Velocity distribution of metal ions in the target region of HiPIMS: the role of Coulomb collisions

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Abstract

High power impulse magnetron sputtering (HiPIMS) discharges have become an important tool for the deposition of thin, hard coatings. Such discharges are operated at a very low working gas pressure in the order of 1 Pa. Therefore, elastic collisions between ions and other heavy particles are often calculated to occur with low frequency, using the hard sphere approximation. However, inside the magnetic trap region of the discharge, a very dense plasma is created and Coulomb collisions become the dominant collision process for ions. In this article, we show that Coulomb collisions are a necessary part of a complete description of ion movement in the magnetic trap region of HiPIMS. To this end, the velocity distribution function (VDF) of chromium and titanium ions is measured using high-resolution optical emission spectroscopy. The VDF of those ions is then described using a simple simulation which employs a direct simulation Monte Carlo scheme. The simulation describes the self-relaxation of the VDF that is initially a Thompson distribution as being created during the sputtering process. Measurement positions inside the discharge are matched to the simulation results choosing an appropriate relaxation time. In this fashion, excellent agreement between simulation and measurement is obtained. We find, that the distribution quickly becomes mostly Maxwellian with a temperature of 9 eV for titanium ions and 4.5 eV in the case of chromium ions. Only the high energy tail of the VDF retains the shape of the initial Thompson distribution. The observed high temperature is explained with an energy redistribution from the highly energetic Thompson distribution into an partly-thermalized Maxwell-like distribution. Finally, the temperature resulting from this energy redistribution is calculated using a simple analytical model which shows good agreement with the measurements.

Keywords: HiPIMS, VDF, sputtering, magnetron, collisions, optical emission spectroscopy, direct simulation Monte Carlo

(Some figures may appear in colour only in the online journal)

1. Introduction

In recent years, high power impulse magnetron sputtering (HiPIMS) has established itself as an interesting and flexible alternative to traditional direct current magnetron sputtering (DCMS). HiPIMS discharges are driven with a higher voltage than a DCMS discharge. However, this high voltage has then to be pulsed, in order not to overheat the target. The high applied voltage results in a very dense plasma and subsequently an increased ionization of sputtered material. Therefore, coatings are partly created by impinging ions instead of only neutrals, thus yielding superior coating quality [1–4]. However, depending on the material and discharge parameters
used, deposition rates during HiPIMS are often found to be smaller than those achieved with DCMS at the same average discharge power [5, 6].

This observed lower deposition rate is usually explained as follows: even outside the cathode sheath there is a strong electric field present in the magnetic trap region of magnetron sputtering discharges. As this field is located in front of the cathode sheath, it is sometimes described as an extended pre-sheath [7]. The field extends over a few centimeters from the target, with the electric field vector pointing towards the target surface [8]. Ions in the magnetic trap region experience this electric field and are accelerated towards the target. Since a large fraction of the sputtered material is ionized during HiPIMS, it is expected that those ions will partly return to the target surface and therefore not reach the substrate [1, 2]. Therefore, the electric field acts as a potential barrier that ions have to overcome in order to leave the magnetic trap region of the discharge. Otherwise, they return to the target surface (return effect [6]).

A detailed understanding of the movement of ions is necessary to overcome this return effect and optimize the deposition rate in HiPIMS. However, this necessitates the measurement of the velocity distribution function of ions (IVDF), which is very challenging.

IVDF measurements can readily be performed using energy resolved mass spectrometry [9, 10]. Mass spectrometers may be used for any ion species and can even detect the small amount of ions with higher charge states [11]. The obtained IVDFs often feature very little noise and high temporal resolution [12, 13]. However, mass spectrometer measurements are necessarily performed on the outside of the magnetic trap region. Because of the potential barrier, only a small subset of ions will leave the magnetic trap and reach the mass spectrometer. These leaving ions are generally not representative for the ions still inside the magnetic trap region or for those returning to the target surface. The interpretation of IVDFs obtained with mass spectrometers is, therefore, rather ambiguous [14].

A more direct access to the IVDFs inside the magnetic trap region is possible using active or passive optical techniques. For example, Desecures et al measured the VDF of metal neutrals [15, 16]. The authors used a tunable diode laser to perform laser induced fluorescence (LIF) measurements to obtain the Doppler broadened excitation line profile which provides the VDF. Similarly, the VDF of titanium ions has been measured by Palmucci et al using a ns pulsed dye laser for LIF [17]. The authors found the width of the VDF to increase during the discharge pulse and then to saturate. The authors explained the initial increase in width with a decreasing amount of argon, present in the magnetic trap. Argon is expelled from the magnetic trap region by collisions with the sputtered material in a process called rarefaction. The saturation plateau in the width of the VDF was interpreted by the authors as complete working gas rarefaction. However, the authors did not attempt to explain the exact shape of the VDF, as the measurements were strongly affected by noise. Later, Britun et al measured maps of both the VDF and the density of titanium ions, time and space resolved, using the same setup [18]. However, the authors neglected all heavy particle collisions in their analysis of the data and could not fully explain the measured IVDF.

In this work we investigate the IVDF using a combined experimental and computational approach. High resolution optical emission spectroscopy is used to determine the VDF of titanium and chromium ions in the discharge by evaluation of emission line profiles. Additionally, the VDF relaxation from the initial Thompson distribution is simulated using a direct simulation Monte Carlo (DSMC) approach. The results from this simulation are used to explain the measured VDFs and to gain a more general insight into the importance of ion collisions in HiPIMS plasmas.

