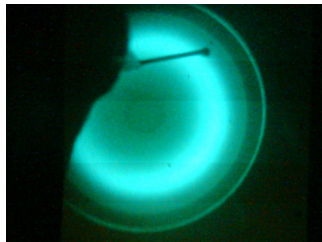
Introduction

- A magnet is placed at the back of the cathode target with the pole pieces at the center and perimeter
- The magnetic field confines the energetic electrons near the cathode
- The electrons undergo numerous ionizing collisions before being lost to a grounded surface



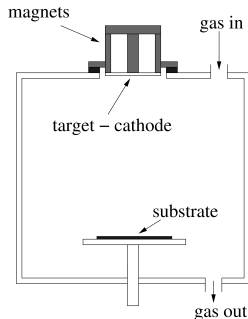
Introduction

- The majority of the ionization events occur in this region where the energetic electrons are trapped
- In the presence of an electric field and magnetic field the electron exhibits a Hall drift, or $\mathbf{E} \times \mathbf{B}$ -drift – this drift is in the azimuthal direction
- If the cathode plate is circular, the magnetic confinement is seen as a torus shaped plasma that hovers in front of the target



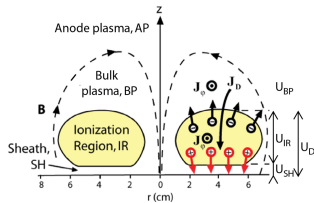
Introduction

- Magnetron sputtering has been a highly successful technique that has a number of industrial applications
- The conventional wisdom is that plasma generation is based on the supply of energy via secondary electrons (SEs) accelerated from the target
- However, one of the remaining fundamental questions is **how electrons are heated in the magnetron sputtering discharge**



Introduction

- A separation of the sputtering magnetron discharge into regions based on the dominating physics
- The sheath (SH) is defined by a substantial charge imbalance such that $(n_i - n_e)/n_e \geq 1$
- The ionization region (IR) is where most of the ionization occurs
- In the bulk plasma (BP), cross-**B** current transport of electrons from the IR to the anode plasma is the key process

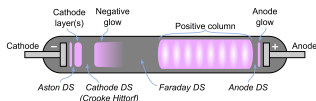


From Huo et al. (2017) JPD (submitted 2017)

dc magnetron sputtering discharge



dc magnetron sputtering discharge



T. J. Petty, LPGP, Université Paris Sud

- A dc discharge with a cold cathode is sustained by secondary electron emission from the cathode by ion bombardment
- The discharge current at the target consists of electron current I_e and ion current I_i or

$$I_D = I_e + I_i = I_i(1 + \gamma_{SE})$$

where γ_{SE} is the secondary electron emission coefficient

- Note that $\gamma_{SE} \sim 0.05 - 0.2$ for most metals, so at the target, the dominating fraction of the discharge current is ion current

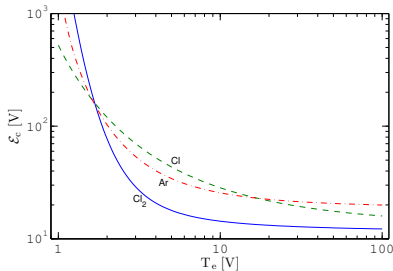


dc magnetron sputtering discharge

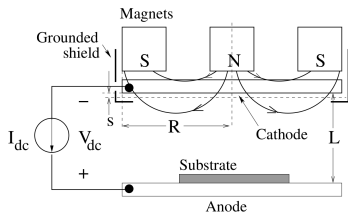
- These secondary electrons are accelerated in the cathode dark space – referred to as primary electrons
- They must produce sufficient number of ions to release more electrons from the cathode
- The number of electron-ion pairs created by each secondary electron is then

$$\mathcal{N} \approx \frac{V_D}{\mathcal{E}_c}$$

where \mathcal{E}_c is the energy loss per electron-ion pair created



dc magnetron sputtering discharge



- To account for the electrons that are not trapped we define an effective secondary electron emission coefficient

$$\gamma_{SE,eff} = m\epsilon_e(1 - r)\gamma_{SE}$$

- ϵ_e is the fraction of the electron energy that is used for ionization before being lost
- m is a factor that accounts for secondary electrons ionizing in the sheath
- r is the recapture probability of secondary electrons

dc magnetron sputtering discharge

- To sustain the discharge the condition

$$\gamma_{SE,eff} \mathcal{N} = 1$$

has to be fulfilled

- This defines the minimum voltage to sustain the discharge as

$$V_{D,min} = \frac{\mathcal{E}_c}{\beta \gamma_{SE,eff}}$$

referred to as Thornton equation

- β is the fraction of ions that return to the cathode

Magnetron sputtering: basic physics and application to cylindrical magnetrons

John A. Thornton

Telic Corporation, 1631 Colorado Avenue, Santa Monica, California 90404
(Received 22 September 1977; accepted 7 December 1977)

