

# The current waveform in reactive high power impulse magnetron sputtering

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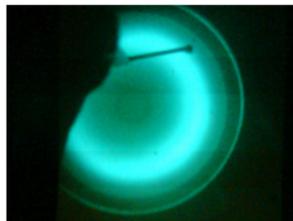
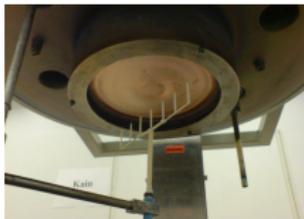
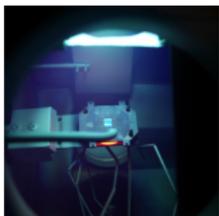
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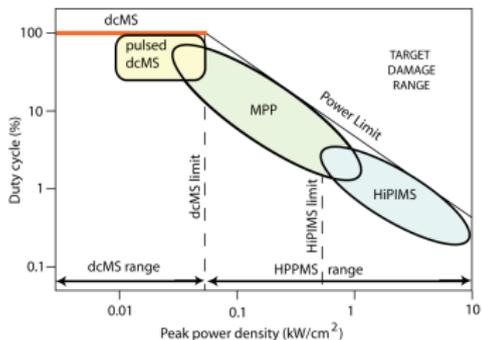
## Introduction



- Magnetron sputtering has been the workhorse of plasma based sputtering methods for over four decades
- For many applications a high degree of ionization of the sputtered vapor is desired
  - controlled ion bombardment of the growing film – controlled by a negative bias applied to the substrate
  - collimation – enhanced step coverage
- Common to all highly ionized magnetron sputtering techniques is a very high density plasma

# Introduction

- In a conventional dc magnetron 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



From 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

## Introduction

- Reactive sputtering, where metal targets are sputtered in a reactive gas atmosphere to deposit compound materials is of utmost importance in various technologies
- In reactive sputtering processes a reactive gas  $O_2$ ,  $N_2$ , or  $CH_4$  etc. is mixed to the noble working gas for oxide, nitride, or carbide deposition
- HiPIMS deposition generally gives denser, smoother films og higher crystallinity than dcMS grown films



Helmersson et al. (2006) Thin Solid Films **513** 1

Magnus et al. (2012) IEEE EDL **33** 1045

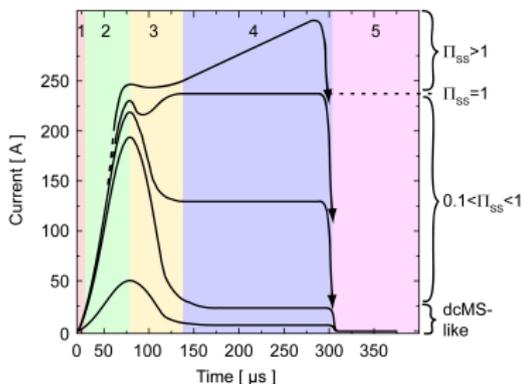


# HiPIMS - Voltage - Current - Time characteristics



## HiPIMS - Voltage - Current - time

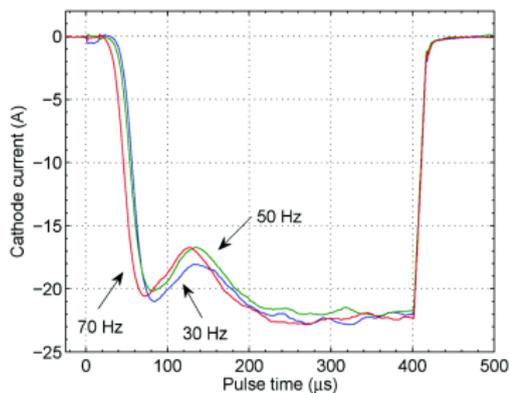
- In **non-reactive** discharge the current waveform shows an initial pressure dependent peak that is followed by a second phase that is power and material dependent
- The initial phase is dominated by gas ions, whereas the later phase has a strong contribution from self-sputtering
- The non-reactive case is well understood