2. Experimental setup

2.1. Chamber and discharge

The discharge was operated in argon at a pressure of 0.5 Pa. The discharge was created using a planar magnetron assembly (Thin Film Consulting IX2U) suitable for round targets with a diameter of 50 mm or 2”. The magnetic field configuration for this magnetron assembly was reconstructed from measurements according to the method of Krüger et al and is shown as part of figure 1(b) [19]. The measurement was performed on a magnetron assembly of the same model (Thin Film Consulting IX2U) using a Hall probe. The figure is true to scale. We define the magnetic trap region as the region between the target surface and the separatrix, which marks the position of the last closed field line. In this magnetic trap, electrons are confined by the magnetic field up to the separatrix on the one side and the target on the other side where electrons are repelled by the strong negative potential. The distance between target and separatrix is about 15 mm at the racetrack position, \( r = 13.5 \text{ mm} \). The magnetic null is located about 18 mm above the target.

Chromium or titanium were used as materials for the 3 mm thick targets. In case of titanium, the discharge was operated with a voltage pulse of 590 V, a pulse length of 100 \( \mu \text{s} \) and a repetition frequency of 40 Hz. The peak current during the pulse was 50 A, or about 2.5 A \( \text{cm}^{-2} \) when normalized to the target surface area. The peak target power density was 1.1 kW \( \text{cm}^{-2} \). The current and voltage time traces for titanium as the target material can be found in a previous publication [20].

For chromium, the applied voltage was 800 V, the pulse length 150 \( \mu \text{s} \) and the repetition frequency 60 Hz. The peak current was 36 A, or 1.8 A \( \text{cm}^{-2} \), respectively. The peak target power density was 1.3 kW \( \text{cm}^{-2} \).

Current and voltage were measured in the cable connecting the power supply (TRUMPF Hütttinger TruPlasma Highpulse 4002) and the magnetron assembly using commercial probes (Tektronix TCP A400, Tektronix P6015A).

2.2. High resolution optical emission spectroscopy and deconvolution

Figure 1(a) shows the setup for the high resolution optical emission spectroscopy. The light, generated by the discharge, is coupled into an optical fibre (800 \( \mu \text{m} \)) using a lens (\( f = 150 \text{ mm} \)). The discharge is observed in the direction parallel to the target surface. The lens limits the field of view of
Figure 1. Experimental setup. (a) Illustration of the setup including the magnetron assembly, chamber, the optical path and the spectrograph. (b) Enlarged view of the magnetic trap region with the magnetic field configuration reconstructed from measurements [19]. The figure is true to scale.

Table 1. Data on the energy levels of the investigated optical emission lines [21, 22]. The electron configuration is given in the notation recommended by NIST [23], originally proposed by Russell et al [24].

| Species | Wavelength (nm) | Upper level energy (eV) | Lower level energy (eV) | Configuration |
|---------|-----------------|-------------------------|-------------------------|---------------|
| Ti II   | 453.396         | 3.97                    | 1.24                    | 3d²(3F)4p     |
| Cr II   | 455.864         | 6.79                    | 4.07                    | 3d⁴(5D)4p     |

In this work, the VDF in target normal direction was not measured. In this direction much stronger gradients in VDF and light intensity are expected. Line integration over these strong gradients would make the interpretation of the obtained VDFs very challenging and prone to errors.

The light was analyzed using a plane grating spectrograph (Zeiss PGS 2, 1300 lines/mm grating) equipped with an iCCD camera (Andor iStar DH320T-25U-A3). The light from a hollow cathode lamp (HCL, Cathodeon 3UNX Ti/Cr) was measured at the same time to provide an unshifted wavelength reference to the plasma emission. The spectrograph was operated in the third diffraction order in order to provide a sufficient spectral resolution of about 1.5 pm pixel-to-pixel. Table 1 lists the investigated emission lines with energy and electron configuration of the levels involved in the transitions.

Figure 1(b) shows an enlarged view of the magnetic trap of the magnetron sputtering discharge. The figure indicates the anode cover, which has a gap in our case, in order to enable the observation of emission located very close to the target surface. The figure also shows an estimation of the field of view of the optical system. All dimensions are true to scale.

The emission line profile of titanium and chromium ions during HiPIMS is mostly determined by instrumental and Doppler broadening, with only minor contributions from other broadening mechanisms [20]. To obtain the VDF, a Wiener deconvolution is performed, thus removing the instrumental profile from the measured line profile. The VDF is then obtained from the line by transforming from wavelength $\lambda$ to velocity $v$ using $v = c(\lambda/\lambda_0 - 1)$. The reference $\lambda_0$, respectively $v_0 = 0$, is obtained by fitting the instrumental profile to the HCL emission, for each measurement. More details on the instrumental profile, the HCL operation and the optical system can be found in a previous publication [20].

