On electron heating in magnetron sputtering discharges

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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 $E \times B$-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 \( \frac{n_i - n_e}{n_e} \geq 1 \)
- The ionization region (IR) is where most of the ionization occurs.
- In the bulk plasma (BP), cross-\( \mathbf{B} \) 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
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

\[ \mathcal{N} \approx \frac{V_D}{E_c} \]

where \( E_c \) is the energy loss per electron-ion pair created.

\[ \begin{align*}
\text{Cl}_2 & \quad \text{Cl} & \quad \text{Ar} \\
1 & \quad 10^1 & \quad 10^2 \\
10 & \quad 10^2 & \quad 10^3 \\
100 & \quad 10^3 & \quad 10^4
\end{align*} \]
To account for the electrons that are not trapped we define an effective secondary electron emission coefficient

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

- $\epsilon_e$ 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

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

has to be fulfilled

This defines the minimum voltage to sustain the discharge as

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

referred to as Thornton equation

- $\beta$ is the fraction of ions that return to the cathode
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

\[
\frac{1}{V_D} = \frac{\beta m \epsilon_e (1 - r)}{\epsilon_c} \gamma_{SE}
\]

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. This requirement can be expressed by the following relationship for the minimum potential to sustain such a discharge.

\[
V_{\text{min}} = \frac{\delta_0}{\Gamma_i \epsilon_i \epsilon_e}
\]
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 $E_c$ are independent of $\gamma_{SE}$.
dc magnetron sputtering discharge

- \( \frac{1}{V_D} \) 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:

$$\frac{1}{V_D} = \frac{\beta \epsilon^H_m (1 - r)(1 - \delta_{IR})}{\varepsilon^H_c} \gamma_{SE} + \frac{\epsilon^C_e \langle I_e/I_D \rangle_{IR} \delta_{IR}}{\varepsilon^C_c}$$

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 $e^C$. 
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 $J_e \cdot E$ to the electrons in the plasma volume outside the sheath.

From Brenning et al. (2016) PSST 25 065024
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 065024
On electron heating in magnetron sputtering discharges

**dc magnetron sputtering discharge**

<table>
<thead>
<tr>
<th>( i_D ) (A)</th>
<th>( p ) (Pa)</th>
<th>Slope ( k )</th>
<th>Intercept ( I )</th>
<th>( \delta_{IR} = \frac{V_{IR}}{V_D} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.4</td>
<td>0.4</td>
<td>0.0117</td>
<td>0.00145</td>
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<td>0.6</td>
<td>0.0140</td>
<td>0.00110</td>
<td>0.15</td>
</tr>
</tbody>
</table>

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

\[
\delta_{IR} = \frac{V_{IR}}{V_D}
\]

can be estimated from

\[
b = \frac{\epsilon_e^C \langle l_e/l_D \rangle_{IR} \delta_{IR}}{\mathcal{E}_c^C}
\]

We assume

\[
\epsilon_e^C = 0.8, \quad \langle l_e/l_D \rangle_{IR} \approx 0.5,
\]

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
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.

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 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

\[ p_t = 0.05 \text{ kW/cm}^2 \text{ dcMS limit} \]

\[ p_t = 0.5 \text{ kW/cm}^2 \text{ HiPIMS limit} \]
Temporal and spatial variation of the electron density
Ar discharge at 20 mTorr, Ti target, pulse length 100 µs
The electron density in the substrate vicinity is of the order of $10^{18} - 10^{19} \text{ m}^{-3}$ – ionization mean free path $\lambda_{iz} \sim 1 \text{ cm}$. 

(After Bohlmark et al. (2005), IEEE Trans. Plasma Sci. 33 346)
Ionization region model studies of 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.

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)

Detailed model description is given in Huo et al. (2017), JPD submitted 2017
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
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
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
A primary current $I_{\text{prim}}$ 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

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

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.

Anders et al. (2012), JPD 45 012003
Huo et al. (2014), PSST 23 025017
For the Al target the critical current is $I_{\text{crit}} \approx 7$ 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 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
The discharge with the Ti target is operated with peak current far above the critical current of $I_{\text{crit}} \approx 19$ 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 $I_{\text{SS-recycle}}$ and working gas-recycling $I_{\text{gas-recycle}}$.

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

Experimental data from Bradley et al. (2015) JPD 48 215202
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
The power transfer to the electrons is given by

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

where

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

and

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

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

\[ I_{e,\text{SH}} \ll I_D / 2 \]

and Ohmic heating is more efficient.
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
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.
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)

It is taken at the end of a 400 $\mu$s 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
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/ranms.html

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References