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

## HiPIMS - Voltage - Current - time

- 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**

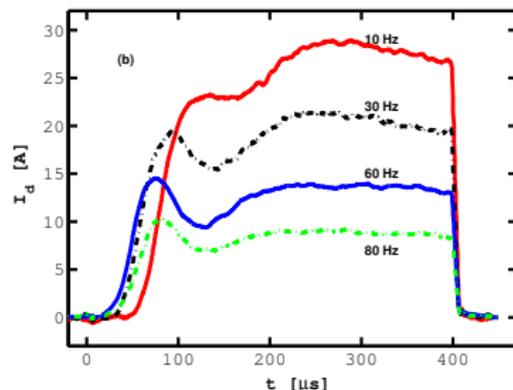


From Magnus et al. (2011) JAP **110** 083306



## HiPIMS - Voltage - Current - time

- During reactive sputtering, a reactive gas is added to the inert working gas
- The current waveform in the reactive Ar/N<sub>2</sub> HiPIMS discharge with Ti target is highly dependent on the pulse repetition frequency
- N<sub>2</sub> addition changes the plasma composition and the target condition can also change due to the formation of a compound on its surface

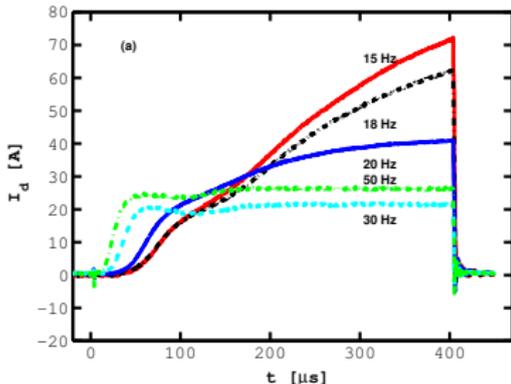


After Magnus et al. (2011) JAP **110** 083306



## HiPIMS - Voltage - Current - time

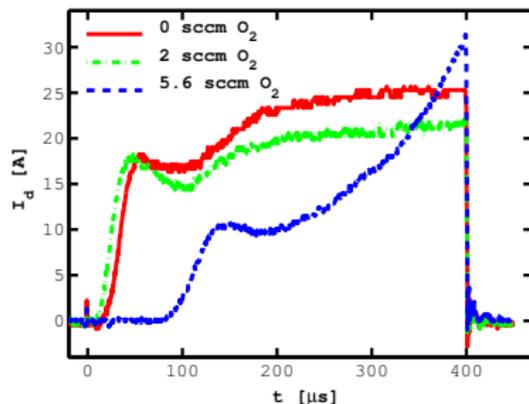
- Similarly for the Ar/O<sub>2</sub> discharge, the current waveform is highly dependent on the repetition frequency and applied voltage which is linked to oxide formation on the target
- The current is found to increase significantly as the frequency is lowered



After Magnus et al. (2012), JVSTA **30** 050601

## HiPIMS - Voltage - Current - time

- As the oxygen flow is increased a transition to oxide mode is observed



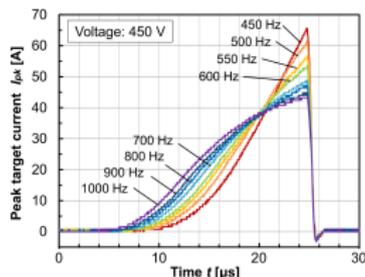
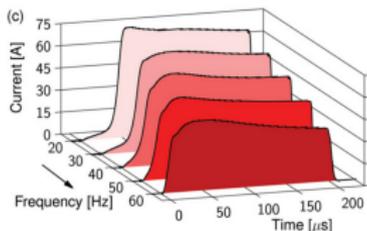
The current waveforms for an Ar/O<sub>2</sub> discharge with a Ti target where the oxygen flow rate is varied – 600 V, 50 Hz and 0.6 Pa

From Gudmundsson et al. (2013), ISSP 2013, p. 192

Gudmundsson (2016) Plasma Phys. Contr. Fus. 58 014002



## HiPIMS - Voltage - Current - time



- Similar behaviour has been reported for various target and reactive gas combinations
  - The current increases with decreased repetition frequency
  - The current waveform maintains its shape for Ar/O<sub>2</sub> discharge with Nb target
- The current waveform becomes distinctly triangular for Ar/N<sub>2</sub> discharge with Hf target