3. Collisions, heating and VDF relaxation model

3.1. Elastic collisions in HiPIMS plasmas

The hard sphere approximation is sometimes employed to calculate elastic collisions between heavy particles when describing the ion movement inside the magnetic trap region of magnetron sputtering discharges [25, 26]. Since magnetron sputtering discharges are in general operated at a very low pressure in the order of 1 Pa, the mean free path $\lambda_{\text{Col}}$ for such collisions then calculates to a few centimeter [27]:

$$\lambda_{\text{Col}} = \frac{1}{n\sigma}$$  \hspace{1cm} (1)

with the particle density $n$ and the cross section $\sigma$:

$$\sigma_{\text{HS}} = \pi(a_1 + a_2)^2.$$  \hspace{1cm} (2)

Here, $a_1$ and $a_2$ are the radii of the colliding particles. For example, a titanium test particle ($a_1 = 246$ pm [28]) in an argon ($a_2 = 183$ pm [28]) background with a density of $1.2 \times 10^{20}$ m⁻³ (0.5 Pa at room temperature) has a mean free path of $\lambda_{\text{Col}} = 14$ mm. This is about the size of the magnetic trap of a 2" magnetron assembly.

However, since the ion density in HiPIMS is very high, Coulomb collisions will be the more important process for heavy particle collisions [29, 30]. The mean free path for such collisions can be estimated using the equation for the
cross section ($\sigma_q$) of many consecutive small angle Coulomb collisions leading to a $90^\circ$ collision [27]:

$$\sigma_q = \frac{8}{\pi} \left( \frac{q_1 q_2}{2 e_0 m_R v_R^2} \right)^2 \ln \Lambda \quad (3)$$

with the reduced mass $m_R$, the charges of collision partners $q_1$, $q_2$, the relative velocity between collision partners $v_R$ and the Coulomb logarithm $\ln \Lambda$ [27]. For example, a titanium ion with a velocity of $v_R = 3 \text{ km s}^{-1}$ (2.3 eV) moving in a resting ion background with a density of $5 \times 10^{20} \text{ m}^{-3}$ has a mean free path of only about 0.5 mm. As a consequence, Coulomb collisions may not be neglected in the description of ion movement in HiPIMS plasmas.

The Coulomb scattering cross section depends strongly on the relative velocity between the colliding particles: $\sigma_q \propto v_R^{-4}$. Therefore, very fast particles can traverse the magnetic trap without collisions. Additionally, the plasma essentially becomes more transparent the hotter it gets. To quantify this effect, figure 2 shows the mean free path for $90^\circ$ Coulomb collisions between a $3 \text{ km s}^{-1}$ test particle and an argon ion background with different temperatures and densities. The mean free path for hard sphere collisions with an argon background at a fixed density of $1.2 \times 10^{20} \text{ m}^{-3}$ is shown for comparison.

The velocity $v_R$ between collision partners was numerically calculated according to

$$v_R = \int_{-\infty}^{\infty} |\vec{v} - \vec{v}_i| f_M(\vec{v}, T_i) d\vec{v} \quad (4)$$

with the Maxwell velocity distribution function $f_M$, the ion background temperature $T_i$, and the velocity of the test particle $|\vec{v}_i| = 3 \text{ km s}^{-1}$. The figure shows that a particle moving in a cold ion background will be much more affected by collisions. However, even for very high temperatures of $T_i = 70000 \text{ K}$ (6 eV), the mean free path for Coulomb collisions is short enough to ensure many collisions to occur inside the magnetic trap region in the HiPIMS regime. Only for an unlikely combination of extremely high temperature and low plasma density would the discharge become collisionless.

In order to assess the importance of Coulomb collisions for the description of ion movement in a HiPIMS discharge, it is, therefore, first necessary to discuss the mechanisms which might affect the effective temperature of ions in HiPIMS discharges.

### 3.2. Ion heating and cooling

In HiPIMS, we need to consider two very different kind of ions: working gas (usually argon) ions are created from the initially cold background gas and are heated up during the discharge pulse. On the other hand, ions created from the sputtered material initially follow a Thomson distribution. These particles are, therefore, initially highly energetic and directed. The average energy contained in the Thomson distribution can be calculated according to [20]:

$$\langle E_f \rangle \approx 2 E_b \ln \left( \frac{E_{\text{max}}}{E_b} \right) - 3 E_b \quad (5)$$

with the surface binding energy $E_b$ and the cutoff energy $E_{\text{max}}$, which is usually the maximum energy transferred by impinging ions [31]. Assuming $E_{\text{max}}$ to be close to the discharge voltage $E_{\text{max}} \approx 500 \text{ eV}$ and using $E_b = 4.6 \text{ eV}$ for titanium, we find $\langle E_f \rangle \approx 30 \text{ eV}$. The ion population created from sputtered particles, thus, starts highly energetic even before any additional heating occurs.

Additional heating might be caused by the strong electric field present in the magnetic trap region of HiPIMS. This Ohmic heating is known to be very important for the heating of electrons [32].

Cooling is mostly caused by interaction with ions created from the cold working gas. Given enough time to equilibrate, the two ion populations (sputtered and working gas) will thermalize to a common temperature. However, it is difficult to quantitatively describe this process, as the ion composition during the HiPIMS pulse is unknown. To our knowledge, no measurement of the argon ion density during HiPIMS exists.

Instead, we will limit our investigation to the case of strong working gas rarefaction which is expected to occur at the end of an high power discharge pulse [33–35]. There, most of the argon should have already been expelled from the magnetic trap region. Simulations predict the argon density to be reduced by as much as 90%, depending on the discharge conditions and the utilized target material [36–38].