Magnetron sputtering sources can be defined as diode devices in which magnetic fields are used in concert with the cathode surface to form electron traps which are so configured that the $\mathbf{E} \times \mathbf{B}$ electron-drift currents close on themselves. Coaxial cylindrical magnetron sputtering sources in which post or hollow cathodes are operated in axial magnetic fields have been reported for a number of years. However, their performance is limited by end losses. A remarkable performance is achieved when the end losses are eliminated by proper shaping of the magnetic field or by using suitably placed electron-reflecting surfaces. High currents and sputtering rates can be obtained, nearly independent of voltage, even at low pressures. This characterizes what has been defined as the *magnetron mode* of operation. This paper reviews the basic principles that underly the operation of dc sputtering sources in the magnetron mode with particular emphasis on cylindrical magnetrons. The important attributes of these devices as sputtering sources are also reviewed.

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

Thornton (1978) JVST **15**(2) 171



dc magnetron sputtering discharge

- The basic assumption is that acceleration across the sheath is the main source of energy for the electrons
- Above breakdown the parameters m , β , ϵ_e and r can vary with the applied voltage
- We can rewrite the Thornton equation for any voltage

$$\frac{1}{V_D} = \frac{\beta m \epsilon_e (1 - r)}{\mathcal{E}_c} \gamma_{SE}$$

A low-pressure cold-cathode discharge is maintained primarily by secondary electrons emitted from the cathode by ion bombardment. These electrons are accelerated in the CDS and enter the plasma where, known as primary electrons, they must produce sufficient ions to release one further electron from the cathode.⁷² This requirement can be expressed by the following relationship for the minimum potential to sustain such a discharge.⁷³

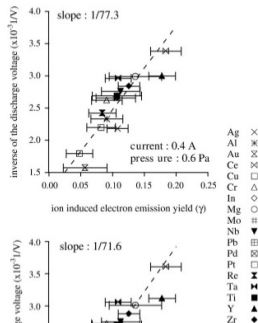
$$V_{\min} = \mathcal{E}_0 / \Gamma_i \epsilon_i \epsilon_e \quad (5)$$

Thornton (1978) *JVST* **15**(2) 171

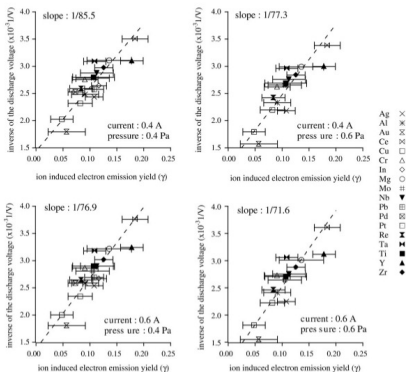


dc magnetron sputtering discharge

- A plot of the inverse discharge voltage $1/V_D$ against γ_{SE} should then give a straight line through the origin
- Depla et al. measured the discharge voltage for a 5 cm diameter target for Ar working gas for 18 different target materials
- Since all the data is taken in the same magnetron, at same current and pressure, the discharge parameters m , β , ϵ_e and \mathcal{E}_c are independent of γ_{SE}



dc magnetron sputtering discharge



From Depla et al. (2009) TSF 517 2825

- $1/V_D$ against γ_{SE} for gas pressures of 0.4 and 0.6 Pa and discharge currents 0.4 A and 0.6 A
- It can be seen that a straight line indeed results, but that it does not pass through the origin

dc magnetron sputtering discharge

- We here propose that the intercept is due to Ohmic heating
- We can now write the inverse discharge voltage $1/V_D$ in the form of a generalized Thornton equation

$$\frac{1}{V_D} = \underbrace{\frac{\beta \epsilon_e^H m (1-r)(1-\delta_{IR})}{\mathcal{E}_c^H}}_a \gamma_{SE} + \underbrace{\frac{\epsilon_e^C \langle I_e/I_D \rangle_{IR} \delta_{IR}}{\mathcal{E}_c^C}}_b$$

