

# On ion recycling and electron heating in high power impulse magnetron sputtering discharges

J. T. Guðmundsson<sup>1,2</sup>

<sup>1</sup>Department of Space and Plasma Physics,  
KTH Royal Institute of Technology, Stockholm, Sweden

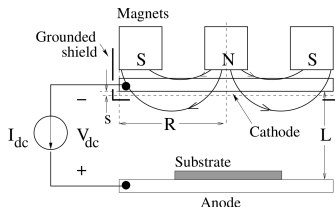
<sup>2</sup> Science Institute, University of Iceland, Reykjavik, Iceland

Faculty of Science, Masaryk University,  
Brno, Czech Republic  
June 12, 2019



# Introduction

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

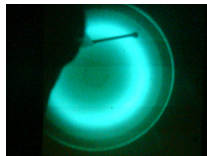
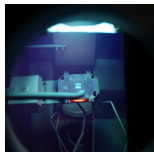
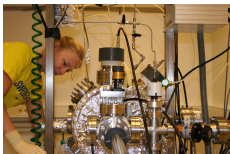


- A magnet is placed at the back of the cathode target with the pole pieces at the center and perimeter
- The magnetic field confines the energetic electrons near the cathode
- The electrons undergo numerous ionizing collisions before being lost to a grounded surface

# The dc magnetron sputtering discharge



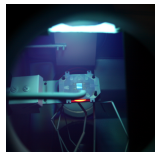
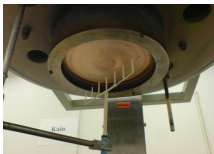
## *The dc magnetron sputtering discharge*



- Magnetron sputtering has been the workhorse of plasma based sputtering methods for over four decades
- Through the years there has been a continuous development of the magnetron sputtering processes to
  - increase the ionization of the sputtered vapor
  - improve target utilization
  - avoid target poisoning in reactive sputtering
  - increase deposition rates

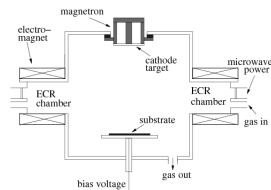
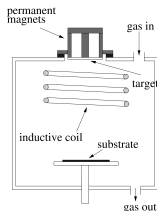
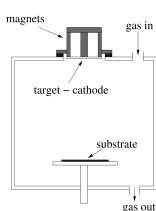


## *The dc magnetron sputtering discharge*



- For many applications a high degree of ionization of the sputtered vapor is desired
  - controlled ion bombardment of the growing film
  - ion energy can be – controlled by a negative bias applied to the substrate
  - collimation – enhanced step coverage
- Ionized flux of the sputtered material introduces an additional control parameter into the deposition process

# The dc magnetron sputtering discharge



From Gudmundsson (2008), J. Phys.: Conf. Ser. **100** 082002

- In magnetron sputtering discharges increased ionized flux fraction is achieved by
  - a secondary discharge between the target and the substrate (rf coil or microwaves)
  - reshaping the geometry of the cathode to get more focused plasma (hollow cathode)
  - increasing the power to the cathode (high power pulse)
- Common to all highly ionized magnetron sputtering techniques is a very high density plasma

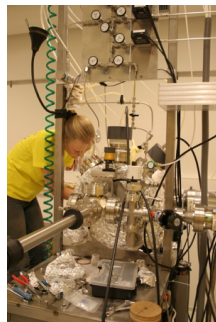


# The high power impulse magnetron sputtering discharge



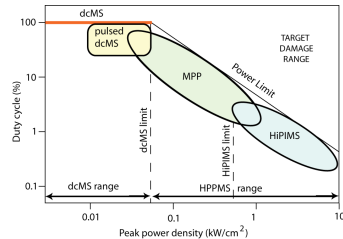
# *High power impulse magnetron sputtering discharge*

- In a conventional dc magnetron discharge the power density is limited by the thermal load on the target
- Most of the ion bombarding energy is transformed into heat at the target
- In a HiPIMS discharge a high power pulse is supplied for a short period
  - low frequency
  - low duty cycle
  - low average power
- The high power pulsed magnetron sputtering discharge uses the same sputtering apparatus except the power supply



# High power impulse magnetron sputtering discharge

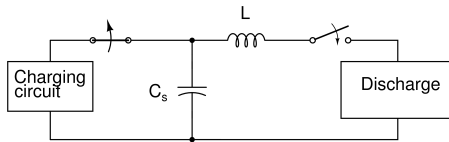
- In dc magnetron sputtering the power density (plasma density) is limited by the thermal load on the target
- High power pulsed magnetron sputtering (HPPMS)
- High power impulse magnetron sputtering (HiPIMS)
  - a pulse of very high amplitude, an impulse, is applied to the cathode and a long pause exists between the pulses
- Modulated pulse power (MPP)
  - the initial stages of the pulse (few hundred  $\mu\text{s}$ ) the power level is moderate (typical for a dcMS) followed by a high power pulse (few hundred  $\mu\text{s}$  up to a ms)



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

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

# High power impulse magnetron sputtering discharge



- The high power impulse magnetron sputtering (HiPIMS) discharge operates with a
  - Cathode voltage in the range of 500 – 2000 V
  - Current densities of 0.5 – 4 A/cm<sup>2</sup>
  - Power densities in the range of 0.5 – 3 kW/cm<sup>2</sup>
  - Average power 200 – 600 W
  - Frequency in the range of 50 – 5000 Hz
  - Duty cycle in the range of 0.5 – 5 %

# High power impulse magnetron sputtering discharge

- The development of high power pulse magnetron sputtering is traced to the Moscow Engineering and Physics Institute (MEPhI)
- The first published record on what we now refer to as a pre-ionized HiPIMS discharge was presented at the XX International Conference on Phenomena in Ionized Gases (ICPIG) Braga , Italy, July 1991 by the group at MEPhI

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QUASI-STATIONARY HIGH CURRENT FORMS OF LOW PRESSURE DISCHARGE IN MAGNETIC FIELD

Petelin I.K., Khodachenko G.V., Mezgrin D.V.