The strong rarefaction is counteracted by a process called working gas recycling: argon ions that reach the target during the initial stages of the discharge are neutralized and diffuse back into the discharge volume, providing a source of fresh neutrals at the target surface [7, 39]. This process might provide an important pathway for a substantial argon ions density during later times of the discharge pulse, where the initial background population of argon neutrals is already mostly expelled. However, measurements indicate that this process can not compensate strong rarefaction [11, 17, 33–35, 40, 41].
Consequently, we assume that the argon ion density will be low compared to the density of metal ions when working at the end of the discharge pulse and operating at a high peak discharge power.

In this case, the problem simplifies to the one of self-relaxation: ions of sputtered particles are now mostly colliding with each other.

3.3. Simulation of VDF self-relaxation

The self-relaxation process of titanium ions is studied using a DSMC approach with three dimensions in velocity space and none in configuration space. The initial velocity of particles is assigned according to the kinetic energy predicted by the Thompson distribution $f_T(E)$:

$$f_T(E) = \frac{2E_{b}E}{(E + E_{b})^2}$$

with: $E =$ kinetic energy of sputtered particles $m =$ mass of sputtered particles $E_{b} =$ surface binding energy of the target.

The distribution of the ejection angle of particles $\theta$ is assumed to follow a cosine distribution. Thus, particles are more likely to be ejected into the direction normal to the target surface. $\theta$ is defined as usual, as the angle between the target surface normal and the direction of ejection. The direction in the surface plane is assumed to be evenly distributed between an angle of $\varphi = 0$ and $\varphi = 2\pi$.

The Thompson distribution is known not to be entirely accurate at the conditions present in this study. A slightly different distribution of ejection angles and a higher energy for oblique ejection angles is expected [42, 43]. However, accurately including these effects would complicate the simple analysis presented here. Additionally, we do not expect these effects to strongly affect the obtained results because discrepancies to the Thompson distribution are mostly found at high energies [42]. These highly energetic particles will not strongly affect the VDF relaxation, since the cross section for Coulomb collisions becomes small for fast relative velocities between colliding particles. However, if ion–neutral or neutral–neutral collisions are important, the more accurate distribution by Stepanova and Dew might be preferable [36, 42].

$3 \times 10^6$ particles are assigned a random set of kinetic energy and angles $\theta$ and $\varphi$. From this, the absolute value of the velocity is calculated and then projected to the $x$, $y$ and $z$ axis in order to assign the particles velocity components in three dimensions.

The population of ions is then subjected to collisions with each other. In each time step, the ion population is split into two equally sized ensembles. Each ion from one ensemble collides with a random ion from the other ensemble until all ions have undergone a single collision. The new velocity after each collision is calculated according to the model by Nanbu [44]. The model allows the treatment of many consecutive small angle collisions as a single large angle binary interaction. This allows for the use of large time steps. In our case, we calculate 400 time steps with a step size of $\Delta \tau = 50$ ns.

The ion density is estimated to $1 \times 10^{20}$ m$^{-3}$, based on probe measurements performed inside the magnetic trap region under similar conditions [45–47]. The uncertainty in this estimate is of no major concern as the collision model scales the deflection angle and momentum transfer proportional to the product of the ion density $n$ and the size of the time step: $\Delta \tau \cdot n$. Thus, if we were to underestimate the ion density for example, we would still reach the same relaxation stage, just at a later point in time.

4. Results

4.1. Simulated VDF relaxation

Figure 3 shows the simulated time evolution of the velocity distribution function (VDF) of titanium ions. Figure 3(a) shows the VDF in the direction normal to the supposed sputtered surface ($z$ direction). In contrast, figure 3(b) shows the VDF in the direction parallel to the sputtered surface ($x$ direction). The simulation was performed with an ion density of $1 \times 10^{20}$ m$^{-3}$ and a time step of $\Delta \tau = 50$ ns. The VDFs are shown as probability densities and, thus, area normalized to unity.

Initially, at $t = 0$ the VDF has a shape described by the Thompson distribution. After only 0.4 $\mu$s, the VDF is already noticeably broadened, as shown by figure 3(a) for the direction normal to the sputtered surface. Subsequently, the VDF becomes broader and broader with time, as the energy contained in the Thompson distribution is redistributed in velocity space. The figure contains an enlarged view of the high velocity tail of the distribution, between 14 km s$^{-1}$ and 32 km s$^{-1}$. Here, the amount of these fast particles between about 15 km s$^{-1}$ and 28 km s$^{-1}$ is reduced after 10 $\mu$s. The energy of these fast particles is redistributed and causes the broadening of the VDF around the peak. For even faster particles with velocities above 30 km s$^{-1}$ the collision cross section becomes small and particles are unlikely to collide. Therefore, the VDF remains largely unaffected by Coulomb collisions at very high velocities.

With increasing relaxation time the peak of the VDF is reduced and shifts towards higher velocities. After 10 $\mu$s, the VDF has mostly the shape of a shifted Maxwellian, centered around a flow velocity of $v_f = 5$ km s$^{-1}$.

In a real HiPIMS discharge, this flow velocity will be affected by the electric field present in the magnetic trap region. For example, a moderate field of 2000 V m$^{-1}$ will shift the center of the VDF to 1 km s$^{-1}$ after only 1 $\mu$s. However, neglecting higher charge states, the electric field will affect all ions the same way. As a consequence, the relative velocity of ions is not changed by the electric field. Therefore, the electric field does not alter the collision processes.

The insight that the ion–ion collisions are largely unaffected by the electric field has three very important consequences:

(a) Close to the target surface and above the racetrack, the electric field points purely in $z$ direction [48]. Therefore, the VDF parallel to the target surface is independent of the electric field and only affected by collisions in this region.
The comparison between experiment and simulation is, therefore, reasonable.