From Hála et al. (2012), JPD **45** 055204

From Shimizu et al. (2016), JPD **49** 065202



## HiPIMS - Voltage - Current - time

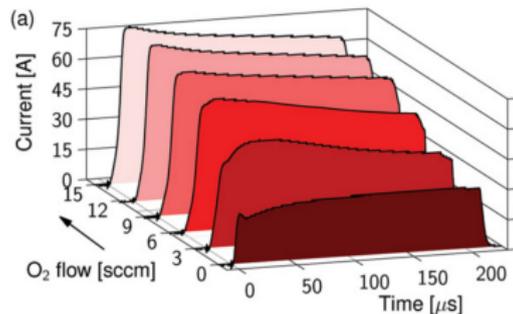
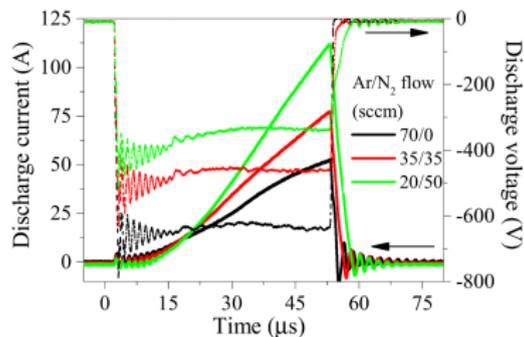
- The current increases with increased partial pressure of the reactive gas

- The current waveform becomes distinctly triangular for Ar/N<sub>2</sub> discharge with Al target

From Moreira et al. (2015), JVSTA **33** 021518

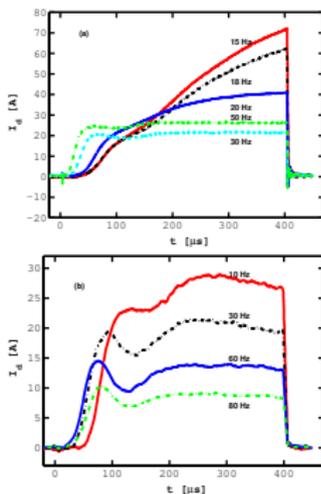
- The current waveform maintains its shape for Ar/O<sub>2</sub> discharge with Nb target

From Hála et al. (2012), JPD **45** 055204



## HiPIMS - Voltage - Current - time

- At high frequencies, nitride or oxide is not able to form between pulses, and self-sputtering by  $Ti^+$ -ions (singly and multiply charged) from a Ti target is the dominant process
- $\gamma_{see}$  is practically zero for singly charged metal ions impacting a target of the same metal
- At low frequency, the long off-time results in a nitride or oxide layer being formed on the target surface and self-sputtering by  $Ti^+$ - and  $N^+$ -ions or  $O^+$ -ions from  $TiN$  or  $TiO_2$  takes place



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

and Magnus et al. (2012), JVSTA **30** 050601

Gudmundsson (2016) PPCF **58** 014002

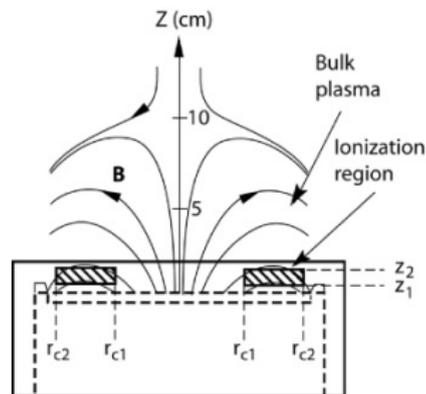


# Ionization region model studies of reactive HiPIMS



## *Ionization region model studies of 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

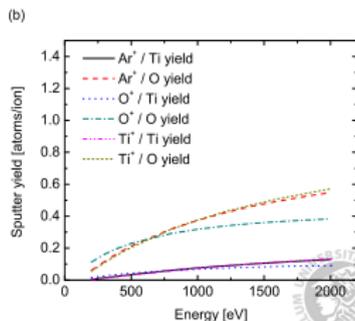
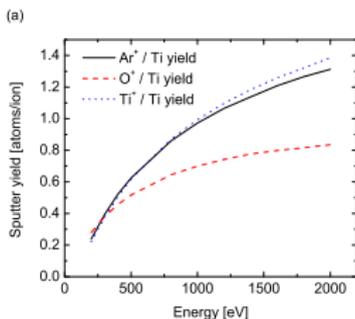
# *Ionization region model studies of reactive HiPIMS*