Department of Plasma Physics, Faculty of Experimental and Theoretical Physics, Moscow Physics Engineering Institute, 10-115489, Moscow, USSR

High-current forms of discharge, which permit to create large volume of high density homogeneous plasma, are widely used in modern technique. These discharges are used as plasma sources of charged particles, and also in various technologies for sputtering, implantation and etching of materials [1].

The possibility of creation of some forms of quasi-stationary discharges in a quadrupole magnetic system, and planar magnetron is described in this paper. The discharge units are shown in fig. 1. The

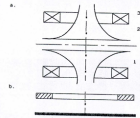


Figure 1. Discharge units. a.-unit with quadrupole magnet; b.-unit with permanent magnet. 1-cathode; 2-planar field coils (permanent magnet).

discharge was created between electrodes 1, 2 in magnetic field, created by either coils or permanent magnet 3. The shape of hollow electrode coincide with that of magnetic field line of force. The shape of hollow electrode provides an orthogonality of Eos field near the cathode surface. This configuration allowed to combine the high-current magnetron discharge with the hollow cathode discharge.

Preliminary. A discharge with current of 0.3 A or less had been initiated between electrodes 1, 2 with stationary power source. Plasma density  $n$  of  $10^{18}-10^{19} \text{ m}^{-3}$  and electron temperature of 3-15 eV was obtained in this discharge, that provided sufficient previous ionization of discharge volume. Afterwards the rectangular current pulse with front of 1-10 ns and plateau of 1-3 ns and amplitude up to 200 A (fig. 2) was superimposed on the

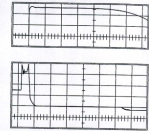


Figure 2. Discharge current (a) and voltage (b) oscillogram. a.-500 A/div; 0.2 ns/div; b.-150 V/div; 0.2 ns/div. Special preliminary discharge characteristics were measured using Rogowski coil-voltage dividers and digital oscilloscope. The pressure was between 10 and 1 Torr. Magnetic field induction was up to 180.

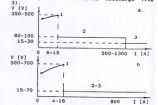


Figure 3. Total discharge characteristics: a.-in quadrupole unit; b.-in the planar magnetron. + - preliminary discharge ( $I=0.05-0.1 \text{ A}$ ). The first form seems to correspond to high-current magnetron discharges with current density up to 0.5 A/cm<sup>2</sup> at higher current (4-18) A the discharge transforms to the second regime with voltage of 100 V and

# High power impulse magnetron sputtering discharge

- The PhD thesis of Dimitry Mozgrin describes a high-current low-pressure quasistationary discharge in a magnetic field
- It was demonstrated for two configurations
  - a planar magnetron device
  - two hollow axisymmetric electrodes immersed in a cusp-shaped magnetic field
- For the planar magnetron device, they reported a peak power of 200 kW (200 A) onto a 120 mm diameter target giving peak a power density of  $1.8 \text{ kW/cm}^2$  and discharge current densities of up to  $25 \text{ A/cm}^2$  at a repetition rate of 10 Hz in a pre-ionized discharge

61-95-1/593-2

МОСКОВСКИЙ ГОСУДАРСТВЕННЫЙ ИНЖЕНЕРНО-ФИЗИЧЕСКИЙ ИНСТИТУТ  
(ТЕХНИЧЕСКИЙ УНИВЕРСИТЕТ)

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МОЗГРИН ДМИТРИЙ ПЕТРОВИЧ

Экспериментальные исследования квазистационарных форм  
напряжённости электрического поля  
в магнетронном поле

01.04.08 - Физика в экстремальных условиях

Диссертация  
на соискание ученой степени  
кандидата физико-математических наук

Автор *Mozgrin*

Научный руководитель -  
доктор физико-математических наук  
профессор Сергей Константинович

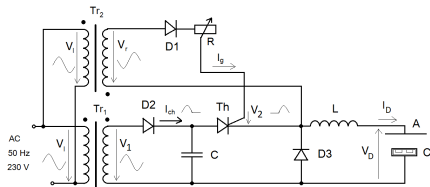
Москва - 1994 г.