(b) Ohmic heating of ions does not occur, since the electric field will only shift the VDF, but not change its shape. This might also indicate little heating to be caused by spokes, low frequency waves or patterns found during HiPIMS which modulate the electric field [46, 49–51].

(c) The electric field does not strongly affect collisions, and in turn, collisions do not strongly affect the overall ion transport. While the VDF is clearly changing due to collisions, the overall ion transport is mostly determined by the average velocity of the ions. This average velocity is strongly affected by the electric field, but not by ion–ion collisions. However, it is unclear how collisions might affect the electric field, thus influencing the ion transport indirectly.

These conclusions are only valid when we neglect the influx of cold argon and freshly sputtered particles into the magnetic trap region while also neglecting ion–neutral collisions and higher charge states.

Argon ions flowing back from the bulk plasma into the magnetic trap region will likely be cold with a low average velocity. Collisions between argon and titanium ions will, therefore, cool the titanium ion VDF and shift the center of the VDF closer to zero. Freshly sputtered material, on the other hand, will heat the VDF and shift the center towards 5 km s\(^{-1}\). Double and triple charged ions will be more strongly accelerated by electric fields and will then dissipate their kinetic energy through collisions, thus causing some Ohmic heating of the single charged ion population. However, we expect all of those effects to be small for the case of a high density, strongly rarefied HiPIMS plasma, at the end of a discharge pulse.

Qualitatively, the relaxation process for metal ions should be the same for different target materials. However, the relaxation times and width of the VDF at a given point in time will differ from material to material, due to the different masses and surface binding energies. Consequently, we find the same trends with chromium as the target material (not shown), but observe a quicker relaxation, as will be explained in detail later on.

4.2. Comparing the measured and calculated VDF

Figure 4(a) shows the width (full width at half maximum—FWHM) of the VDFs obtained from optical emission spectroscopy measurements as a function of target distance. These VDF widths were calculated after smoothing the measured VDFs with a Savitzky–Golay filter and interpolating the gaps between measurement points using B-splines. Error bars in the figure indicate the standard deviation of widths obtained from three consecutive measurements. Figure 4(b) shows an enlarged view of the chromium measurement again to make small changes more visible. The width of the titanium ion VDF initially increases with target distance form 8.9 km s\(^{-1}\) to 10.7 km s\(^{-1}\) at \(z = 7\) mm. After that point, the width of the VDF begins to reduce and reaches 9.2 km s\(^{-1}\) at \(z = 15\) mm.

Under comparable conditions, Palmucci et al observed a width of 11 ± 2 km s\(^{-1}\) for the VDF of titanium ions, using LIF at a target distance of 15 mm [17]. This is an excellent agreement to the results presented here, considering the large measurement uncertainty of LIF when applied during the on-time of HiPIMS.

For the measurement with chromium as the target material, a similar trend can be observed. Here, the width increases from 7.3 km s\(^{-1}\) to 7.5 km s\(^{-1}\) and then drops to 7.0 km s\(^{-1}\).

For both target materials, the initial rise in width can be explained with a redistribution of energy from the high energetic Thompson distribution. As particles are created at the target surface and travel through the discharge, they become ionized and will subsequently start to collide with other ions. The further these ions travel, the more time has passed for the VDF to relax.

Subsequently, ions will also be affected by the electric fields, present in the magnetic trap region of HiPIMS, and will be back attracted towards the target surface. The VDF of these ions is already close to relaxation, leading to a mix....
of newly created and nearly thermalized ions close to the target surface. Assuming the ionization frequency to decrease with target distance, the fraction of freshly ionized particles will also decrease with target distance. Thus the average lifetime of ions is increasing with target distance and a larger target distance may be correlated with longer average relaxation time and, therefore, an increased FWHM, as shown in figure 3(b).

Another possible explanation for the increasing VDF width might be the influence of argon recycling: argon ions that were implanted earlier during the discharge pulse are expected to continuously outgas from the target, entering the discharge again as neutrals. If the contribution of these particles dominates over the argon refill from the bulk plasma, then we might expect the density of argon neutrals and likely also ions to decrease with target distance. Because we identified interaction with initially cold argon ions as the most important cooling mechanism for energetic sputtered particles, this possible gradient in argon density might explain the observed increase in metal ion VDF width with target distance.

The increase of VDF width with target distance implies that Ohmic heating only plays a minor role. Otherwise, the VDF width should be decreasing with target distance, as the electric field strength has also shown to be decreasing with target distance [48, 52].

The decrease of VDF width for target distances larger than 8 mm, might be explained by the decreasing influence of Ohmic heating. However, a more likely cause is cooling by interaction with cold working gas particles, which should become more numerous with increasing target distance. Therefore, we will limit the comparison between simulation and measurement to the first 8 mm target distance, where the influence of the working gas seems to be small.

Figure 5 shows the measured VDFs for four different target distances. Each of the graphs shows three consecutive measurements, shown separately to give an impression of the measurement precision. For each measurement distance, the simulations relaxation time was varied until good agreement between simulated and measured VDF was reached. The figure also shows the initial Thompson distribution at $t = 0$ for titanium ($E_b = 4.6$ eV) and a Maxwell distribution ($T = 9$ eV) for comparison. The Thompson distribution was calculated as $f_T(v_x) \propto \left(\frac{v_x^2 + 2E_b/m}{E_b/m}\right)^{-3/2}$ [53]. The measurement and the simulated VDF are both individually area normalized to unity. The unlabeled y-axis of the graphs are in units of $10^{-5}$ s m$^{-1}$. The Thompson and Maxwell distributions are height normalized to the simulation results and are, thus, in arbitrary units.

