Synchronising optical emission spectroscopy to spokes in magnetron sputtering discharges

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Abstract
Spokes are patterns of increased light emission, observed to rotate in front of the targets of magnetron sputtering discharges. They move through the plasma at velocities of several km s⁻¹ in or against the \( \vec{E} \times \vec{B} \) direction of the discharge. The high velocity and their initial creation at arbitrary positions render measurements of spokes challenging. For more demanding plasma diagnostic techniques that require data acquisition over multiple discharge pulses, synchronisation to the spoke movement is necessary. In this publication, we present optical emission spectroscopy of spokes in both high power impulse magnetron sputtering (HiPIMS) as well as direct current magnetron sputtering (DCMS) discharges, performed by triggering a camera on the spoke movement. Optical filters between plasma and camera allow us to isolate emission lines of metal and working gas neutrals and ions. Based on these optical measurements and previous probe studies, the dynamics of electrons drifting through spokes in both DCMS and HiPIMS is discussed. In HiPIMS, the much shorter mean free path for inelastic electron collisions enables strong ionisation inside the spoke, causing a sudden variation in electron density which leads to the distinct spoke shape. In contrast, the spoke shape for DCMS discharges seems to rather be indicative of electron energy variations.

Keywords: magnetron sputtering, spokes, HiPIMS, instability, optical emission spectroscopy, electron drift, DCMS

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

1. Introduction
Magnetron sputtering discharges feature a configuration of crossed electric and magnetic fields. Such plasmas are prone to self-organisation phenomena, that appear as instabilities and waves [1].

In recent years, high power impulse magnetron sputtering (HiPIMS) emerged as a popular variant of magnetron sputtering. Here, the applied peak power density to the plasma is increased by the application of high voltage pulses rather than one continuous voltage. The repetition frequencies are usually of a few Hz to a few kHz, with pulse lengths ranging from a few to a few hundred microseconds. The peak power densities usually reach kW cm⁻² [2]. This increased discharge power leads to a large ionisation degree up to 90% among the sputtered particles [3–5]. The high ion fluxes resulting from a HiPIMS-discharge can lead to coatings of higher quality, when compared to those deposited by direct current magnetron
sputtering (DCMS) [6–8]. Recent investigations revealed that the deposition rate of HiPIMS can be higher than DCMS by changing the applied magnetic field structure [9]. However, when using a more conventional magnetic field structure, the deposition rate is usually lower [10, 11].

Early on, Lundin et al [12] observed potential fluctuations in their HiPIMS discharge, which the authors identified as a modified two stream instability. The authors also found low frequency fluctuations which might correspond to rotating spokes, that are commonly found in Hall thruster discharges [13]. Later, these spokes were observed as distinct zones of increased emission intensity when monitoring the plasma with a fast camera [14–16].

Two to five of these spokes usually rotate along the race-track of a HiPIMS discharge at velocities of about 10 km s$^{-1}$ in $\vec{E} \times \vec{B}$ direction, using small, 2 inch magnetrons [14–17].

Subsequently, spokes were also found in DCMS and in radio frequency magnetron sputtering (RFMS) [18–20]. In these cases, spokes rotate in retrograde $\vec{E} \times \vec{B}$ direction with a lower phase velocity compared to HiPIMS [21–24]. Moreover for DCMS, fewer spokes are usually found to exist simultaneously compared to HiPIMS: on a 2 inch target, often only a single spoke is present at a time [19, 24]. Simulations by Boeuf and Takahashi [25] showed that the origin of spokes is similar for both HiPIMS and DCMS. Instabilities in the plasma lead to the formation of an electric double layer that enables electron loss. This leads to the formation of spokes in the discharge. The authors claim that the change in propagation direction with increasing current is caused by a change in the location of a plasma density maximum with respect to the overall spoke and from the ratios of the axial and azimuthal electric fields inside the spoke.

Spokes are most clearly visible close to the target surface, where the most intense light emission originates [26]. The shape of spokes is sometimes distinctly triangular, with a sharp drop in emission intensity at the trailing edge of the structure for HiPIMS [27], or at the leading edge for DCMS [19]. This distinct shape has inspired much of the physical interpretation of the phenomenon.

Anders et al [28] interpreted the sudden emission intensity drop at the trailing edge of HiPIMS spokes as indicative of a double layer. Hecimovic et al [27] initially correlated the shape of the spoke structure with the target material. However, a newer study by Hnilica et al [29] could indicate that the optical appearance of the spoke also strongly depends on working gas pressure, magnetic field strength and discharge power. The authors proposed that the triangular spoke shape may be created by increased argon ion emission at the corners of the sharp edge of the spoke.

The origin of the different spoke shapes could be identified by performing optical emission spectroscopy (OES). However, the stochastic nature of spokes, which are initially created at arbitrary positions above the racetrack, makes it particularly difficult to perform such demanding measurements.

$^1$ We could not find the propagation direction of spokes in RFMS documented anywhere in the literature. However, using the setup from [24] we found them to rotate in retrograde direction.

Andersson et al [26] used different optical filters combined with an ICCD camera to perform OES of spokes above an aluminium target in a HiPIMS discharge. They found the emission of low energy aluminium neutral transitions (Al I) to be broadly distributed along the racetrack. Transitions from higher energy levels, particularly of argon neutrals (Ar I) and ions (Ar II) and of aluminium ions (Al II), were found to be much more concentrated at distinct regions, which correspond to spokes. However, the employed camera could only produce a single picture per discharge pulse, which means that the authors had to compare the filtered emission from different discharge pulses with each other. This makes it very challenging to judge which part of the spoke produces increased light emission for a given optical filter and the corresponding emission line.

Hecimovic used a setup of multiple cameras and beam splitters to record the plasma emission of HiPIMS spokes with four cameras simultaneously [30]. Each of the cameras was combined with a different optical filter, making OES of the same spoke possible. With this setup, the author found the sharp trailing edge of the spoke to mostly contain Ar I and Ar II emission, while the leading edge of the spoke was dominated by Al I emission. However, the setup was limited to four cameras, which means that no attempt was made at recording the emission of Al II. Additionally, this approach reduced the amount of light which could reach each of the cameras, leading to a poor signal-to-noise ratio.