- The species assumed in the IRM are
  - electrons
  - argon atoms  $\text{Ar}(3s^23p^6)$ , warm argon atoms in the ground state  $\text{Ar}^W$ , hot argon atoms in the ground state  $\text{Ar}^H$ ,  $\text{Ar}^m$  ( $1s_5$  and  $1s_3$ ) (11.6 eV), argon ions  $\text{Ar}^+$  (15.76 eV)
  - titanium atoms  $\text{Ti}(a^3F)$ , titanium ions  $\text{Ti}^+$  (6.83 eV), doubly ionized titanium ions  $\text{Ti}^{2+}$  (13.58 eV)
  - oxygen molecule in the ground state  $\text{O}_2(X^3\Sigma_g^-)$ , the metastable oxygen molecules  $\text{O}_2(a^1\Delta_g)$  (0.98 eV) and  $\text{O}_2(b^1\Sigma_g)$  (1.627 eV), the oxygen atom in the ground state  $\text{O}(^3P)$ , the metastable oxygen atom  $\text{O}(^1D)$  (1.96 eV), the positive ions  $\text{O}_2^+$  (12.61 eV) and  $\text{O}^+$  (13.62 eV), and the negative ion  $\text{O}^-$

# *Ionization region model studies of reactive HiPIMS*

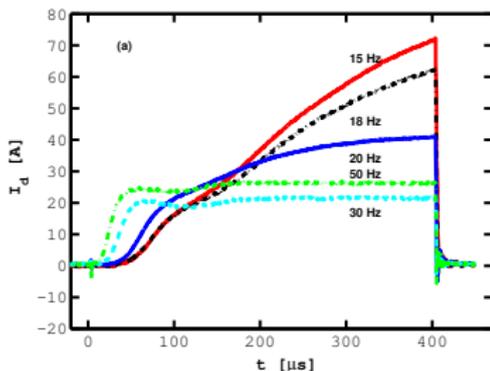
- The sputter yield for the various bombarding ions was calculated by TRIDYN for
  - **Metal mode** – Ti target
  - **Poisoned mode** – TiO<sub>2</sub> target
- The yields correspond to the extreme cases of either clean Ti surface and a surface completely oxidized (TiO<sub>2</sub> surface)
- The sputter yield is much lower for poisoned target

The sputter yield data is from Tomas Kubart, Uppsala University



## ***Ionization region model studies of reactive HiPIMS***

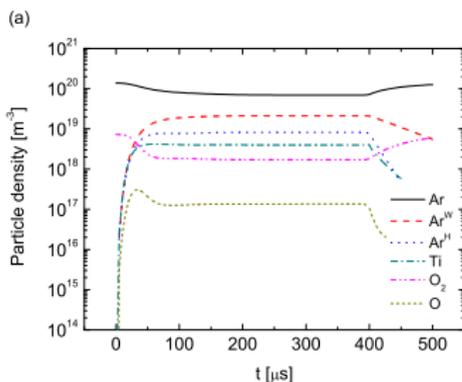
- The model is applied to explore Ar/O<sub>2</sub> discharge with Ti target in both metal mode and oxide (poisoned) mode
- The IRM is a semi-empirical model in the sense that it uses a measured discharge current waveform as a main input parameter
- For this study we use the measured curve for Ar/O<sub>2</sub> with Ti target at 50 Hz for metal mode and at 15 Hz for poisoned mode



After Magnus et al. (2012), JVSTA **30** 050601

# *Ionization region model studies of reactive HiPIMS*

- The gas rarefaction is observed for the argon atoms but is more significant for the  $O_2$  molecule
- The density of Ti atoms is higher than the  $O_2$  density
- The atomic oxygen density of is over one order of magnitude lower than the molecular oxygen density – the dissociation fraction is low



