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Spoke rotation reversal in magnetron discharges of aluminium, chromium and titanium

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Abstract
The rotation of localised ionisation zones, i.e. spokes, in magnetron discharge are frequently observed. The spokes are investigated by measuring floating potential oscillations with 12 flat probes placed azimuthally around a planar circular magnetron. The 12-probe setup provides sufficient temporal and spatial resolution to observe the properties of various spokes, such as rotation direction, mode number and angular velocity. The spokes are investigated as a function of discharge current, ranging from 10 mA (current density 0.5 mA cm$^{-2}$) to 140 A (7 A cm$^{-2}$). In the range from 10 mA to 600 mA the plasma was sustained in DC mode, and in the range from 1 A to 140 A the plasma was pulsed in high-power impulse magnetron sputtering mode. The presence of spokes throughout the complete discharge current range indicates that the spokes are an intrinsic property of a magnetron sputtering plasma discharge. The spokes may disappear at discharge currents above 80 A for Cr, as the plasma becomes homogeneously distributed over the racetrack. Up to discharge currents of several amperes (the exact value depends on the target material), the spokes rotate in a retrograde $\mathbf{E} \times \mathbf{B}$ direction with angular velocity in the range of 0.2–4 km s$^{-1}$. Beyond a discharge current of several amperes, the spokes rotate in a $\mathbf{E} \times \mathbf{B}$ direction with angular velocity in the range of 5–15 km s$^{-1}$. The spoke rotation reversal is explained by a transition from Ar-dominated to metal-dominated sputtering that shifts the plasma emission zone closer to the target. The spoke itself corresponds to a region of high electron density and therefore to a hump in the electrical potential. The electric field around the spoke dominates the spoke rotation direction. At low power, the plasma is further away from the target and it is dominated by the electric field to the anode, thus retrograde $\mathbf{E} \times \mathbf{B}$ rotation. At high power, the plasma is closer to the target and it is dominated by the electric field pointing to the target, thus $\mathbf{E} \times \mathbf{B}$ rotation.

Keywords: HiPIMS, spokes, ionisation zones, $\mathbf{E} \times \mathbf{B}$ drift, spoke rotation reversal, magnetron sputtering discharges

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

1. Introduction
In a magnetron discharge, permanent magnets placed behind the target generate a curved magnetic field over the target, confining the electrons and enhancing ionisation. Magnetic field strengths are commonly up to 100 mT [1], which is sufficient to magnetise the electrons, but insufficient to confine ions due to the larger Larmor radius, comparable to the experimental chamber. Nonetheless, electrons and ions are bound by ambipolar fields [2]. The complex geometry of the magnetic field induces several electron drifts oriented in the same direction, contributing to a total electron drift velocity. Most prominent is the $\mathbf{E} \times \mathbf{B}$ drift, induced by the electric field directed to the target, produced by the negative potential applied to the target, in conjunction with the magnetic field parallel to the target. The curved magnetic field and the gradient of the magnetic field.
field towards the target induce two other electron drifts. These cumulative drifts induce a total electron drift velocity in the range 10^{-5}–10^{-3} m s^{-1} [3, 4]. In a planar circular magnetron the electron motion describes a ‘closed drift’, resulting in the formation of an azimuthal current, referred to as Hall current [5]. Typically, the Hall current density is about 10 times higher compared to the discharge current density normal to the target [5, 6].

The \( \mathbf{E} \times \mathbf{B} \) drift has been experimentally confirmed by Tomasel et al [7] by supplying electrons to a planar magnetron by an external arc. However, several publications report on spokes propagating in the opposite, retrograde \( \mathbf{E} \times \mathbf{B} \) direction. This trend has been observed experimentally in magnetic micro-discharges [8] and in magnetron plasmas [9], and as an outcome of a particle-in-cell Monte-Carlo simulation method of a magnetron plasma [10]. Ito and Cappelli [8] suggested that the retrograde rotation is due to fluctuations in plasma density, which they attribute to drift waves driven by density gradients. Pflug et al [10] and Yang et al [9] offer a qualitative explanation; the plasma density deviation is maintained by a self-sustaining positive feedback mechanism, which can be described as an ionisation wave propagating in the opposite direction to the electron drift current.