In this work, we synchronise the camera to the movement of spokes, enabling us to record the average spoke emission over hundreds of discharge pulses. The synchronisation is similar to our approach used in recent probe measurements, which were performed under precisely the same discharge conditions, enabling us to directly compare both measurements [31]. This synchronisation scheme creates the picture of an average spoke which is highly reproducible. Performing this synchronised measurement with different optical filters in the beam path, allows us to investigate the different emission components which contribute to the overall appearance of the spoke, while maintaining an excellent signal-to-noise ratio. This approach allows us to not only investigate spokes in HiPIMS, but also the much less bright spokes in DCMS discharges.

2. Experimental setup

2.1. Discharge

Figure 2 illustrates the experimental setup. The employed vacuum chamber was evacuated to a base pressure of $10^{-6}$ Pa. Argon was used as the working gas at a flow rate of 41 sccm, resulting in a pressure of 0.5 Pa. The 2 inch magnetron (Thin Film Consulting IX2U) was inserted horizontally into the chamber. A round aluminium target with a diameter of 50 mm and a thickness of 3 mm was used. Due to the magnetic arrangement of the magnetron, the racetrack had a radius of about $r = 13.5$ mm. A metal ring was used as the anode cover, providing mostly unobstructed optical access to the region close to the target surface, when imaging the plasma from the
field configuration. An anode cover ring is situated at about $z = 4$ mm infront of the target. The cylindrical coordinate system, used to denote positions in the discharge is also shown. The coordinate $r$ points in radial direction with $r = 0$ being the middle of the target and $z = 0$ denoting the target surface itself. $\phi$ points in azimuthal direction.

side of the magnetron. Figure 1 shows a sketch of the target region, including the anode cover and the magnetic field configuration, analytically reconstructed from measurements (using the method from [32]).

A cylindrical coordinate system, shown in figure 1 is used to denote positions in the discharge. The coordinate $z$ is normal to the target surface, with $z = 0$ denoting the surface itself. The coordinate $r$ points in the radial direction with $r = 0$ being the middle of the target and $\phi$ points in azimuthal direction.

For HiPIMS, current and voltage were measured in the cable between magnetron and power supply (Trumpf Hüttenger TruePlasma Highpulse 4002) using commercial probes (Tektronix TCP404XL + TCPA400, Tektronix P6015A). An additional inductance was present between the power supply and the magnetron to increase the discharge stability by limiting large current changes. Details on this inductance can be found elsewhere [33]. The pulse parameters were set to a length of 100 $\mu$s, with a repetition rate of 30 Hz. The applied voltage was 670 V, which reduced to about 570 V after breakdown. Current onset was after about 18 $\mu$s, reaching a peak current of 40 A or a peak target current density of 2 A cm$^{-2}$ at the end of the discharge pulse. The peak target area normalised power density was 1.15 kW cm$^{-2}$. The current waveform can be found in a recent publication using the same discharge parameters [31].

For DCMS, a different power supply was used (Trumpf Hüttenger TruePlasma Highpulse 4001 G2) and the discharge was excited without the external inductance between power supply and magnetron. Voltage and current were again measured in the cable (Elditest CP6550, Tektronix P6015A). The discharge was operated at 305 V, yielding a current of 60 mA or a target normalised current density of 3 mA cm$^{-2}$.

2.2. Flat probe and photomultiplier

The spoke synchronisation was carried out using either a photomultiplier (PMT) or a flat probe. Both diagnostics show a peak in the signal whenever a spoke moves past the monitored region. These peaks were used to trigger the camera, thus providing synchronisation between spoke appearance and diagnostic. The flat probe measurements introduced a larger delay and jitter than the PMT; because of the time ions need to reach the probe which was located outside of the magnetic trap. Therefore, the PMT was used to create the trigger signal in the case of HiPIMS. However for DCMS the spokes were not bright enough and the flat probe was used instead.

PMT and camera observed the discharge via different windows, as indicated in figure 2. The PMT was combined with an optical fiber and lens ($f = 135$ mm) to limit the field of view to a small part of the racetrack. The field of view had a diameter of about 0.8 mm on the target surface. The PMT was connected to an oscilloscope (Teledyne Waverunner 8404M-MS) and terminated with 500 $\Omega$, to provide a strong enough signal while maintaining a fast time response. The oscilloscope then created a trigger signal for the camera.

For DCMS, the emission intensity is much lower, increasing the noise in the PMT signal and rendering the triggering on this signal unreliable. Therefore, a flat probe, positioned at the side of the magnetron, was used to monitor the ion saturation current of the discharge. The probe potential was set to $-40$ V (compared to ground), yielding strong fluctuations in the ion saturation current whenever a spoke moved past the probe [34]. The flat probe setup has been described in detail in previous publications [15, 27]. The probe was connected to the oscilloscope (Teledyne Waverunner 8404M-MS) which created the trigger signal.

2.3. Camera and optical filter

The camera (Andor iStar sCMOS 18U-E4) was arranged to observe the discharge from two different positions, as indicated in figure 2. In the head-on position, the camera is pointed directly at the target surface, enabling us to observe the $r-\phi$ plane of the discharge. When the camera is instead positioned in the side-on position, we can image the $r-z$ plane of the discharge to investigate the spatial extent of the spoke structure in the target normal direction. Considering figure 1 the $r-\phi$ plane describes the target surface, while the $r-z$ plane is depicted in the figure itself.

For the HiPIMS discharge, the gate width was 100 ns and the camera intensifier was adjusted for the different filters, to provide a good signal without oversaturating the camera. For the less bright DCMS discharge, the camera intensifier was always operated at the maximum value and the gate width was varied between 200 ns and 1000 ns to acquire enough signal without blurring the spoke movement, which might occur for longer gate widths. Images were averaged over 200 to 500 discharge pulses, again depending on the emission intensity.

Table 1 lists the wavelengths ($\lambda_{F}$) of the utilised optical filters as well as their widths ($\Delta \lambda_{F}$, FWHM) and the emitter species of the optical emission line that passes each of these filters. The table also gives the energies of the upper level of the emission line transition. This information will be important for the interpretation of the data as it indicates how much electron energy is required to excite the emitter into the corresponding level. For argon neutrals, different filters were utilised depending on the discharge operation: for DCMS a
Figure 2. Illustration of the experimental setup. The spoke movement was monitored with either a photomultiplier tube (PMT) or a flat probe, for HiPIMS and DCMS operation, respectively. The signal from either the PMT or the flat probe was analysed by the oscilloscope. Whenever a spoke moved past the monitored region, the oscilloscope created a trigger signal which was sent to the camera. The camera was either positioned facing the target (head-on) or observing the discharge from the side (side-on). Optical filters were positioned between plasma and camera to measure different emission components separately.