or

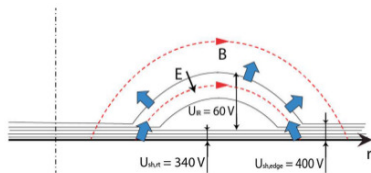
$$\frac{1}{V_D} = a \gamma_{SE} + b$$

- We associate a with hot electrons e^H , sheath acceleration
- We associate b with the Ohmic heating process and cold electrons e^C



dc magnetron sputtering discharge

- The figure shows schematically the magnetic field lines and the electric equipotential surfaces above the racetrack
- A potential V_{SH} falls over the sheath, and the rest of the applied voltage, $V_{IR} = V_D - V_{SH}$, falls across the extended pre-sheath, the ionization region (IR), $\delta_{IR} = V_{IR}/V_D$
- Ohmic heating, the dissipation of locally deposited electric energy $\mathbf{J}_e \cdot \mathbf{E}$ to the electrons in the plasma volume outside the sheath



From Brenning et al. (2016) PSST **25** 065024

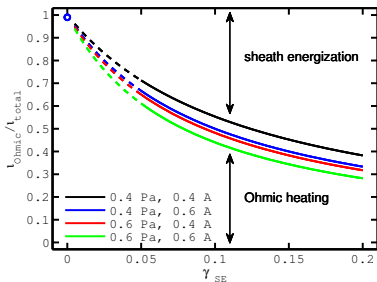
dc magnetron sputtering discharge

I_D (A)	p (Pa)	Slope k	Intercept l	$\delta_{IR} = U_{IR}/U_D$
0.4	0.4	0.0117	0.00145	0.19
0.4	0.6	0.0129	0.00120	0.16
0.6	0.4	0.0130	0.00130	0.17
0.6	0.6	0.0140	0.00110	0.15

- It follows that the fraction of the total ionization that is due to Ohmic heating can be obtained directly from the line fit parameters a and b
- This can be written as a function of only the secondary electron yield

 γ_{SE}

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



From Brenning et al. (2016) PSST 25 063024



dc magnetron sputtering discharge

I_D (A)	p (Pa)	Slope k	Intercept l	$\delta_{IR} = U_{IR}/U_D$
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0.4	0.6	0.0129	0.00120	0.16
0.6	0.4	0.0130	0.00130	0.17
0.6	0.6	0.0140	0.00110	0.15

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

$$\delta_{IR} = \frac{V_{IR}}{V_D}$$

can be estimated from

$$b = \frac{\epsilon_e^C \langle I_e/I_D \rangle_{IR} \delta_{IR}}{\mathcal{E}_C^C}$$

- We assume

$$\epsilon_e^C = 0.8, \quad \langle I_e/I_D \rangle_{IR} \approx 0.5,$$

and

$$\mathcal{E}_C^C = 53.5 \text{ V for } T_e = 3 \text{ V}$$

which gives

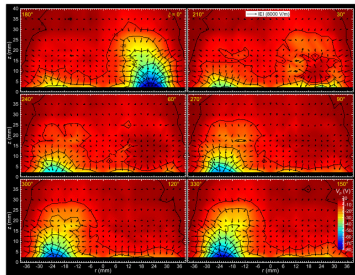
$$\delta_{IR} = 0.15 - 0.19$$

- 15 - 19 % of the applied discharge voltage fall over the ionization region



dc magnetron sputtering discharge

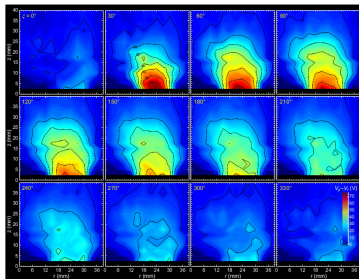
- Recent measurements have revealed strong electric fields parallel and perpendicular to the target of a dc magnetron sputtering discharge
- The largest **E**-fields result from a double layer structure at the leading edge of an ionization zone
- It is suggested that the double layer plays a crucial role in the energization of electrons since electrons can gain several tens of eV when crossing the double layer



From Panjan and Anders (2017) JAP 121 063302

dc magnetron sputtering discharge

- Electrons gain energy when they encounter an electric field – a potential gradient, such as the field in the double layer
- The potential in the double layer jumps by 30 – 70 V ($\delta_{\text{IR}} = 11 - 25 \%$) in the region up to 20 mm over the racetrack area
- The electron heating power $\mathbf{J}_e \cdot \mathbf{E}$ is associated with an acceleration of electrons in the electric field – this electron energization in a double layer is Ohmic heating