In magnetron discharges, the spokes have been observed at both low discharge powers [11], and at high powers in high-power impulse magnetron (HiPIMS) discharges [12–14]. In a DC magnetron discharge, Martines et al [11] observed spoke rotation in the direction of the \( \mathbf{E} \times \mathbf{B} \) drift, while Yang et al [9] observed rotation in the retrograde \( \mathbf{E} \times \mathbf{B} \) drift direction. The spokes observed in HiPIMS plasmas, sometimes referred to as spokes, due to the similarity with the spokes in Hall thrusters, rotate in the \( \mathbf{E} \times \mathbf{B} \) drift direction [13–15].

A comprehensive investigation of the spoke properties for different target materials and power levels is lacking. The purpose of this paper is the investigation of rotation direction, speed and mode number of spokes on a planar circular magnetron. The experiments have been performed for three target materials, Al, Cr and Ti, and for a wide range of discharge currents, from 10 mA (current density 0.5 mA cm^{-2}) to 140 A (7 A cm^{-2}), which is the upper limit before the discharge starts arcing. The data in our experiment offer the unique possibility to develop a consistent model, because spoke velocities are measured over a very large parameter space ranging from a pure argon-dominated discharge to a metal-dominated discharge for three different target materials.

2. Experimental setup

The experiments have been performed in a cylindrical chamber of 35 cm in diameter and 35 cm in height. Vacuum conditions were obtained by a vacuum turbo-molecular pump backed up by a roughing pump to obtain a base pressure of \( 1 \times 10^{-4} \) Pa. The working gas was Ar, pressure was kept at 0.5 Pa, and the 2" target materials were Al, Cr and Ti. The discharge current varied from 10 mA (current density 0.5 mA cm^{-2}) to 140 A (7 A cm^{-2}). In the range from 10 mA to 600 mA the plasma was sustained in a DC mode, and in the range from 1 A to 140 A the plasma was sustained in the HiPIMS mode. In the HiPIMS mode, the pulse duration was 200 \( \mu \)s, and the frequency was varied between 2 Hz and 300 Hz to ensure stability of the current waveform.

Figure 1(a) shows the experimental setup consisting of the 2" magnetron, a 12 flat probe array (FPA) mounted around the magnetron, and a photomultiplier tube (PMT). The PMT measures the plasma light emission. The spatial resolution was achieved by using a set of two apertures, transmitting only the light coming from an area on the racetrack of about 3 mm in diameter. Figure 1(b) shows a 3D schematic of the magnetron static magnetic field and a Hall current due to the \( \mathbf{E} \times \mathbf{B} \) drift.

The floating potential was measured by the 12 FPA consisting of 12 flat probes placed azimuthally around the magnetron, separated by 30°, covering the complete circle around the target, shown in figure 1(c). The flat probes, 3 mm in diameter, are placed at a distance of 17 mm from the racetrack and 5 mm above the target surface, as shown in figure 1(d). Each probe measures the floating potential \( V_f \) oscillations during the pulse independently. Within a single HiPIMS pulse the 12 \( V_f \) signals are measured using twelve 14-bit AD converters, with an acquisition rate of 100 MS s^{-1}. The floating potential is used as a fingerprint for the spikes, since the correlation between the periodicity of the floating potential oscillations and the spoke rotation has been established previously [16].