Table 1. Properties of employed optical filters and corresponding emission lines. For the filters, the central wavelength ($\lambda_F$) and width of transmission ($\Delta\lambda_F$) is presented (FWHM). The energy of the upper level of the corresponding transition line, $E_i$, was taken from NIST [35]. In case multiple lines pass through the filter, the energy of the transitions was comparable and the entry corresponds to the most prominent line.

| Species | $\lambda_F$ (nm) | $\Delta\lambda_F$ (nm) | $E_i$ (eV) |
|---------|-----------------|------------------------|------------|
| Ar I    | 750             | 1                      | 13.5       |
| Ar I    | 760             | 10                     | 13.2       |
| Ar II   | 488             | 3                      | 19.7       |
| Al I    | 396             | 3                      | 3.1        |
| Al II   | 560             | 9                      | 15.5       |

A filter around 760 nm was used. Multiple strong Ar I lines pass through this filter, yielding a strong signal, which is important for the faint DCMS discharge. However, the emission of metal species becomes much stronger in HiPIMS, leading to a mixture of aluminium and argon lines to pass the 760 nm filter. Thus, a more narrow filter around 750 nm was used for HiPIMS operation.

2.4. Spoke synchronisation

For the HiPIMS operation, a PMT was used to detect spokes moving past a certain position of the racetrack. Whenever a spoke moves past the PMT’s field of view, a peak in the PMT signal is created, as recorded by the oscilloscope. When the oscilloscope is triggered, the trigger signal is also relayed to the camera, which acquires an image of the spoke located at the position that the PMT monitors.

In order to create an accurate trigger signal, the advanced trigger options of the oscilloscope were utilised. This is illustrated in figure 3 and involved two steps:

(i) Initially the scope received a trigger signal produced by the magnetron power supply, indicating the beginning of the discharge pulse. Once the scope received this signal, a hold-off time of 90 $\mu$s began, ensuring that the spoke, which was going to be triggered upon, occurred at the end of the 100 $\mu$s discharge pulse.

(ii) The second condition for the trigger process was a rising flank in the PMT signal with a preset threshold intensity. After the hold-off time the scope waited until a spoke signal rose strong enough to surpass this threshold. Only then did it trigger a measurement and sent a trigger signal to the camera.

Since the windows of the reactor chamber became constantly coated the longer the measurements progressed, the PMT-signal level reduced with time. All measurements were therefore performed in rapid succession, ensuring a negligible change in the signal’s intensity.

To ensure that the trigger process does not create a significant delay between spoke detection and image acquisition, the speed of the trigger process was examined. The process creates a delay of approximately 100 ns and a jitter of up to 50 ns. Given the average signal time of a spoke, which is in the $\mu$s range, this speed is sufficient.

To investigate the trigger process further, we compared spoke triggered images and single-shot images of HiPIMS spokes to each other. For this comparison, the contrast of the different spoke signals was evaluated. Since the spoke intensity varies stochastically from one pulse to the next, the contrast depends sensitively on the set trigger level. A higher trigger level leads to a higher contrast, since small spokes with intensities that do not exceed this level are skipped. However, if the trigger level is too high, no spoke within the 10 $\mu$s trigger windows between the hold-off time and the pulse end might have the required intensity. In this case, no image is acquired during the discharge pulse and the acquisition is
Figure 3. Illustration of the trigger process for the HiPIMS discharge. The time axis is relative to the beginning of the discharge pulse. Initially, the scope awaits the magnetron power supply to signal the beginning of the discharge pulse. Once this signal is received, the scope waits for a predefined hold-off time after which a rising flank above the trigger threshold will cause the trigger to fire. The scope then sends the trigger signal to the camera.

instead reset for the next pulse. Consequently, the measurement time increases as the trigger level is increased, since the oscilloscope begins to skip more and more pulses.

The contrast values of single-shot measurements and spoke-triggered measurements were calculated by dividing the maximum intensity of each image by the respective minimum. In comparison, the spoke-triggered measurements still retained a good 40% to 80% of the single-shot contrast, depending on the chosen trigger threshold. For the best compromise between measurement time and contrast, the trigger threshold was kept at a level that ensured a 75% contrast. This trigger level is shown in figure 3. In this case, almost no discharge pulse was skipped.

For DCMS, the PMT signal was not strong enough to produce a reliable trigger signal. Therefore, the spoke induced fluctuations in the ion saturation current onto a flat probe were used. A different trigger option of the oscilloscope was then used, as illustrated in figure 4. In this mode, the oscilloscope is only triggered, if the observed signal is above the trigger level for a specified length of time. The oscilloscope channel was used in AC mode to remove the average ion saturation current and only retain the fluctuations caused by the spokes. The trigger level was then set to 0 V, so that the scope triggered if the ion saturation current was a certain time above the average and then fell below the average value. The trigger option was set to be active for fluctuations widths between 24.9 μs and 25.1 μs, which was about the mean width of spoke fluctuations under these conditions.

3. Results and discussion

We will first discuss the results for the HiPIMS discharge. Recently published probe measurements, performed for HiPIMS under the same conditions, will guide our interpretation of the results [31]. We will then move to the results for DCMS and lastly compare both discharge conditions to draw general conclusions about the spoke phenomenon.

3.1. HiPIMS

Figure 5(a) shows a single-shot image of three HiPIMS spokes in front of the aluminium target, observed from the head-on configuration. The emission intensity is shown as a false colour image, with the intensity scale normalised to the maximum. The picture shows the spectrally integrated plasma emission without an optical filter in the beam path. The image was recorded at the end of the discharge pulse, 95 μs after its onset. The gate width was 100 ns. An arrow in the figure indicates the $\vec{E} \times \vec{B}$ direction of the discharge configuration, which is also the direction of spoke movement for spokes in HiPIMS.
Figure 5. False colour image of the plasma emission resulting from spokes rotating in front of an aluminium target. The white circle indicates the edge of the target. (a) Single-shot image of the spokes in front of the aluminium target. It was recorded using the same delay and gate width as the averaged picture. Three triangular spokes are distinctly visible. (b) Image averaged over 250 pictures using the trigger setup. Only a single spoke intensity maximum is fully maintained, while all other spoke maxima are strongly attenuated due to the fluctuations in the spoke mode number.