## High power impulse magnetron sputtering discharge

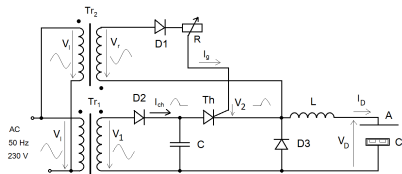
- The original concept of a HiPIMS power supply, which was based on thyristor switches and can deliver extremely high currents during the pulse
- The capacitor  $C$  had a value around  $10 - 20 \mu\text{F}$
- There are two transformers  $\text{Tr}_1$  and  $\text{Tr}_2$  working with the line frequency  $50 - 60 \text{ Hz}$
- This type of HiPIMS power supply is capable of delivering pulse powers of up to  $P_p \approx 1 \text{ MW}$



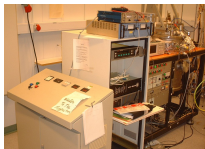
Based on Kouznetsov (2001) U.S. Patent no. 6,296,742 B2

## High power impulse magnetron sputtering discharge

- In this construction it is difficult to control the length of the active pulse, it is given by the time constant of the plasma impedance and the values of  $C$  and  $L$
- Furthermore, the pulse repetition frequency is fixed by the frequency of the ac line supply
- This type of power supply was used in the early demonstration of the HiPIMS technique performed at Linköping University

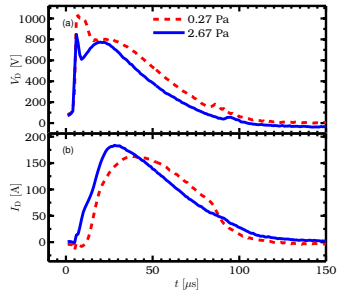


Based on Kouznetsov (2001) U.S. Patent no. 6,296,742



## High power impulse magnetron sputtering discharge

- The discharge voltage  $V_D$  and current  $I_D$  for an argon discharge at 0.27 and 2.67 Pa with 150 mm diameter tantalum target driven by an early thyristor-based power supply
- We see an initial voltage peak in the kilovolt range which is followed by a drop in the voltage to several hundred volts
- The voltage then drops even further to values that are typical operating voltages for a dcMS discharge
- As the voltage drops, the discharge current increases up to a peak value followed by a gradual decay of the current

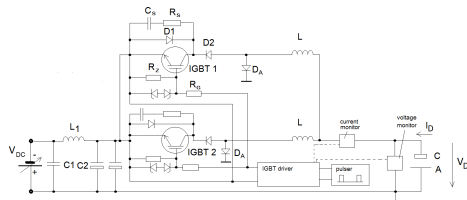


From Gudmundsson et al. (2002) SCT 161 249



# High power impulse magnetron sputtering discharge

- Modern concepts of pulsed power supplies utilize insulated-gate bipolar transistors (IGBTs) as high power switches
- Also, the previously used small capacitor  $C$  is substituted by a large capacitor bank composed of low-impedance electrolytic capacitors
- A typical circuit diagram of a HiPIMS power supply based on IGBT switches

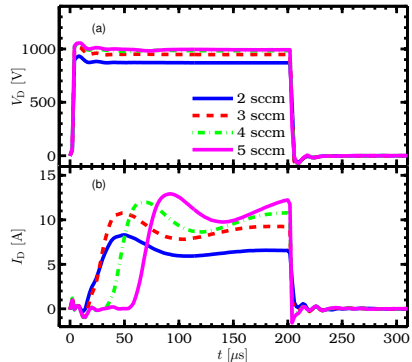


Hubička et al. (2019) in High Power Impulse Magnetron

Sputtering Discharge, Elsevier, 2019

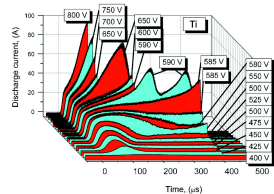
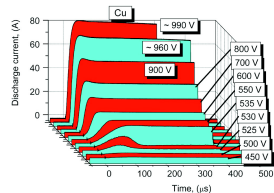
# High power impulse magnetron sputtering discharge

- The discharge voltage  $V_D$  and current  $I_D$  for an argon discharge mixed with nitrogen at different flow rates and vanadium target
- The discharge is driven by an IGBT based power supply with a large capacitor bank
- The total gas pressure is 0.9 Pa, the argon flow rate is 40 sccm, the voltage pulse is 200  $\mu$ s long and the pulse frequency is 100 Hz.



# High power impulse magnetron sputtering discharge

- In **non-reactive** discharge the current waveform shows an initial pressure dependent peak that is followed by a second phase that is power and material dependent
- The initial phase is dominated by working gas ions, whereas the later phase has a strong contribution from self-sputtering
- For some materials, the discharge switches into a mode of **sustained self-sputtering**



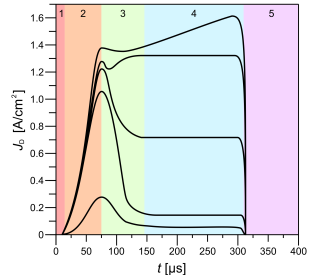
From Anders et al. (2007),

JAP 102 113303 and JAP 103 039901



# High power impulse magnetron sputtering discharge

- The initial phase has a contribution from the working gas ions, whereas the later phase has a strong contribution from self-sputtering at high voltage
- The current is generally characterized by an initial peak followed by a more or less stable current plateau (bottom current curves)
- In other cases it shows an initial peak followed by a second increase of the discharge current





## *High power impulse magnetron sputtering discharge*

- The self-sputtering can operate in a self-sustained mode, when the ions of the sputtered vapor are created at high enough rate that the ions of the working gas are not needed
- The condition for sustained self-sputtering is expressed as

$$\Pi_{ss} = \alpha \beta_t Y_{ss} = 1$$

where

- $\alpha$  is the probability of ionization of the sputtered atom
- $\beta_t$  is the probability that the newly formed ion of the sputtered vapor returns to the target
- $Y_{ss}$  is the self-sputter yield of the ion
- This is a steady state situation and the current remains constant