From Panjan and Anders (2017) JAP **121** 063302

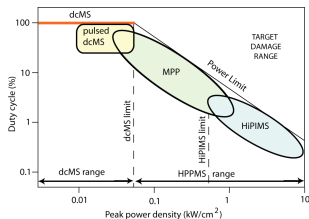
- The distribution of $V_p - V_f \propto \langle E \rangle$ in the $r-z$ plane for a dcMS operated at 270 V and 0.27 Pa

High power impulse magnetron sputtering discharge



High power impulse magnetron sputtering discharge

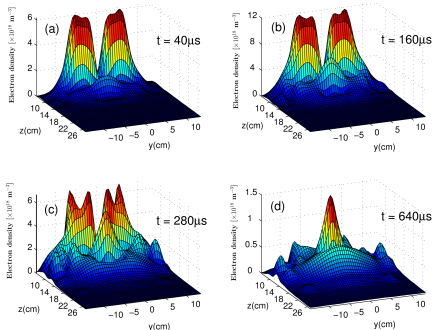
- High ionization of sputtered material requires very high density plasma
- In a conventional dc magnetron sputtering discharge the power density (plasma density) is limited by the thermal load on the target
- High power pulsed magnetron sputtering (HPPMS)
- In a HiPIMS discharge a high power pulse is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



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

- Power density limits
 $\rho_t = 0.05 \text{ kW/cm}^2$ dcMS limit
 $\rho_t = 0.5 \text{ kW/cm}^2$ HiPIMS limit

dc magnetron sputtering discharge



(After Bohlmark et al. (2005), IEEE Trans. Plasma Sci. **33** 346)

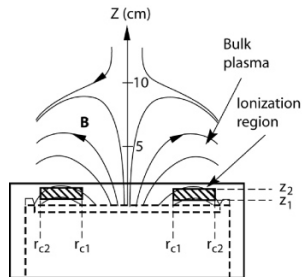
- Temporal and spatial variation of the electron density
- Ar discharge at 20 mTorr, Ti target, pulse length $100 \mu\text{s}$
- The electron density in the substrate vicinity is of the order of $10^{18} - 10^{19} \text{ m}^{-3}$ – ionization mean free path $\lambda_{iz} \sim 1 \text{ cm}$

Ionization region model studies of non-reactive HiPIMS



Ionization region model non-reactive HiPIMS

- The ionization region model (IRM) was developed to improve the understanding of the plasma behaviour during a HiPIMS pulse and the afterglow
- The main feature of the model is that an ionization region (IR) is defined next to the race track
- 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



Ionization region model non-reactive HiPIMS

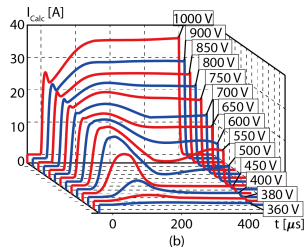
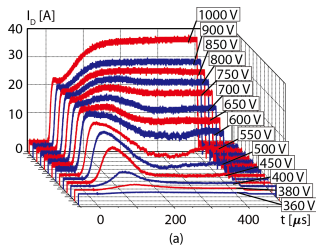
- The temporal development is defined by a set of ordinary differential equations giving the first time derivatives of
 - the electron energy
 - the particle densities for all the particles
- The species assumed in the non-reactive-IRM are
 - cold electrons e^C (Maxwellian), hot electrons e^H (sheath acceleration)
 - argon atoms $Ar(3s^23p^6)$, 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)
 - titanium atoms $Ti(a^3F)$, titanium ions Ti^+ (6.83 eV), doubly ionized titanium ions Ti^{2+} (13.58 eV)
 - aluminium atoms $Al(^2P_{1/2})$, aluminium ions Al^+ (5.99 eV), doubly ionized aluminium ions Al^{2+} (18.8 eV)

Ionization region model non-reactive HiPIMS

- The model is constrained by experimental data input and fitted to reproduce the measured discharge current and voltage curves, $I_D(t)$ and $V_D(t)$, respectively
- Two model fitting parameters were found to be sufficient for a discharge with Al target
 - V_{IR} accounts for the power transfer to the electrons
 - β is the probability of back-attraction of ions to the target