In contrast, figure 5(b) shows an average of 250 pictures recorded using the described trigger scheme. The hold-off time was set to 90 µs and the gate width was again 100 ns. The figure now displays only a single average spoke at the top of the racetrack. This is the position monitored by the PMT, used to create the trigger signal. Evidently, the trigger and averaging process sustains the image of only a single spoke, while attenuating all other spokes. This is caused by changes in spoke mode number from one discharge pulse to the next: changes in the mode number also cause changes in the distances in between the spokes. Therefore these additional spokes leading and trailing the triggered spoke will be averaged out.

Comparing this average spoke to the single-shot picture of figure 5(a), we find that the average spoke shows a less distinctive asymmetry and exhibits a less abrupt emission intensity decrease at the trailing end of the spoke. This more diffuse shape is presumably caused by variations in spoke shape and size from one pulse to the next and cannot be avoided when averaging over many spokes. However, the general shape and size of the structure is well preserved by our approach. It should be emphasised that this average spoke picture is highly reproducible and appears the same from one measurement to the next and from day to day, as long as the same discharge parameters are adjusted. This enables us to perform multiple comparable measurements with different optical filters in the optical path.

3.1.1. Filtered emission. To compare the shapes of the different emission lines more conveniently, we transformed the images to polar coordinates, as previously done by Biskup et al [4] and Panjan and Anders [19]. The polar transformed image of the unfiltered emission is shown in the bottom of figure 6(a). The azimuthal axis $\phi$, points in $-\vec{E} \times \vec{B}$-direction, with $0^\circ$ starting at the bottom of the racetrack and reaching the trigger position, at the top of the racetrack, at $180^\circ$.

The transformed results of the filtered measurements are plotted in figure 6(b). A dotted line was drawn at $180^\circ$, indicating the position monitored by the PMT creating the trigger signal. In this representation the spokes move from right to left in $\vec{E} \times \vec{B}$ direction, defining a leading edge on the left and a trailing edge on the right side of the emission maximum.

Comparing the emission passing the different optical filters, presented in figure 6(b), we see some noteworthy differences. Considering the spatial variation of the Al I emission, it seems that its behaviour is characterised by a minimum in intensity rather than an emission maximum as observed in the unfiltered emission. This minimum is located at $205^\circ$ and, thus, slightly behind the trailing edge of the spoke. An additional intensity maximum can be observed at the leading edge of the spoke at $160^\circ$. However, this maximum is rather weak compared to the more distinctive minimum at the trailing edge.

This emission intensity minimum observed for the Al I emission is rather unusual, as spokes are usually identified as a maximum in the emission intensity. However, the same behaviour was recorded by Hecimovic, while investigating the same wavelength with a four-camera setup [30]. Equally, the measurements by Andersson et al also reveal diffuse areas of higher intensity that are intersected by sharp minima in emission intensity, for the same emission lines [26, 36].

We find two possible explanations for this strong Al I intensity minimum right behind the trailing edge of the spoke:

(i) An increased ionisation rate inside the spoke causes a reduction of aluminium neutral density after the spoke has past.

(ii) Since the energy of the upper emission level with 3.1 eV is rather small, most electrons are able to populate it. Thus, the emission pattern closely follows the electron density rather than the electron temperature, as one would usually expect in low temperature plasmas.
Figure 6. (a) The top of the figure shows the unfiltered average spoke created by triggering the camera on the spoke appearance and then averaging over 500 acquisitions. The edge of the target is indicated by the white circle. The arrow indicates the $\vec{E} \times \vec{B}$ direction which is also the direction of spoke propagation. Focusing on the top of the racetrack the spokes therefore move from right to left. The bottom part of the figure shows the same image in polar coordinates. (b) Spoke triggered and averaged images for different optical filters between plasma and camera. The vertical line marks the $180^\circ$ position at the very top of the racetrack.

To investigate these two possibilities we consult our recent probe measurements, which were performed under exactly the same discharge conditions [31]. Figure 7 shows the emission of the Al I line as a line graph, obtained by integrating the coordinate transformed image from figure 6(b) over the radial axis. For comparison, the figure also shows electron density and temperature measured with a Langmuir probe positioned 8 mm above the racetrack centre ($r = 13.5$ mm, $z = 8$ mm). Using the spoke velocity $v_s$ obtained from the probe measurements, the time axis of the probe data was converted to azimuthal position using:

$$\varphi(t) = \left(t - t_{ref}\right) \frac{360}{2\pi R} v_s + \varphi_{ref}$$  \hspace{1cm} (1)

with the racetrack radius, $R = 13.5$ mm and the reference time $t_{ref}$ and angle $\varphi_{ref}$ marking the position of the spoke in space and time, as determined by the synchronisation approaches for the probe and the camera measurements, respectively. Since the probe was located at the $\varphi = 180^\circ$ position and both measurements were synchronised using a photomultiplier, the measurements should correspond to equivalent average spokes at the same location.

Comparing the Al I emission to the probe results presented in figure 7, we find that the maximum in emission actually occurs before the peak in electron temperature and density. The emission peaks at $165^\circ$ (left dotted line) and then decreases while electron density and temperature still increase into the spoke structure. Once density and electron temperature have also peaked around $190^\circ$, the decrease in Al I emission becomes even more steep. The distinctive minimum in emission intensity coincides with the strong electron density minimum at the trailing edge of the spoke, at $205^\circ$ (right dashed line).

The initial decrease in emission intensity between the intensity maximum at $165^\circ$ and about $190^\circ$ is observed, although both electron density and temperature are still increasing, which should lead to an increase in excitation rate. An emission intensity decrease simultaneously to an excitation rate increase points towards a decrease in emitter density: the aluminium neutral density decreases so rapidly, that the increased excitation rate is apparently overcompensated. This neutral depletion is likely caused by strong ionisation, in agreement with Anders’ et al [28] interpretation of spokes as ionisation zones.

For positions $>190^\circ$, electron density and temperature also begin to decrease, resulting in a slightly steeper decrease in Al I emission intensity, as the excitation rate reduces. After the minimum, the emission intensity increases again towards the leading edge of the next spoke structure, following behind.