## *High power impulse magnetron sputtering discharge*

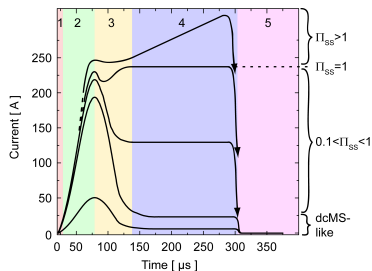
- Note that since  $\alpha < 1$  and  $\beta_t < 1$  the condition  $Y_{ss} > 1$  is necessary but not sufficient for achieving sustained self-sputtering
- The transient phase of self-sputtering runaway occurs when  $\Pi_{ss} > 1$
- Self-sputtering runaway occurs at a well-defined threshold power, determined by the discharge voltage and is readily obtained for high sputter yield materials
- But runaway can also occur at lower threshold voltages than for pure self-sputtering as well as for transition metals and target materials of low sputter yield due to what is referred to as 'gas recycling' runaway

Anders (2011), SCT 205 S1, Anders et al. (2012) JPD 45 012003



# High power impulse magnetron sputtering discharge

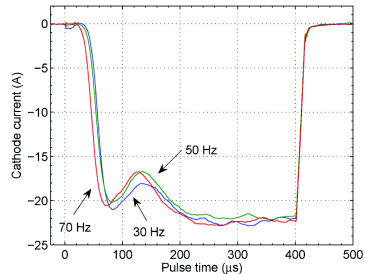
- The bottom curve represents a range of low self-sputtering,  $\Pi_{ss} < 0.1$  and the discharge physics in the plateau/runaway phase is dcMS-like
- The middle range of power densities, with  $0.1 < \Pi_{ss} < 1$ , represents partially self-sputtering discharge
- The top curve represents self-sputtering runaway which requires  $\Pi_{ss} > 1$  and a self-sputter yield  $Y_{ss} > 1/(\alpha\beta_t) > 1$



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

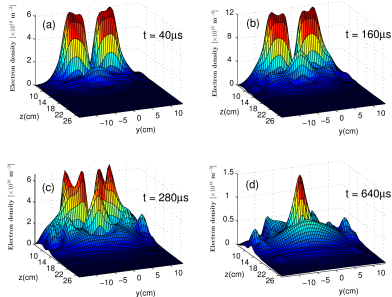
# High power impulse magnetron sputtering discharge

- Ar discharge with Ti target
- The initial peak in current results large flux of atoms from the target
- Collisions of the sputtered atoms with the working gas result in heating and expansion of the working gas – **rarefaction**
- A significant fraction of the sputtered atoms experience electron impact ionization (the ionization mean free path  $\sim 1$  cm) and are attracted back to the target to participate in the sputtering process – **self-sputtering**



From Magnus et al. (2011) JAP **110** 083306

# High power impulse magnetron sputtering discharge

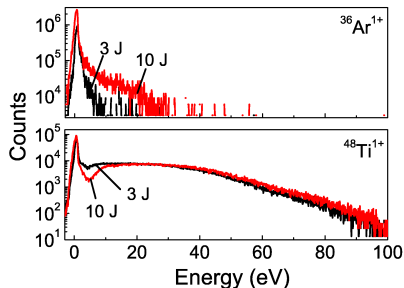


(After Bohlmark et al. (2005), IEEE Trans. Plasma Sci. **33** 346)

- 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  $\lambda_{iz} \sim 1 \text{ cm}$

# High power impulse magnetron sputtering discharge

- The time averaged ion energy distribution for  $\text{Ar}^+$  and  $\text{Ti}^+$  ions
- The working gas pressure was 3 mTorr, pulse energy 3 J and 10 J and the target made of Ti
- The ion energy distribution is broad to over 100 eV
- About 50 % of the  $\text{Ti}^+$  ions have energy  $> 20$  eV



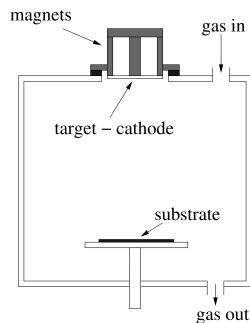
From Bohlmark et al. (2006) TSF 515 1522

# Electron power absorption in magnetron sputtering discharges



# Introduction

- The conventional wisdom is that plasma generation in magnetron sputtering discharges is based on the supply of energy via secondary electrons (SEs) accelerated from the target
- However, one of the remaining fundamental questions is **how power is absorbed by the electrons in the magnetron sputtering discharge**

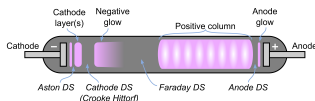




# dc magnetron sputtering discharge



## *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  $\gamma_{SE}$  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

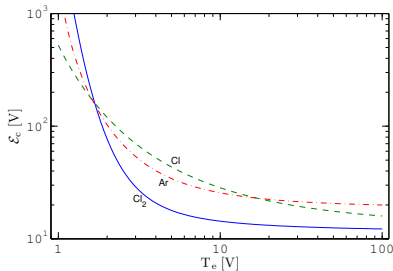


## *dc magnetron sputtering discharge*

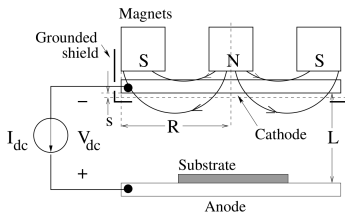
- These secondary electrons are accelerated in the cathode dark space – referred to as primary electrons
- They must produce sufficient number of ions to release more electrons from the cathode
- The number of electron-ion pairs created by each secondary electron is then