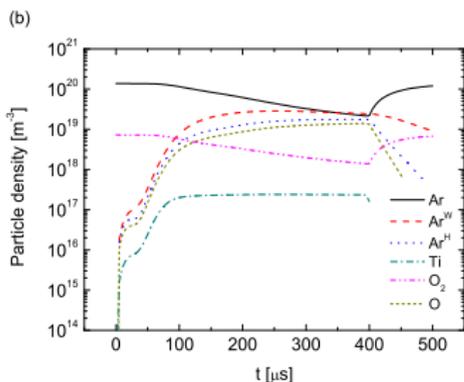
The temporal evolution of the neutral species with 5 % oxygen partial flow rate for Ar/ $O_2$  discharge with Ti target in **metal mode**.

Gudmundsson et al. (2016), PSST, submitted 2016



## ***Ionization region model studies of reactive HiPIMS***

- Gas rarefaction is observed for both argon atoms and O<sub>2</sub> molecules
- The density of Ti atoms is lower than both the O<sub>2</sub> density and atomic oxygen density
- The atomic oxygen density is higher than the O<sub>2</sub> density towards the end of the pulse



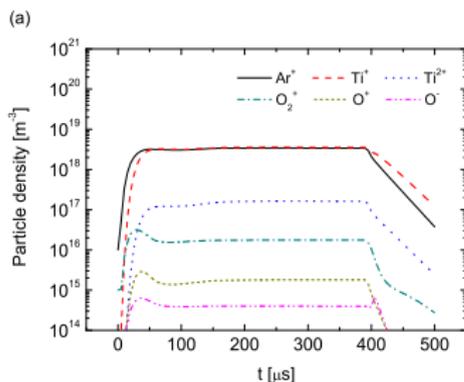
The temporal evolution of the neutral species with **5 % oxygen partial flow** rate for Ar/O<sub>2</sub> discharge with Ti target in **poisoned mode**.

Gudmundsson et al. (2016), PSST, submitted 2016



# ***Ionization region model studies of reactive HiPIMS***

- $\text{Ar}^+$  and  $\text{Ti}^+$ -ions dominate the discharge
- $\text{Ti}^{2+}$ -ions follow by roughly an order of magnitude lower density
- The  $\text{O}_2^+$  and  $\text{O}^+$ -ion density is much lower



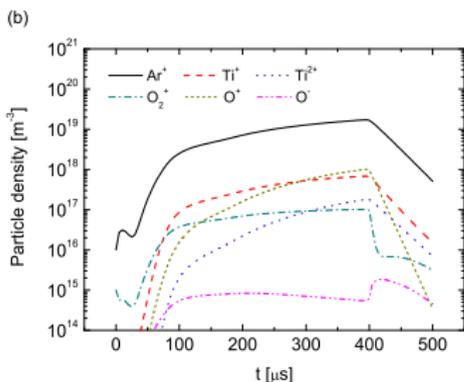
The temporal evolution of the neutral species with **5 % oxygen partial flow rate** for  $\text{Ar}/\text{O}_2$  discharge with Ti target in **metal mode**.

Gudmundsson et al. (2016), PSST, submitted 2016



# ***Ionization region model studies of reactive HiPIMS***

- Ar<sup>+</sup>-ions dominate the discharge
- Ti<sup>+</sup>, O<sup>+</sup>, have very similar density, but the temporal variation is different, and the O<sub>2</sub><sup>+</sup> density is slightly lower
- The Ti<sup>2+</sup>-ion density increases fast with time and overcomes the Ti<sup>+</sup> density towards the end of the pulse



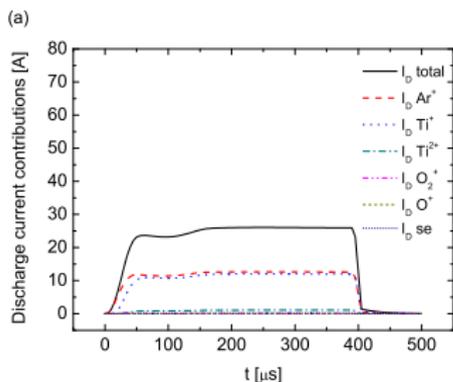
The temporal evolution of the neutral species with 5 % oxygen partial flow rate for Ar/O<sub>2</sub> discharge with Ti target in **poisoned mode**.