It should be noted, that simulation and experiment that are compared here do not describe precisely the same situation. In the simulation we introduce freshly sputtered and ionized particles once and then follow the VDF evolution. In contrast, each position in the experiment contains a mixture of freshly ionized particles and older ions that already had more opportunity to collide. However, both cases describe a mixture of particles which experienced a different amount of collisions, depending on the average lifetime of the ion population. As such, the comparison should be valid.

Figure 5(a) shows the target parallel VDF in a target distance of only 1.5 mm. Already at this position, $z = 1.5$ mm, the VDF is considerably wider than the Thompson distribution, shown for comparison. The simulation (DSMC) result for the self-relaxation of $1 \times 10^{20}$ m$^{-3}$ titanium ions over the relaxation time of 2.5 $\mu$s shows excellent agreement with the measurement.

Figures 5(b)–(d) show the VDFs for increasing distance to the target surface. As the target distance increases, the VDF becomes wider, more Maxwellian, and agreement to longer relaxation times in the simulation is found. This is expected, since the ionization mean free path for the sputtered metal is likely small and ionization occurs very close to the target surface. Consequently, a greater target distance correlates with a longer average time that particles remain inside the plasma and a lower fraction of newly ionized particles. In all
Figure 5. Comparison of titanium ion VDFs obtained from the simulation (DSMC) and the measurement at different relaxation times and target distances, respectively. Measurement and simulation results are independently area normalized to unity. The initial Thompson distribution and a Maxwell distribution with a temperature of $T = 9$ eV is shown for comparison. Thompson and Maxwell distribution are height normalized to the simulation results to keep the graph readable. The graphs show the measured and simulated VDFs in units of $10^{-5}$ sm$^{-1}$. The Thompson and Maxwell distributions are presented in arbitrary units.

In cases, excellent agreement between measurement and simulation can be obtained, using the relaxation time as a fitting parameter.

At a target distance of $z = 7$ mm, shown in figure 5(d), the VDF follows closely the shape of a Maxwell distribution with a temperature of $T = 9$ eV, shown for comparison. Differences between the distributions are only found at higher velocities around 10 km s$^{-1}$. This is caused by the strong velocity dependence of the Coulomb cross section (equation (3)), which leads slow particles to collide much more often than fast particles.

The VDF in the direction normal to the target surface cannot be easily measured using OES, since the technique will line integrate over regions where ions experienced different relaxation times and different electric field strengths. However, using the relaxation times obtained from the comparison between measurement and simulation in the direction parallel to the target surface enables us to speculate on the shape of the VDF in the direction normal to the target surface. Since we obtained a relaxation time of more than 10 $\mu$s at $z = 7$ mm, the VDF should be mostly relaxed towards a Maxwellian in all directions, as shown in figure 3. In reality, the situation will be much more complex because of the strong electric field present in the magnetic trap region of HiPIMS discharges. Nevertheless, we would expect only minor traces of the initial Thompson distribution to remain in the ion VDF after crossing the magnetic trap. However, the high energy tail of the Thompson distribution should remain intact, given the much smaller cross section for Coulomb collisions between fast particles. Indeed, mass spectrometer measurements often show remains of the initial Thompson distribution only for high velocities [25, 30, 54].

Figure 6 shows the measured VDFs for chromium as the target material for two different target distances. The figure also
Figure 6. Comparison of chromium ion VDFs obtained from the simulation (DSMC) and the measurement at different relaxation times and target distances, respectively. Measurement and simulation results are independently area normalized to unity. The initial Thompson distribution and a Maxwell distribution with a temperature of $T = 4.5$ eV is shown for comparison. Thompson and Maxwell distribution are height normalized to the simulation results to keep the graph readable. The unlabeled y axis of the graphs are in units of $10^{-5}$ sm$^{-1}$ for the measurement and the simulation, only.

shows the VDF obtained from the simulation, with the relaxation time adjusted for best agreement with the respective measurement. Again, the figure also shows the initial Thompson distribution for chromium ($E_b = 1.65$ eV [55]) and a Maxwell distribution ($T = 4.5$ eV) for comparison. As above, the measurement and the simulated VDF are both individually area normalized to unity. The unlabeled y axis of the graphs are in units of $10^{-5}$ s m$^{-1}$. The Thompson and Maxwell distribution are height normalized to the simulation results and are, thus, in arbitrary units.

Figure 6(a) shows the VDF obtained from the measurement in a target distance of only 1 mm together with the simulation result for a self-relaxation in $1 \times 10^{20}$ m$^{-3}$ ions, over 8 $\mu$s. Surprisingly, the VDF is nearly Maxwellian, even this close to the target surface. For target distances larger than 3 mm, the VDF becomes nearly indistinguishable from a Maxwell distribution with a temperature of $T = 4.5$ eV, as shown in figure 6 for $z = 5$ mm.