The interpretation of the spatial profile of the aluminium neutral lines shows that the line indeed closely follows the electron density but is also strongly affected by depletion of the aluminium atom density caused by ionisation. The strong
electron temperature gradient or a gradient in the ionisation radial direction between ions and neutrals might indicate an emission line at 394 nm. This difference in the width in

**Figure 7.** Comparison between the emission of Al I and Al II and the electron density and electron temperature measured using a Langmuir probe (adapted from [31]). The maximum and minimum in emission intensity are indicated by vertical lines. The reduction in emission intensity during the spoke is caused by both the depletion of neutral density by strong ionisation and the sudden drop in electron density at the trailing edge of the spoke.

intensity minimum at the trailing edge of the spoke is a consequence of the combination of both of these processes.

This observation is supported by further probe measurements that were performed with titanium and chromium as the target material (not shown here). For these materials, symmetrical spokes were observed with the camera, without the distinctive sharp edge at the trailing end of the spoke. In the probe measurements, the electron density fluctuations were then also symmetrical, indicating that it is indeed the asymmetric variation in electron density that causes the distinctive triangular spoke shape.

Considering the emission of Al II as shown in figure 6, we find a clear maximum at the trigger position of 180°. Since this emission originates from an upper energy level of 15.5 eV, this maximum corresponds to the peak in electron temperature found by the probe measurements, as can be seen in figure 7. Another remarkable difference to the spatial emissions profile of Al I is the width that the emission occupies in the radial direction. While the emission of Al II is very localised to the racetrack centre, since the density of emitters is higher. This explains the difference between the spatial emission of argon ions and neutrals. The maximum in Ar II emission in azimuthal direction is located between the emission maxima of Al II and of Ar I. Again, the reason behind this can be found in the spatial extent of the emission in the axial or target–normal direction. Our head-on measurements necessarily perform a line-of-sight integration over this direction. We, therefore, turn to the side-on measurements, which can provide further insight.

### 3.1.2. Differential emission imaging

**Figure 8(a) shows the spoke synchronised measurements from the side-on position, averaged over 200 discharge pulses and without filter in the optical path.** The measurement was performed with an additional delay of 3 μs between spoke detection and image acquisition. During this time, the spoke moves from the 180° position, (compare figure 6) to 90°, so that we observe the side of the spoke. The spoke movement direction is marked in the figure. An emission intensity minimum can be observed at about z = 4 mm, which is caused by light obstruction the anode cover ring (see figure 1).

Despite the synchronisation on the spoke movement, a localisation of emission around 90° is hardly visible in figure 8(a). As the measurement collects light from both sides of the racetrack simultaneously, the strong background emission may conceal the increase in emission caused by the spoke nearest to the camera. To separate the emission of the spoke degree, with a higher electron temperature or ionisation degree located in the centre of the racetrack leading to increased emission from Al II. The higher excitation rate in the racetrack centre could be caused by secondary electrons ejected from the cathode preferentially at this region of strongest sputtering [37].

Regarding the Ar I emission in radial r-direction—rather than in azimuthal φ-direction—it exhibits a remarkably broad profile along the radius. Although the energy of the upper level 13.5 eV is close to the energy of the aluminium ion level, the spatial emission profile for Ar I is much wider. This might be caused by a larger ionisation degree at the centre of the racetrack or by rarefaction taking place in front of the target, which reduces the argon density especially in front of the racetrack’s centre. These effects counteract the increased excitation rate at the centre of the racetrack and can lead to the observed wide profile. The peak in the Ar I emission component is located to the right of the trigger position at 180° and appears at the trailing edge of the spoke. We will later find that a large part of the emission originates from argon neutrals that are located comparatively far away from the target surface, as will be discussed below.

Regarding the Ar II emission in radial direction, we find a narrow emission profile localised at the racetrack centre, in contrast to the emission of Ar I. For the Ar II emission, three processes seem to be relevant: rarefaction can still play a crucial part in the density of argon ions, however the centre of the racetrack will feature both an increased excitation rate, as well as a higher degree of ionisation. The larger degree of ionisation leads to a more centralised ion emission at the racetrack centre, since the density of emitters is higher. This explains the difference between the spatial emission of argon ions and neutrals. The maximum in Ar II emission in azimuthal direction is located between the emission maxima of Al II and of Ar I. Again, the reason behind this can be found in the spatial extent of the emission in the axial or target–normal direction. Our head-on measurements necessarily perform a line-of-sight integration over this direction. We, therefore, turn to the side-on measurements, which can provide further insight.
from this background, the spoke-triggered measurements are combined with pulse-triggered, averaged measurements shown in figure 8(b). These measurements are performed at 95 μs after the beginning of the discharge pulse, without spoke synchronisation. Averaging over many consecutive pulses will average out the spoke intensity variations. If this pulse-triggered and averaged image is now subtracted from the spoke synchronised measurement (a), the emission variation caused by the spoke is recovered.

This yields an image of differential emission, as shown as figure 8(c). This differential emission shows the regions that are brighter or fainter than the average in the presence of the spoke. We, therefore, use a diverging colour map that shows regions of higher than average emission in red and regions of lower than average emission in blue.

In figure 8(c), the spoke is now clearly visible as the red region of increased light emission. Because of the 3 μs delay between spoke detection and image acquisition, the spoke is located at around 90° and is observed from the side. In this graphical presentation, the spoke maintains the asymmetric shape with a slow intensity increase at the leading edge (bottom) and a more sudden drop in emission intensity at the trailing edge (top). Additionally, the structure appears to be tilted backwards by about 6°. This tilt might represent the drift direction of electrons as they move through the spoke. The azimuthal E-fields around the spoke lead to a weak drift component in axial direction, in addition to the mostly azimuthal drift caused by the combination of $\vec{E} \times \vec{B}$, diamagnetic and curvature drifts.

The side-on measurements were performed for the same set of optical filters between plasma and camera as the head-on measurements. The spoke averaged emission (figure 8(b)) was obtained for each filter separately, since the different emission lines naturally also feature spatial differences not caused by the spokes [34].