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

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

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

- $\epsilon_e$  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

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

has to be fulfilled

- This defines the minimum voltage to sustain the discharge as

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

referred to as Thornton equation

- $\beta$  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  $m$ ,  $\beta$ ,  $\epsilon_e$  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.<sup>72</sup> This requirement can be expressed by the following relationship for the minimum potential to sustain such a discharge:<sup>73</sup>

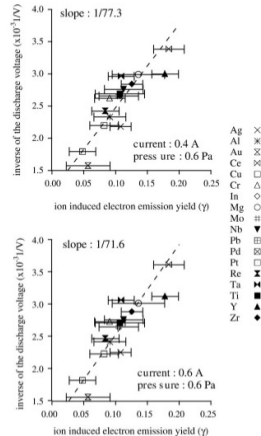
$$V_{\min} = \mathcal{E}_0 / \Gamma_i \epsilon_i \epsilon_e \quad (5)$$

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

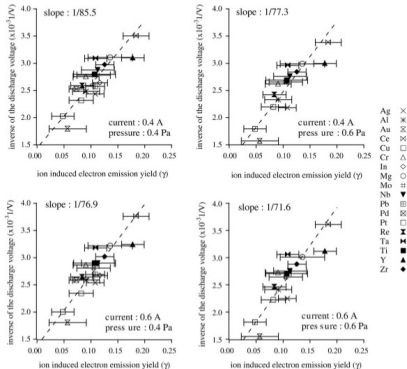


## *dc magnetron sputtering discharge*

- A plot of the inverse discharge voltage  $1/V_D$  against  $\gamma_{SE}$  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  $m$ ,  $\beta$ ,  $\epsilon_e$  and  $\mathcal{E}_c$  are independent of  $\gamma_{SE}$



# dc magnetron sputtering discharge



From Depla et al. (2009) TSF 517 2825

- $1/V_D$  against  $\gamma_{SE}$  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

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

or

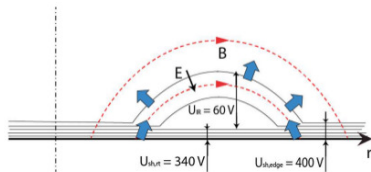
$$\frac{1}{V_D} = a \gamma_{SE} + b$$

- 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



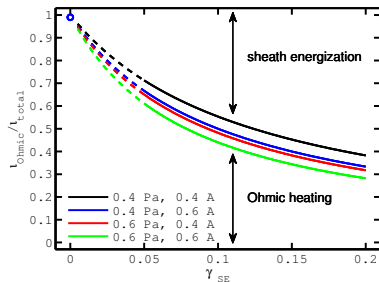
From Brenning et al. (2016) PSST **25** 065024

# 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  $\gamma_{SE}$

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



From Brenning et al. (2016) PSST 25 06:0024

## *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

- 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

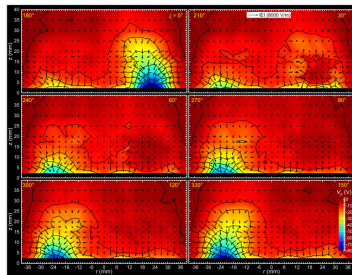
$$\delta_{IR} = 0.15 - 0.19$$

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



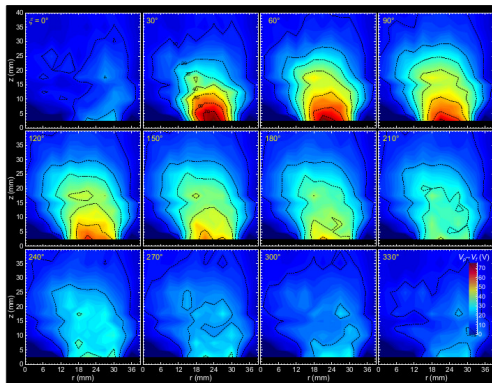
## *dc magnetron sputtering discharge*

- 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*

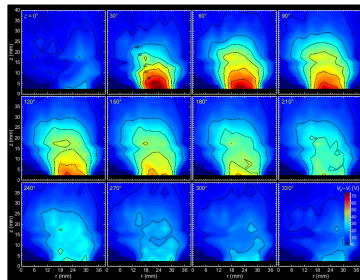


From Panjan and Anders (2017) JAP 121 063302

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

## *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 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

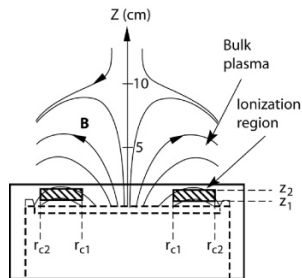
# Ionization region model studies of HiPIMS discharges





## *Ionization region model of 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 of HiPIMS*

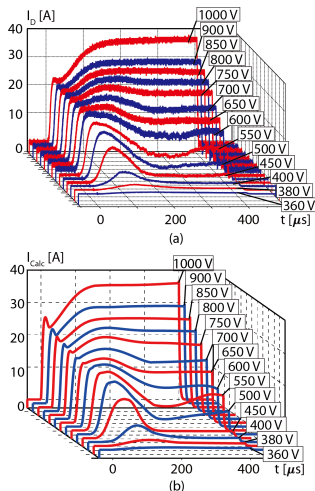
- The temporal development is defined by a set of ordinary differential equations giving the first time derivatives of
  - the electron energy
  - the particle densities for all the particles
- The species assumed in the of-IRM are
  - cold electrons  $e^C$  (Maxwellian), hot electrons  $e^H$  (sheath 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 of HiPIMS***