From Huo et al. (2017), JPD submitted 2017

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

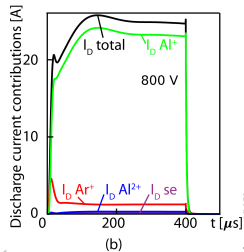
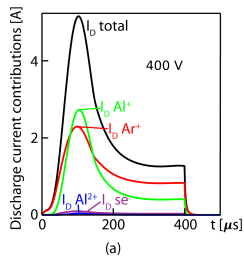


Ionization region model non-reactive HiPIMS

- A **non-reactive** discharge with Al target
- When the discharge is operated at 400 V the contributions of Al^+ and Ar^+ -ions to the discharge current are very similar
- At 800 V Al^+ -ions dominate the discharge current (**self-sputtering**) while the contribution of Ar^+ is below 10 % except at the initiation of the pulse

From Huo et al. (2017), JPD submitted 2017

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

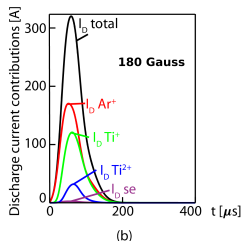
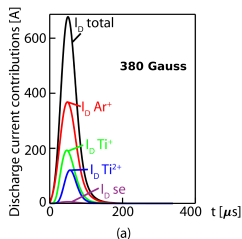


Ionization region model non-reactive HiPIMS

- A **non-reactive** discharge with Ti target
- The contributions to the discharge current for two cases, weak (180 Gauss) and strong (380 Gauss) magnetic field, at 75 Hz pulse frequency
- Stronger magnetic field leads to a higher discharge current
- Higher magnetic field strength leads to higher relative contribution of Ti^{2+} while it lowers the relative contribution of Ti^+

From Huo et al. (2017), JPD submitted 2017

Experimental data from Bradley et al. (2015) JPD **48** 215202



Ionization region model non-reactive HiPIMS

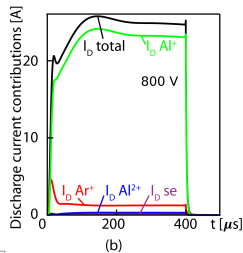
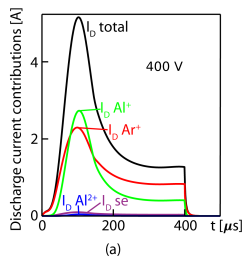
- A primary current I_{prim} is defined as ions of the working gas, here Ar^+ , that are ionized for the first time and then drawn to the target
- This is the dominating current in dc magnetron sputtering discharges
- This current has a critical upper limit

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

- 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

Ionization region model non-reactive HiPIMS

- For the Al target the critical current is $I_{crit} \approx 7 \text{ A}$
- The experiment is operated from far below I_{crit} to high above it, up to 36 A.
- With increasing 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 $I_{SS\text{-}recycle}$ dominates



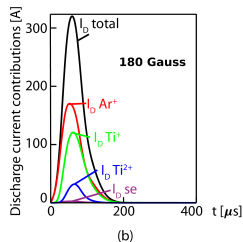
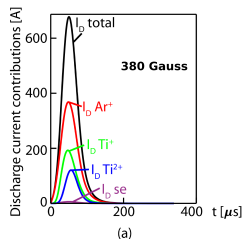
From Huo et al. (2017), JPD submitted 2017

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



Ionization region model non-reactive HiPIMS

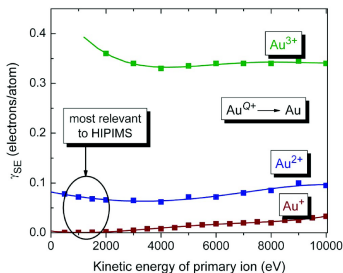
- The discharge with the Ti target is operated with peak current far above the critical current of $I_{crit} \approx 19$ A
- The contribution of Ar^+ and the sum of the contributions of Ti^+ and Ti^{2+} are of similar magnitude
- Higher magnetic field strength enhances the Ti^{2+} -ion density
- This discharge shows close to a 50/50 combination of **self-sputter recycling** $I_{SS-recycle}$ and **working gas-recycling** $I_{gas-recycle}$



From Huo et al. (2017), JPD submitted 2017

Ionization region model non-reactive HiPIMS

- Recall that singly charged metal ions cannot create the secondary electrons – for metal self-sputtering (γ_{SE} is practically zero)
- The first ionization energies of many metals are insufficient to overcome the workfunction of the target material
- For the discharge with Al target operated at high voltage, self-sputter dominated, the effective secondary electron emission is essentially zero