3.1.3. Differential emission maps, side-on. To compare the emissions of several species in a single figure, we performed further post processing. For each species, differential emission images were created as described above. Then regions with an emission intensity increase of less than 10% were removed, to only show the regions of increased light emission in the presence of the spoke. Every emission line was assigned a different colour and the opacity was adjusted to reflect the emission intensity. This way multiple species can be presented in a differential emission map that shows which species’ emission intensity is increased in the presence of a spoke. Importantly, the map also reflects the amount and position of the intensity’s increase. However, the intensity modulation for each species is normalised individually so that the relative intensity modulation strengths between the species are not preserved (for example, the broad, but not very strong peak in the Al I emission will appear as strong as the very distinct peak in Al II emission—compare figure 6—which should be kept in mind). Additionally, the distinct minimum of the Al I emission cannot be displayed in this presentation.

Figure 9 shows the differential emission map for the side-on position. The outline of the spectrally integrated spoke emission is plotted as a black dotted line and the light grey background shows the averaged plasma emission, without spoke synchronisation.

Figure 9(a) presents the emission increase of Al I and Al II. Because aluminium ions are only excited by highly energetic electrons, the Al II emission increase is located between
Figure 9. Differential emission maps for the side-on position. The contour plot indicates an increased emission in presence of a spoke, compared to the average plasma emission. High differential emission is assigned higher opacity and, thus, a more intense colour. The pictures were taken using a delay of 3 μs between spoke detection and camera acquisition, in which time the spoke moved to the 90° position (compare figure 2). Thus, the spoke is observed from the side and is moving downwards in the image. The unfiltered spoke shape is plotted as a black dotted line. (a) Differential emission of Al I and Al II emissions. The Al I emission peaks at the leading edge of the spoke, while the Al II emission is increased in the spoke centre and at the trailing edge. (b) Differential emission of Ar I and Ar II emissions. The argon emissions make up the trailing edge of the spoke. The Ar I emission reaches very far into the reactor volume, rising up to 18 mm.

The nature of the spoke as an ionisation zone becomes particularly apparent, as the Al II emission only begins to increase as the emission of Al I is beginning to faint. The sharp trailing edge of the unfiltered spoke emission (black dotted line) coincides with the strong reduction in Al II emission, indicating that these ions are partly responsible for this distinctive feature. In fact, the strong reduction in electron density at the trailing edge of the spoke indicates a reduction of both electron and ion density, since the electric fields inside of a spoke are not strong enough to violate quasi neutrality [31]. Despite the electron energy being high enough to excite the upper level of the transition, the decrease of aluminium ion and electron density predominates. This reduction of both densities leads to a sharp decrease in the emission intensity of Al II at the trailing edge of the spoke as was visible in figure 6.

Figure 9(b) displays the emission increase of both Ar I and Ar II. Both emissions peak at the trailing edge of the spoke, following the course of the electron temperature since higher energy electrons are required to excite both argon species. The Ar I emission increase reaches far from the target surface up to the last closed magnetic field line of the magnetic trap region at about z = 18 mm. This would indicate that spokes are not located exclusively close to the target surface but extend over the whole magnetic trap region. This is not always visible since the emissions originating from the target surface are much brighter so that the region close to the magnetic null remains obstructed. In the measurements of Andersson et al [26], so called flares were reported which caused increased light emission far beyond the magnetic trap region. These features were not observed here, likely because the shape of these flares seems to be very variable so that they are removed by averaging over many spokes.

The large spatial extent of the Ar I emission in target normal direction also explains why the neutral emission seemingly lags behind the Ar II emission in the head-on measurement shown in figure 6: the tilt of the spokes causes the emissions at greater distances from the target surface to appear as if they were produced at or after the trailing edge of the spokes. However, the side-on measurements reveal that both Ar I and Ar II feature increased emission at the same azimuthal position, located around the maximum in electron temperature at the trailing edge of the spoke.

The large spatial extent of the light increase in target–normal direction caused by the spokes becomes even more apparent when the spoke is observed from yet another angle. To this end, measurements from the side-on position were also performed without the time delay (Δt = 0) between spoke detection and camera acquisition. In this case, the spoke is located at the 180° position and viewed from the side-on position the spoke is now moving towards the camera. The
differential emission maps of this configuration are shown in figure 10, where the spokes are now, accordingly, moving out of the paper plane. The figure additionally shows the magnetic field lines, which were reconstructed from measurements according to the method of Krüger et al [32]. The weak emission at the 0° position—or \( r = -5 \) to \( r = -15 \) mm in figure 10—is caused by all spokes, simultaneously present on the racetrack, which were not completely averaged out.

Figure 10(a) shows the differential emission increase for Al I and Al II. The differential emission in radial direction, as shown here, is difficult to interpret: while our post processing approach of subtracting the average emission from the spoke triggered images overcomes the line integration over the background emission, line of sight integration over the complete spoke structure is still performed. Since in figure 10, we are looking at the tip of the advancing spoke structure, line of sight integration is performed over both the moderate emission maximum in the Al I emission, as well as the more distinct emission intensity minimum at the trailing edge of the spoke. Consequently, any spoke induced light intensity modulation is actually removed for the Al I emission, which is only shown in figure 10(a) for the sake of completeness.

The Al II emission is similarly affected by the problem of line-of-sight integration, as the species also feature an intensity minimum after the spoke. However, the minimum is less pronounced, so that the emission increase remains visible. As from the other measurements, the Al II emission is located close to the target and at the racetrack centre, where an elevated electron temperature and an increased ionisation degree are suspected.

For most species, an additional intensity increase can be observed at the 0° position. This is caused by additional spokes present on the racetrack, which are mostly, but not completely averaged out by our synchronisation approach. These features are also visible from the head-on position, but only in the differential emission, where the background emission is removed and much smaller light intensity changes become visible.

Figure 10(b) shows the spoke induced emission intensity increase for Ar I and Ar II. The most striking aspect is that the emission of Ar I is bent inwards, closely following the curvature of the magnetic field lines. The emission increase nearly extends to the magnetic null position at a target distance of about 18 mm. Since the intensity increase strictly follows the magnetic field lines, it likely marks the actual spoke structure, and not flares. This would mean that the electron density, electron temperature and potential fluctuations caused by spokes might also extend throughout the entire magnetic trap region, instead of being mostly located close to the target surface, as the unfiltered emission might imply.