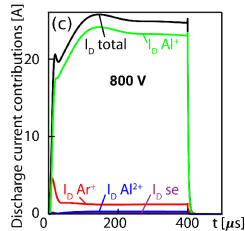
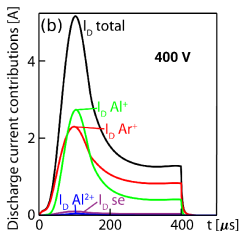
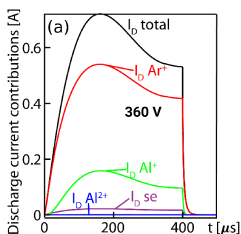
- The model is constrained by experimental data input and fitted to reproduce the measured discharge current and voltage curves,  $I_D(t)$  and  $V_D(t)$ , respectively
- Two model fitting parameters were found to be sufficient for a discharge with Al target
  - $V_{IR}$  accounts for the power transfer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

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

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



# ***Ionization region model of HiPIMS***



- A **non-reactive** discharge with 50 mm diameter Al target
- Current composition at the target surface

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

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

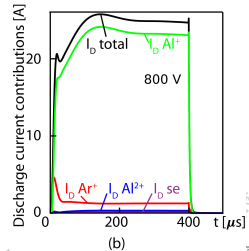
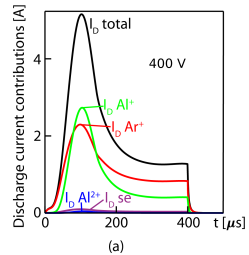


## ***Ionization region model of HiPIMS***

- When the discharge is operated at 400 V the contributions of  $\text{Al}^+$  and  $\text{Ar}^+$ -ions to the discharge current are very similar
- At 800 V  $\text{Al}^+$ -ions dominate the discharge current (**self-sputtering**) while the contribution of  $\text{Ar}^+$  is below 10 % except at the initiation of the pulse

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

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



## *Ionization region model of HiPIMS*

- A primary current  $I_{\text{prim}}$  is defined as ions of the working gas, here  $\text{Ar}^+$ , 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

$$I_{\text{crit}} = S_{\text{RT}} e p_g \sqrt{\frac{1}{2\pi m_g k_B T_g}} = S_{\text{RT}} e n_g \sqrt{\frac{k_B T_g}{2\pi m_g}}$$

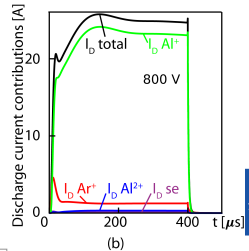
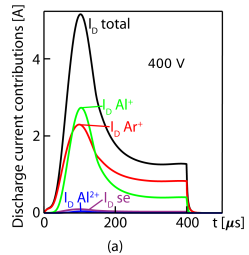
- Discharge currents  $I_D$  above  $I_{\text{crit}}$  are only possible if there is some kind of recycling of atoms that leave the target, become subsequently ionized and then are drawn back to the target

# ***Ionization region model of HiPIMS***

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

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

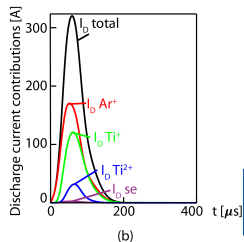
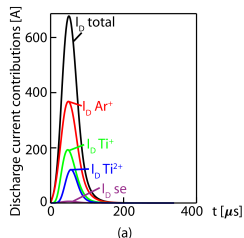
Experimental data from Anders et al. (2007) JAP **102** 113303



## ***Ionization region model of HiPIMS***

- For discharges with Ti target the peak current is far above the critical current (up to 650 A, while  $I_{\text{crit}} \approx 19$  A)
- However, this discharge shows close to a 50/50 combination of **self-sputter recycling**  $I_{\text{SS-recycle}}$  and **working gas-recycling**  $I_{\text{gas-recycle}}$
- Almost 2/3 of the current to the target is here carried by  $\text{Ar}^+$  and  $\text{Ti}^{2+}$ -ions, which both can emit secondary electrons upon target bombardment, and this gives a significant sheath energization

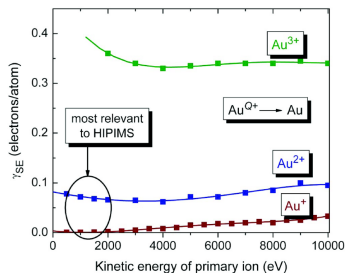
From Huo et al. (2017) JPD **50** 354003





## *Ionization region model of HiPIMS*

- Recall that singly charged metal ions cannot create the secondary electrons – for metal self-sputtering ( $\gamma_{SE}$  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

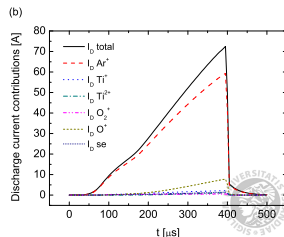
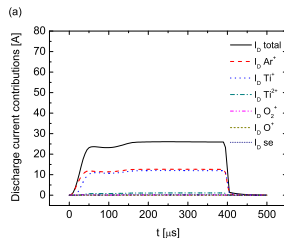