This fast relaxation implies a much higher collision frequency for the chromium discharge compared to the discharge using titanium as the target material. The most obvious reason for this higher collision frequency would be a higher ion density. To test this hypothesis, Langmuir probe measurements were performed under the same discharge conditions, using a setup introduced in a previous publication [46]. At a target distance of 8 mm, we found an ion density of $n = 5 \times 10^{19}$ m$^{-3}$ for titanium and $n = 1 \times 10^{20}$ m$^{-3}$ for chromium as the target material. This density difference, which can likely be explained by the difference in sputter yield for the two materials, will lead to a higher collision frequency by a factor of two in the case of chromium, compared to titanium as the target material.

Another factor contributing to the larger collision frequency in the case of chromium is the generally lower velocity of chromium ions. The clean chromium target surface has a surface binding energy of only 1.65 eV$^1$, compared to 4.6 eV for titanium [55, 57]. Therefore, the initial VDF in surface normal direction peaks at 1.3 km s$^{-1}$ for chromium and 2.4 km s$^{-1}$ for titanium. Since the Coulomb cross section for collisions is much larger for slow particles the collision frequency for chromium ions is higher than for titanium ions.

The difference in surface binding energies between the two target materials also causes the difference in VDF widths, shown in figure 7. Since chromium has a much smaller surface binding energy, the VDF will be less energetic. This also results in the different temperatures of the Maxwell distributions, towards which the two ion species relax: $T = 9$ eV for titanium and $T = 4.5$ eV in the case of chromium. Qualitatively, this difference can again be explained by the lower average energy of particles following a Thompson distribution with a lower value of $E_b$. As this average energy is redistributed in velocity space, the resulting temperature is a function of $E_b$.

4.3. Model of energy redistribution and thermalization temperature

The average energy of the Thompson distribution $f_T$ can be calculated as:

$$\langle E_{f_T} \rangle = \frac{\int_0^{E_{max}} E f_T dE}{\int_0^{E_{max}} f_T dE}$$

with the cutoff energy $E_{max}$, which is usually the maximum transferred energy from the impinging ion to the sputtered particle [31]. For titanium, we would set $E_{max} \approx 500$ eV and calculate the average energy to about $\langle E_{f_T} \rangle \approx 30$ eV (see

---

$^1$ It should be noted, that the surface binding energy for a clean chromium surface (1.65 eV [55]) differs substantially from the sublimation energy (4.1 eV [56]), which is often used as an estimation for the surface binding energy.
section 3.2). However, this is much higher than the average energy of the observed Maxwell distribution: \( \langle E_{f,i} \rangle = \frac{3}{2} k_B T = 13.5 \text{ eV} \).

This discrepancy can be resolved by taking into account that not all ions of the initial Thompson distribution contribute to the relaxed VDF. The strong velocity dependence of the Coulomb cross section makes it unlikely for fast, energetic particles to collide. This can be observed in figure 3(a), where the high energy tail of the distribution above \( 30 \text{ km} \cdot \text{s}^{-1} \) remains unaffected by collisions. To take this into account, the integration in equation (7) should be performed to a value for \( E_{\text{max}} \) which describes the most energetic ions still colliding in a given relaxation time. Setting the relaxation time equal to the mean time between collisions and using equation (3) we find:

\[
\tau = (n \sigma v_R)^{-1} = \frac{\pi (4 \pi \epsilon_0)^2 m_{R}^2 v_R^3}{32 q^2 q^2 n \ln \Lambda} \quad (8)
\]

with: 
- \( m_{R} \): reduced mass 
- \( v_R \): relative velocity 
- \( n \): ion density 
- \( \ln \Lambda \): Coulomb logarithm.

This equation is similar to the equation for the time needed to fill the Maxwell tail of a distribution under relaxation obtained by MacDonald et al from the Fokker–Planck equation [58, 59]. Using \( m_R = \frac{1}{2} m \) for equal particles and solving for \( v_R \) yields:

\[
v_R = \left[ \frac{128 q^2 q^2 \tau n \ln \Lambda}{\pi (4 \pi \epsilon_0)^2 m^2} \right]^{1/3} \quad (9)
\]

Here, \( v_R \) is the relative velocity between a fast particle with the velocity \( v_c \) and the rest of the VDF, \( v_c \) can then be interpreted as the highest velocity a particle may have to still collide in a time of \( \tau \). Assuming \( v_c \) to be much larger than the thermal velocity of the VDF, we can approximate \( v_R \approx v_c \) and calculate the corresponding kinetic energy:

\[
E_{\text{max}} = \frac{1}{2} m v_c^2 \approx \frac{1}{2} m v_R^2 = 8 m \left[ \frac{2 q^2 q^2 \tau n \ln \Lambda}{\pi (4 \pi \epsilon_0)^2 m^2} \right]^{2/3} \quad (10)
\]

The integrations in equation (7) can be performed analytically to [20]:

\[
\langle E_{f,i} \rangle = \left[ E_b^3 3 E_b + 4 E_{\text{max}} E_b + E_{\text{max}}^3 \right] + 2 E_b \ln (E_b + E_{\text{max}}) \\
- 3 E_b - 2 E_b \ln (E_b) \\
\cdot \left[ -E_b^2 \left( E_b + E_{\text{max}} \right) + 1 \right]^{-1} \quad (11)
\]

Substituting \( E_{\text{max}} \) from equation (10) into equation (11) yields an expression for the average energy of the already relaxed part of the VDF at a given density and after a certain relaxation time. Figure 8 shows this average energy as an equivalent temperature \( k_B T = \frac{1}{2} \langle E_{f,i} \rangle \), for titanium and chromium during self-relaxation assuming an ion density of \( 1 \times 10^{20} \text{ m}^{-3} \).