This large extent of spokes in axial direction would explain why measurements of spoke induced electron density fluctuations at the target surface [38], the middle of the magnetic trap [31] and at the magnetic null [39] all found a very similar modulation strength of about 25%.
Figure 11. Differential emission maps for the head-on position. The contour plot indicates an increased emission in presence of a spoke, compared to the average plasma emission. High differential emission is assigned higher opacity and, thus, a more intense colour. The shape of the unfiltered differential emission is plotted as a black dotted line. The shape of the average plasma emission is indicated as a grey background. (a) Differential emission of Al I and Al II emissions. The Al I emission rises first and fills out the rising slope of the spoke emission very broadly. The Al II emission follows suit making up the centre of the spoke emission. (b) Differential emission of Ar I and Ar II emissions both located at the trailing edge of the spoke. Ar I reveals an increased intensity in the lower left of the picture. This appearance is due to the Ar I spoke emission being relatively low compared to the attenuated signal (compare figure 6(b)). The signal of other spokes being present on the racetrack is therefore not entirely averaged out.

3.1.4. Differential emission maps, head-on. While we have already discussed the head-on measurements without subtracting the average plasma emission, it is still useful to also create differential emission maps for this observation direction. This helps to increase the contrast of the images and facilitate the comparison of the different species more easily.

Thus, figure 11(a) shows the differential emission for the head-on measurements of Al I as well as Al II. As before, the unfiltered differential emission is plotted as a black dotted line. While the Al I emission is mostly characterised by the strong minimum at the trailing edge of the spoke (compare figure 6), figure 11(a) now only shows the regions of increased emission intensity. For Al I, this increased emission intensity is located broadly at the leading edge of the spoke and its edges. In contrast, the emission intensity increase of Al II peaks at the very centre of the spoke, both radially as well as in azimuthal direction. At this position, we expect a combination of high ionisation degree, electron density and electron temperature, which is necessary to first ionise the aluminium atoms and then excite the upper level of the transition (15.5 eV). The Al I emission, in turn, is strongly reduced in the centre of the spoke (not visible in this representation). We explained this with the depletion of aluminium neutral density by rapid ionisation as well as with the sudden decrease of electron density at the trailing edge of the spoke. That the Al I emission is only reduced at the (radial) centre of the spoke indicates that the emission reduction is at least partly caused by strong ionisation. At the sides of the racetrack, both the electron density as well as the electron temperature appear to be lower than at the centre. In turn, this leads to a lower ionisation rate than at the centre of the racetrack and the Al I emission intensity remains high.

Figure 11(b) shows the emission intensity increase of Ar I and Ar II. Ar I reveals an increased intensity in the lower left of the picture, which is due to the Ar I emission caused by the triggered spokes being relatively low compared to the attenuated signal (compare figure 6(b)). The signal of the other spokes present on the racetrack is therefore not entirely averaged out.

Regarding the emission at the top of the racetrack both species show increased emission at the trailing edge of the spoke, where the electron temperature is higher. The Ar II emission is larger at the radial centre of the spoke and towards the trailing edge. The emission of Ar I seems to peak after the trailing edge of the spoke. As discussed above, this is mostly an artefact created by line-of-sight integration over the backwards tilted spoke structure in combination with the far reaching emission intensity increase of Ar I in axial direction. Additionally, the Ar I emission is distributed much more broadly in radial direction. This is partly due to the increased ionisation degree in the centre of the racetrack, which causes ion emission to be generally more localised at the middle of the racetrack. Additionally, the line-of-sight integration over the Ar I emission, which is arced along the magnetic field lines (compare figure 11(b)) creates an artificially broad profile for the head-on measurement.
3.2. DCMS

Figure 12 shows a false colour image of a spoke in a DCMS discharge, operated at low current. Under such conditions, the spoke rotates in retrograde $\vec{E} \times \vec{B}$ direction and moves much slower ($\sim 3$ km s$^{-1}$) than spokes in HiPIMS. The spoke maintains a distinctly asymmetric shape, but the more rapid emission intensity change is now located at the leading edge of the spoke.

Figure 13(a) shows the corresponding average spoke, created by triggering on the spoke appearance as detected by the flat probe and averaging over 500 acquisitions. This average spoke has lost much of the distinctive asymmetry and is considerably wider than in the single-shot image. This is partly caused by the less reliable triggering—since spokes are more difficult to detect in DCMS—and partly by the fact that the sharp edge of the spoke is less distinctive for DCMS than for HiPIMS, at least for our experimental conditions (compare figure 5(a)). The average spoke for DCMS is very broad, which prevents us from performing side-on measurements. Since the spoke occupies more than half of the racetrack, side-on measurements always suffer from line-of-sight integration over different parts of the spoke which make the evaluation of the results nearly impossible. For side-on measurements of DCMS spokes, a larger magnetron would be needed to ensure that the spoke always occupies less than half of the racetrack area.

The average spoke is now located at the bottom of the racetrack, at 30°, even tough the flat probe, used for detecting the spoke movement, was located at the top of the racetrack, at 180°, which is the same detection position as for the HiPIMS discharge. The difference in the position of the average spoke is caused by the different trigger scheme used in the case of DCMS, which effectively only fires the trigger after the spoke has completely passed the flat probe position. Nevertheless, the position of the spoke is fixed on the racetrack, and enables us to compare the different emission components by bringing optical filters in the beam path.

3.2.1. Filtered emission. Figure 13(b) shows the coordinate transformed images filtered for the different emission components. The spoke moves in retrograde $\vec{E} \times \vec{B}$ direction as indicated in the coordinate transformed pictures. As for the HiPIMS discharge, a shift between the different components is noticeable, in particular for the emission of Al II, which seem to precede the other emission components. The comparison however, is impeded by the broad spoke structure, which makes it difficult to judge the positional shift of the different emission components. Thus, we will again subtract the averaged emission without synchronisation, to create differential emission maps, as we did for the HiPIMS results.

3.2.2. Differential emission maps, head-on. Figure 14(a) shows the differential emission of Al I and Al II. The spectrally integrated differential emission is shown as a black dotted outline. The Al II emission increase is mostly located at the leading edge of the spoke, where the sharp emission intensity variation is observed in the single-shot picture (figure 12). The emission intensity increase of Al I is, in contrast, entirely located at the trailing edge of the spoke and makes up the slow, elongated emission intensity variation observed there.