From Anders (2008) APL **92** 201501



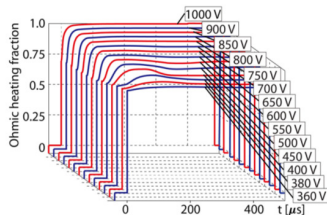
# ***Ionization region model of HiPIMS***

- **Reactive HiPIMS**
- Ar/O<sub>2</sub> discharge with Ti target
- For this system  $I_{\text{crit}} \approx 5$  A
- In the metal mode Ar<sup>+</sup> and Ti<sup>+</sup>-ions contribute roughly equally to the current – combined **self-sputter recycling** and **working gas recycling**
- In the poisoned mode the current increases and Ar<sup>+</sup>-ions dominate the current – **working gas recycling**



## *Ionization region model of HiPIMS*

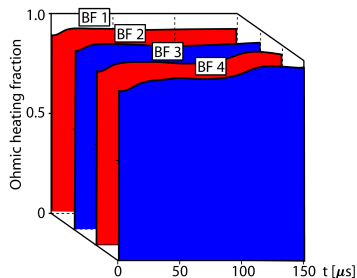
- For the Al target, Ohmic heating is in the range of 87 % (360 V) to 99 % (1000 V)
- The domination of  $\text{Al}^+$ -ions, which have zero secondary electron emission yield, has the consequence that there is negligible sheath energization
- The ionization threshold for twice ionized  $\text{Al}^{2+}$ , 18.8 eV, is so high that few such ions are produced



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

## *Ionization region model of HiPIMS*

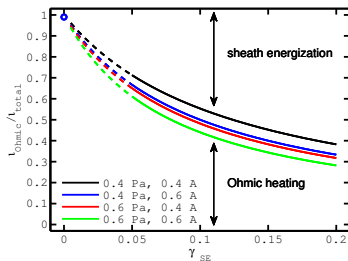
- For a Ti target Ohmic heating is about 92 %
  - Both  $\text{Ar}^+$  and  $\text{Ti}^{2+}$ -ions contribute to creation of secondary electrons
- For Ti target in  $\text{Ar}/\text{O}_2$  mixture
  - In the metal mode Ohmic heating is found to be 90 % during the plateau phase of the discharge pulse
  - For the poisoned mode Ohmic heating is 70 % with a decreasing trend, at the end of the pulse



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

# *Ionization region model of HiPIMS*

- Ohmic heating is also very significant in dc magnetron sputtering discharges
- The relative contributions to the total ionization  $\iota_{\text{total}}$  due to Ohmic heating,  $\iota_{\text{Ohmic}}$ , and sheath energization,  $\iota_{\text{sheath}}$
- A blue circle marks the HiPIMS study modelled by Huo et al. (2013)
- Note that this HiPIMS case  $\gamma_{\text{SE,eff}}$  is consistent with the dcMS cases



From Brenning et al. (2016) PSST 25 065024

# The generalized recycling model



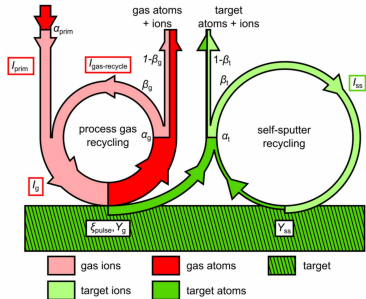
# Generalized recycling

- A working gas-sputtering parameter

$$\pi_g = \alpha_g \beta_g \xi_{\text{pulse}}$$

where

- $\alpha_g$  is ionization probability
- $\beta_g$  is back attraction probability
- $\xi_{\text{pulse}} = 1$  is return fraction in a pulse
- The total current carried by working gas ions



$$I_g = I_{\text{prim}} + I_{\text{gas-recycle}} = I_{\text{prim}} \left( 1 + \frac{\pi_g}{1 - \pi_g} \right)$$

From Brenning et al. (2017) PSST 26 125003

# Generalized recycling

- The total self-sputter current is

$$I_{SS} = I_g \left( \frac{Y_g}{Y_{SS}} \frac{\pi_{SS}}{1 - \pi_{SS}} \right)$$

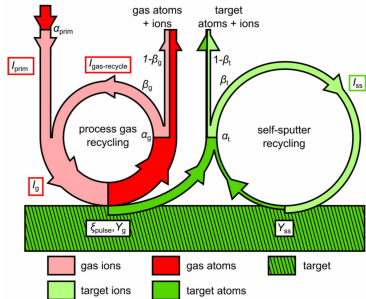
where the self-sputter parameter is

$$\pi_{SS} = \alpha_t \beta_t Y_{SS}$$

- The total discharge current is

$$I_D = I_{prim} + I_{gas-recycle} + I_{SS}$$

$$= I_{prim} \left( 1 + \frac{\pi_g}{1 - \pi_g} \right) \left( 1 + \frac{Y_g}{Y_{SS}} \frac{\pi_{SS}}{1 - \pi_{SS}} \right)$$