The temperature in figure 8 initially increases quickly with increasing relaxation time but shows a much smaller increase at later times. For large relaxation times, the relaxation temperatures increase only with \( T \propto \ln \tau \). This temperature increase would theoretically continue until \( E_{\text{max}} \) reaches the maximum energy transferred during the sputter process, i.e. until the energy of the Thompson distribution is completely redistributed to a Maxwell distribution and the VDF is totally thermalized.

For chromium, after a relaxation time of 10 \( \mu \text{s} \), a temperature of 4.5 eV is obtained from the model. For titanium, a value of about 9 eV can be found after 15 \( \mu \text{s} \). This is in good agreement with the measured and simulated VDFs shown in figures 5 and 6. The differences between the VDFs of the two materials are well reproduced by this simple model.
In the case of a strongly rarefied HiPIMS plasma, equations (10) and (11) may be used to obtain a rough estimate for the effective ion temperature of the relaxed part of the VDF inside the magnetic trap.

4.4. Ohmic heating and spokes

While the results presented here indicate that the effective temperature of sputtered and ionized particles is mostly defined by the initial energy with which particles were released from the target surface, some heating caused by the electric field is to be expected. This contribution is likely small, otherwise the VDF width should decrease with target distance together with the reducing electric field strength [48]. However, a minor contribution of Ohmic heating, caused by the electric field, would not be noticed by the comparison of measurement and simulation presented here. This is because the agreement between simulation and measurement was reached by adjusting the relaxation time. If the residence time of ions in the discharge is lower than the time predicted by us, a smaller part of the total VDF width would be caused by the energy redistribution by collisions and the remaining width could be explained by Ohmic heating. Additionally, it is possible that the cooling provided by the remaining argon ions in the magnetic trap is counteracting the heating provided by the electric field.

An additional source of heating is expected to be provided by spokes, long wavelength oscillations found in HiPIMS plasmas and DCMS [49, 50]. Spokes are known to cause fluctuations in the plasma potential and introduce an azimuthal electric field which can cause a periodic acceleration of ions [46, 60]. Spokes move through the discharge with velocities in the order of 10 km s\(^{-1}\) in \(\vec{E} \times \vec{B}\) direction. Consequently, due to their inertia, ions will not follow the spoke movement. Instead, most ions will only be slightly accelerated by the electric field at the leading edge of the spoke, only to be decelerated by the same amount on the trailing edge of the spoke, which features an electric field of the same strength but reverse orientation. Since all ions are accelerated simultaneously, their relative velocities remain unchanged and little heating is to be expected from this process, when only considering Coulomb collisions alone. However, a randomization of the direction of ion movement by other types of collisions could enable such a spike induced heating.

In contrast, ions with a high velocity in \(\vec{E} \times \vec{B}\) direction might be sufficiently accelerated by the advancing electric field of the spoke to reach the spoke velocity. Such ions can then spend a long time in the advancing electric field at the leading edge of the spike and can gain up to the entire potential difference between the inside of the spike and the surrounding plasma as kinetic energy. Anders et al described such ions as ‘surfing on the potential gradient’ of the advancing spokes [61]. The amount of particles that are fast enough to follow the spoke movement depends on the width of the VDF as well as the velocity of spokes.

However, in presence of a high collision frequency it is to be expected that the accelerated ions quickly collide with the slower ions population. Depending on the amount of ions that are fast enough to follow the spoke potential maximum, this effect could lead to additional heating of the ion VDF.

5. Conclusion

In this paper, we investigated the VDF of titanium and chromium ions in a strongly rarefied HiPIMS plasma.

After discussing possible collision processes, Coulomb collisions were identified as the primary mechanism for ion collisions in the magnetic trap region of HiPIMS. The hard sphere approximation is not a suitable model for collisions inside the dense magnetic trap region as it does not take the relative velocity of colliding particles into account and will strongly underestimate the collision frequency.

Using a DSMC approach, the self-relaxation of the VDF was simulated. After a relaxation time of 10 \(\mu\)s to 15 \(\mu\)s in an ion density of \(1 \times 10^{20} \text{ m}^{-3}\), the VDF was mostly Maxwellian, with a high temperature of 4.5 eV and 9 eV for chromium and titanium, respectively. Only for high velocities, the VDFs remained close to their initial Thompson distribution.

The simulated VDFs were then compared to measurements using high-resolution optical emission spectroscopy. Excellent agreement between measured and simulated VDFs were found, validating our hypothesis that the energy of metal ions is primarily determined by their initial energy gained during the sputter process prior to ionization. For a high degree of ionization and strong rarefaction, Ohmic heating seems to play only a minor role.

Finally, the differences between the VDFs for chromium and titanium were explained using a simple energy conversion model. The model might be useful to provide a rough estimation of effective ion temperatures in strongly rarefied HiPIMS discharges.

In summary, our work shows that the ion movement in HiPIMS discharges is not only affected by strong electric fields, but also by a high amount of Coulomb collisions. Those collisions redistribute the initially directed movement of ions in velocity space and might contribute to the lower deposition rate commonly observed in HiPIMS plasmas.

Based on these findings, a collisionless treatment of ions in the target region of HiPIMS plasmas might not produce reliable results. Instead, the VDF of ions is much closer to a Maxwell distribution than previously thought. Therefore, a fluid description of ion dynamics in HiPIMS appears promising.

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