Figure 14(b) shows the emission intensity increase of Ar I and Ar II. The emission increase for both species is located in the azimuthal centre of the spoke structure, indicating an increased electron temperature. The argon neutrals are located slightly more towards the trailing edge of the spoke. At the leading edge of the spoke structure, the emission of argon species is preferentially increased at the sides of the racetrack. This causes the substantial widening of the emission structure in radial direction in the presence of the spoke (compare figure 12).

The different emission features leading to the overall appearance of a spoke in DCMS indicate a similar internal structure compared to spokes in HiPIMS: a large electron energy at the sharp edge of the spoke and a more gradual decrease of electron density causing the gently intensity variation at the other side. While the spokes are moving in different directions, the excitation dynamics are likely mostly caused by the azimuthal drifts of electrons through the spoke structure. With estimated drift velocities in the order of 100 km s$^{-1}$, the spoke is basically stationary relative to the movement of electrons in both DCMS and HiPIMS. Thus, the propagation direction of the spoke plays a minor role.

3.3. Electron dynamics

In order to understand the emission structure of the DCMS spoke, we may consult the probe measurements by Panjan and Anders who combined an emissive probe and a floating probe to measure both the plasma potential as well as the floating potential [19]. The difference of both potentials was also used as an measure of the electron energy. The authors found a sharp increase in the plasma potential at the sharp leading edge of the spoke. Close to the target, the potential increased from about $-80$ V to nearly zero. This potential increase was also accompanied by an equally sharp increase in electron energy. Both potential and electron energy then decreased much more slowly until the beginning of the next spoke.
Figure 13. (a) The top of the figure shows the unfiltered average spoke created by triggering the camera on the spoke appearance and then averaging over 500 acquisitions. The edge of the target is indicated by the white circle. Arrows indicate the $\vec{E} \times \vec{B}$ direction as well as the direction of spoke propagation. The bottom part of the figure shows the same image in polar coordinates. (b) Spoke triggered and averaged images for different optical filters located between plasma and camera.

The internal potential and density structure we propose based on our OES results and the probe measurements of Panjan and Anders is shown in figure 15. Electrons following the different drifts ($\vec{E} \times \vec{B}$, diamagnetic, curvature) enter the spoke structure at the sharp edge and instantly gain energy through the change in plasma potential. In the figure this is indicated by the change in arrow colour from blue (cold) to red (hot/energetic). As electrons cross the jump in potential, they drift downwards as the $\vec{E}_\phi \times \vec{B}$ drift points towards the target during the potential change [36]. However, electrons also continue their movement in azimuthal direction, as the about equally important diamagnetic drift and curvature drift remain unchanged [40].

The potential jump causes an electron energy increase, while the flux density is conserved. This leads to an initial electron density minimum, as the constant flux of electrons is accelerated. The hot electrons can efficiently excite aluminium ions and argon particles leading to the observed emission maxima of these species. The observation that the Al II emission seems to be located closer to the sharp leading edge of the spoke than the argon species might be caused by the (axial) downwards movement of electrons: as the aluminium ions are mostly located close to the target surface, the initial downwards movement of electrons leads to a preferential excitation of these species. However, after the potential maximum, the electric field vector is reversed and the $\vec{E} \times \vec{B}$ vector now points partly away from the target surface, leading to a slow upwards drift of electrons as they continue their movement in azimuthal direction.

At larger distances from the target, the electrons first encounter more argon ions and eventually argon neutrals, causing the emission sequence shown in figure 14. During this movement, electrons continuously lose energy, both due to their movement into regions of lower plasma potential and due to inelastic collisions with all species, causing excitation and ionisation. Both the ionisation, as well as the slowing down of electrons lead to an increase in electron density, that explains the observed maximum in Al I emission at the trailing edge of the spoke. Subsequently, as the ionisation rate plummets due to the decreased electron energy, the electron density also begins to decrease. This leads to the long, gradually vanishing emission tail of the spoke.

For HiPIMS spokes, the picture is very similar. The main difference is that the much shorter mean free path for inelastic electron collisions leads to a more rapid ionisation which causes the observed sharp edge in electron density (compare figure 7). The strong ionisation also drives the plasma potential back towards the baseline, causing the complete spoke structure to be shorter in azimuthal direction. A similar argument has been made before by both Panjan and Anders et al [20, 41]. An additional difference to spokes in DCMS is that in HiPIMS the emission is also affected by aluminium neutral depletion, due to the strong ionisation. This makes it more difficult to
understand the created emission patterns, but the basic processes leading to the distinct shape of a spoke in both DCMS and HiPIMS seem to be the same.

4. Conclusion

We presented OES of spokes in both HiPIMS and DCMS discharges. A gated and intensified camera was triggered on the movement of spokes, enabling us to perform averaging to increase the signal-to-noise ratio without averaging out the spoke signal. Placing optical filters in between the plasma and the camera revealed which part of the spoke emission was caused by which of the different excited species.

For both DCMS and HiPIMS spokes, the gradual emission variation on one side of the spoke is caused by emission from low lying energy levels of aluminium neutrals, which closely follow the evolution of the electron density in the spoke. Emission from aluminium ions and argon species occupies the centre of the spoke, where the electron temperature is high. In DCMS, Al I emission is missing from this region because the electron density is very low. In HiPIMS, the neutral density is depleted by the strong ionisation, also reducing the Al I emission.

For HiPIMS, the sharp edge of the spoke is caused by the strong drop in electron density at the trailing edge of the spoke. Probe measurements showed an electron density decrease of 50% creating an emission intensity reduction of about similar strength.

For DCMS, in contrast, we expect the electron density to not feature a fast increase at the sharp edge of the spoke, as the mean free path for inelastic collisions is large and electrons can move much further along the racetrack. Instead, the sudden increase in electric potential causes an increase in electron energy. This sudden increase in electron energy leads to an increase in the emission of all species, with the exception of Al I, which more closely represent the trend in electron density.

Summarising, the sharp edge in the emission of the spoke in DCMS is indicative of strong variations in electron energy. For HiPIMS, the sharp edge is instead indicative of a strong
variation in electron density. However, both the variations in electron density as well as electron energy are driven by the strong fluctuations in the plasma potential. Consequently, the sharp edge in the emission intensity is also, indirectly, an indication for the variation in electric potential. A more distinctive spoke shape with a more pronounced edge hints at stronger variations in plasma potential.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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