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



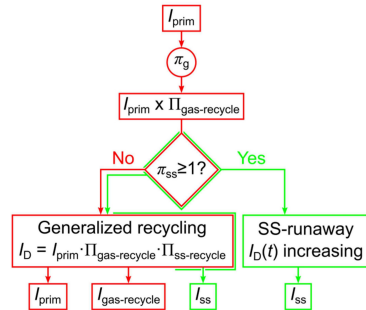


# Generalized recycling

- The discharge current

$$I_D = I_{\text{prim}} \Pi_{\text{gas-recycle}} \Pi_{\text{SS-recycle}}$$

- $I_{\text{prim}}$  is the seed current that acts as a seed to the whole discharge current and has an upper limit  $I_{\text{crit}}$
- $I_{\text{prim}} \Pi_{\text{gas-recycle}}$  is the seed current for the self-sputter process
- If  $\pi_{\text{SS}} > 1$  the discharge goes into SS-runaway

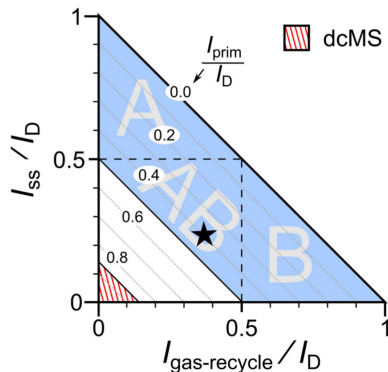


From Brenning et al. (2017) PSST 26 125003



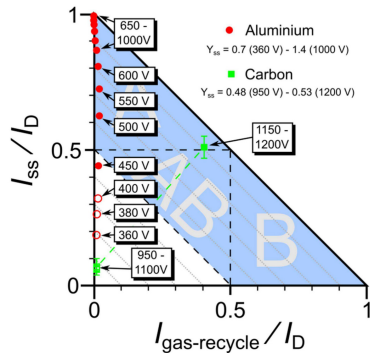
# Generalized recycling

- Recycling map
- A graph in which the ion current mix of  $I_{\text{prim}}$ ,  $I_{\text{gas-recycle}}$ , and  $I_{\text{SS}}$  to the target in a magnetron discharge is defined by a point
- The value of  $I_{\text{prim}}/I_D = 39\%$ , can be read on the diagonal lines ( $Y_{\text{SS}} = 0.5$ )
- $I_{\text{prim}}/I_D \geq 0.85$  defines the dcMS regime
- For  $I_{\text{SS}}/I_D > 0.5$  we have the SS-recycle dominated range A
- For  $I_{\text{gas-recycle}}/I_D > 0.5$  we have the gas-recycle dominated range B



## Generalized recycling

- The discharge with Al target moves from the dcMS regime to the HiPIMS discharge regime with increased discharge voltage – **type A**
- A discharge with carbon target jumps from the dcMS regime to the HiPIMS regime – both SS recycling and working gas recycling play a role – intermediate **type AB**

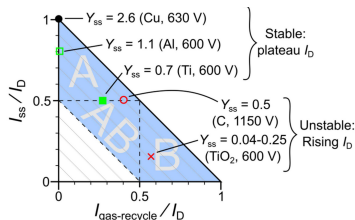


# Generalized recycling

- 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 –  $Y_{SS} = 0.5$
  - TiO<sub>2</sub> –  $Y_{SS} = 0.04 - 0.25$

- 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**

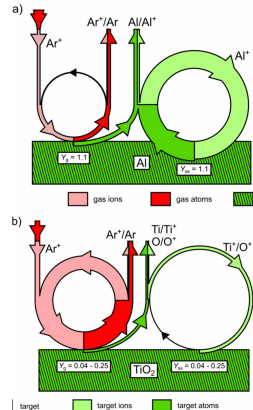


From Brenning et al. (2017),

PSST 26 125003

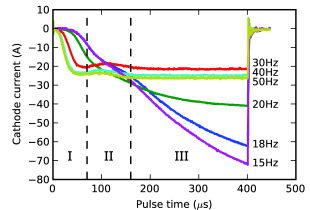
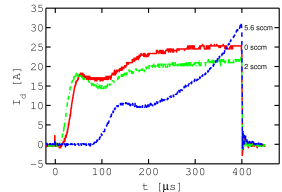
## Generalized recycling

- Recycling loops
- Discharge with Al target – SS recycling dominates
  - high self sputter yield
- Reactive discharge with  $\text{TiO}_2$  target – working gas recycling dominates
  - low self sputter yield



## HiPIMS - Voltage - Current - time

- For Ar/O<sub>2</sub> discharge with Ti target
- At high frequencies, oxide is not able to form between pulses, and **self-sputtering recycling** by Ti<sup>+</sup>-ions is the dominant process
- At low frequency, the long off-time results in an oxide layer being formed (TiO<sub>2</sub>) on the target surface and **working gas recycling dominates** – triangular current waveform



# 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





## Summary

- For very high self-sputter yields, above approximately  $Y_{SS} \approx 1$ , the discharges above  $I_{crit}$  are of type A with
  - dominating SS-recycling
  - very little secondary electron emission
  - little sheath energization of electrons
- For very low self-sputter yields, below approximately  $Y_{SS} \approx 0.2$ , the discharges above  $I_{crit}$  are of type B with
  - dominating working gas recycling
  - significant secondary electron emission
  - significant sheath energization of electrons.

## Thank you for your attention

The slides can be downloaded at

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

The work is in collaboration with

- Dr. Daniel Lundin, Université Paris-Sud, Orsay, France
- Prof. Nils Brenning, KTH Royal Institute of Technology, Stockholm, Sweden
- Dr. Michael A. Raadu, KTH Royal Institute of Technology, Stockholm, Sweden
- Prof. Tiberu Minea, Université Paris-Sud, Orsay, France

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