From Anders (2008) APL 92 201501



Ionization region model non-reactive HiPIMS

- The power transfer to the electrons is given by

$$P_e = P_{SH} + P_{Ohm} = I_{e,SH} (V_D - V_{IR}) + \frac{I_D V_{IR}}{2}$$

where

$$P_{SH} = I_{e,SH} V_{SH} = \left(I_{Ar^+} \gamma_{Ar^+,eff} + \frac{1}{2} I_{M^{2+}} \gamma_{M^{2+},eff} \right) V_{SH}$$

and

$$P_{Ohm} = I_{e,IR} V_{IR} = \left\langle \frac{J_e}{J_D} \right\rangle I_D V_{IR}$$

- Then $I_{e,SH} \sim \gamma_{SE} \epsilon_e m (1 - r) I_D \sim 0.05 I_D$ and $I_{e,SH} \ll I_D/2$ so that

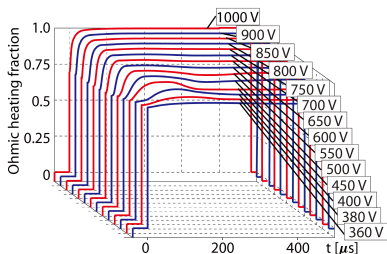
$$I_{e,SH} \ll I_D/2$$

and Ohmic heating is more efficient



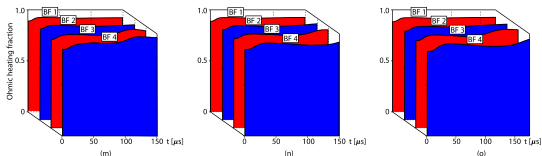
Ionization region model non-reactive HiPIMS

- For the Al target, the fraction of the total electron heating that is attributable to Ohmic heating is found in the range of 0.87 (360 V) to 0.99 (1000 V).
- The domination of Al^+ -ions, which have zero secondary electron emission yield, has the consequence that there is negligible sheath energization
- The ionization threshold for twice ionized Al^{2+} , 18.8 eV, is so high that few such ions are produced



From Huo et al. (2017), JPD submitted 2017

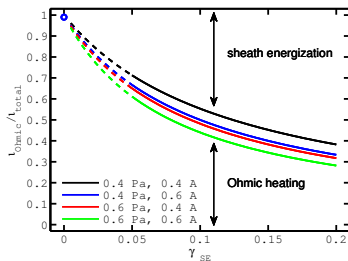
Ionization region model non-reactive HiPIMS



- For the discharge with Ti target more Ar^+ -ions contribute to the current and the ionization degree of Ti^{2+} is more than order of magnitude larger than the ionization degree of Al^{2+} , so there are more secondary electrons
- The fraction of the total electron heating that is attributable to Ohmic heating is about 0.92
- Decreasing the magnetic field strength (BF1 to BF4) slightly reduces the Ohmic heating fraction

Ionization region model non-reactive HiPIMS

- The relative contributions to the total ionization ι_{total} due to Ohmic heating, ι_{Ohmic} , and sheath energization, ι_{sheath}
- A blue circle marks the HiPIMS study modelled by Huo et al. (2013)
- It is taken at the end of a 400 μs long pulse when the discharge was deep into the self-sputtering mode
- A large fraction of Al^+ ions here gives $\gamma_{\text{SE,eff}}$ close to zero
- Note that this HiPIMS case $\gamma_{\text{SE,eff}}$ is consistent with the dcMS cases



From Brenning et al. (2016) PSST 25 065024



Ionization region model non-reactive HiPIMS

- The model results show that for an argon discharge with Al target the contribution of Al^+ -ions is over 90 % at 800 V, while Al^+ -ions and Ar^+ -ions contribute roughly equally to the discharge current at 400 V
- For high currents the discharge with Al 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 Ti target, a self-sputter yield significantly below unity makes working gas-recycling necessary at high currents.
- The model results show that Al^{2+} -ions contribute negligibly, while Ti^{2+} -ions effectively contribute to the production of secondary electrons
- The fraction of the total electron heating that is attributable to Ohmic heating is over 90 %



Summary



Summary

- It has been demonstrated that Ohmic heating of the electrons can play a significant role in conventional dc magnetron sputtering discharges
- We used a ionization region model to explore the plasma composition and the electron heating mechanism in a high power impulse magnetron sputtering (HiPIMS) discharge
 - For high currents the discharge with Al 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**
 - The fraction of the total electron heating that is attributable to Ohmic heating is over 90 % in the HiPIMS discharge



Thank you for your attention

The slides can be downloaded at

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

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