Gudmundsson et al. (2016), PSST, submitted 2016



# *Ionization region model studies of reactive HiPIMS*

- $\text{Ar}^+$  and  $\text{Ti}^+$ -ions contribute most significantly to the discharge current



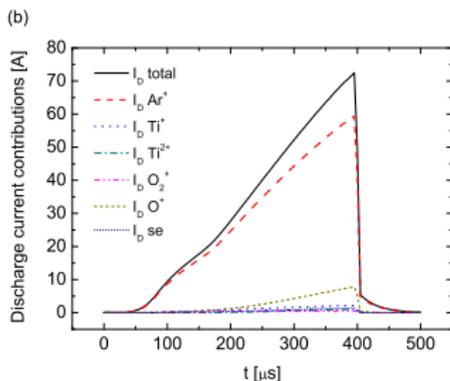
The temporal evolution of the neutral species with **5 % oxygen partial flow rate** for  $\text{Ar}/\text{O}_2$  discharge with Ti target in **metal mode**.

Gudmundsson et al. (2016), PSST, submitted 2016



## ***Ionization region model studies of reactive HiPIMS***

- $\text{Ar}^+$  contribute most significantly to the discharge current – almost solely
- The contribution of secondary electron emission is very small



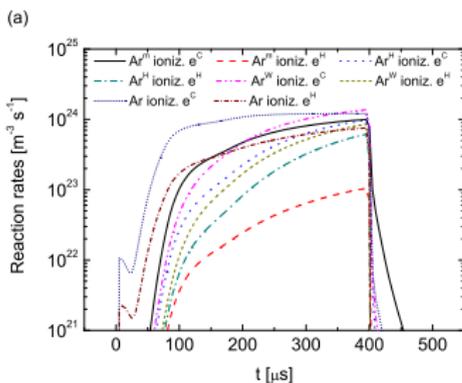
The temporal evolution of the neutral species with **5 % oxygen partial flow** rate for  $\text{Ar}/\text{O}_2$  discharge with Ti target in **poisoned mode**.

Gudmundsson et al. (2016), PSST, submitted 2016



# *Ionization region model studies of reactive HiPIMS*

- Recycling of ionized atoms coming from the target are required for the current generation in both modes of operation
- In the metal mode self-sputter recycling dominates and in the poisoned mode working gas recycling dominates
- The dominating type of recycling determines the discharge current waveform



The temporal variations of the reaction rates for electron impact ionization of the argon atoms (ground state plus metastable) in poisoned mode.

Gudmundsson et al. (2016), PSST, submitted 2016



# Summary



## Summary

- An ionization region model was used to explore the plasma composition during the high power pulse
- Comparison was made between the metal mode and the poisoned mode
  - In metal mode  $\text{Ar}^+$  and  $\text{Ti}^+$ -ions dominate the discharge and are of the same order of magnitude
  - In poisoned mode  $\text{Ar}^+$ -ions dominate the discharge and two orders of magnitude lower,  $\text{Ti}^+$ ,  $\text{O}^+$ , have very similar density, with the  $\text{O}_2^+$  density slightly lower
  - In the metal mode  $\text{Ar}^+$  and  $\text{Ti}^+$ -ions contribute most significantly to the discharge current while in poisoned mode  $\text{Ar}^+$  dominate
- In the metal mode self-sputter recycling dominates and in the poisoned mode working gas recycling dominates – the dominating type of recycling determines the discharge current waveform



The slides can be downloaded at

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

- The experimental work was made in collaboration with
  - Dr. Fridrik Magnus, Uppsala University, Uppsala, Sweden
  - Tryggvi K. Tryggvason, University of Iceland
- We got help with the sputtering yields from
  - Dr. Tomas Kubart, Uppsala University, Uppsala, Sweden
- and the project is funded by
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