In a HiPIMS pulse, the plasma is ignited from Ar gas, sometimes aided by sputtered metal ions remaining from the previous pulse, depending on the target material, pressure and repetition frequency [17]. In contrast to a DC discharge where the plasma density is in a steady state, during a HiPIMS pulse \( V_f \) exhibits a very dynamic behaviour [18]. \( V_f \), measured in front of the target, drops to a negative value during the pulse ignition phase, which can be explained by the presence of energetic electrons being accelerated in a collapsing sheath [18], settling to levels in the range of minus tens of V to 0 V. A typical \( V_f \) obtained in our experiment by flat probes mounted close to the target edge is shown in figure 2(a). As expected, large negative values of \( V_f \) are observed at the beginning of the pulse, followed by an increase in \( V_f \) to values of several volts. Positive values of \( V_f \) could be understood by considering the location of the flat probes measuring \( V_f \). \( V_f \) is expected to be dependent on electron energy [19]: energetic electrons in the magnetic trap lead to larger negative values, while electrons reaching the target edge are less energetic and less abundant, leading to \( V_f \) values close to 0 V [19]. Since the flat probes in our experiment are placed at the target edge, a small positive value could originate from a low energy of electrons at the magnetron edge.

The amplitude of the floating potential oscillations, which are correlated with the spikes, is small compared to the background floating potential. In order to investigate the spikes, the empirical mode decomposition (EMD) method [20] is used to remove the background floating potential, and to emphasise the oscillations. Figure 2 shows an example of the EMD technique. Figure 2(a) shows the measured floating potential, and the straight line represents the sum of the last two EMD modes, representing the background floating potential signal.
These last two EMD modes are subtracted from the measured floating potential, resulting in a signal showing floating potential oscillations correlated with spokes, shown in figure 2(b).

All 2D maps of spokes presented in section 3 are data after EMD subtraction.

The rotation direction of spokes can be deduced from the tilt of the 12 signals plotted on the same graph. Figure 3 shows a simulation where 12 sinusoidal waves are shifted in phase, demonstrating how the tilt of the signals indicates the spoke rotation direction. The signals are plotted in a 2D plot with colour-coded intensity. The x-axis shows the time, and the y-axis shows the probe number, from 1 to 12, and

Figure 2. Example of empirical mode decomposition (EMD). (a) Measured floating potential and discharge current; the straight line overlaying $V_f$ represents the sum of the last two EMD modes. (b) Signal obtained by baseline correction based on EMD.

Figure 3. Simulations showing examples of spoke rotation. The tilt of the floating potential signal reveals the rotation direction of the spokes: (a) retrograde $\mathbf{E} \times \mathbf{B}$ direction, (b) stationary oscillations—pulsing plasma, (c) $\mathbf{E} \times \mathbf{B}$ direction.

These last two EMD modes are subtracted from the measured floating potential, resulting in a signal showing floating potential oscillations correlated with spokes, shown in figure 2(b).

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the corresponding azimuthal angle, 0 to 2π, as shown in figure 1(c). The tilt of the fluctuations to the left, as shown in figure 3(a), indicates rotation in a clockwise direction, which is opposite to the \( \mathbf{E} \times \mathbf{B} \) drift direction, i.e. the retrograde \( \mathbf{E} \times \mathbf{B} \) drift direction. A vertical signal without any tilt, as shown in figure 3(b), indicates a pulsing plasma, as the signal simultaneously oscillates on all probes. The tilt of the fluctuations to the right, as shown in figure 3(c), indicates the spoke rotation direction in the \( \mathbf{E} \times \mathbf{B} \) drift direction.

In a planar circular magnetron the spokes are rotating in a closed loop, which means that, e.g. for spokes propagating in the \( \mathbf{E} \times \mathbf{B} \) direction, the oscillation detected on probe 12 will be next detected on probe 1, 2, etc. Therefore, stacking the same time series, i.e. connecting oscillations on probe 12 to probe 1, enables visualisation of the spoke propagation in the azimuthal direction. Figure 4 shows the same time series stacked for three full cycles. For example, the floating potential maximum detected on probe 1 at 130 \( \mu \)s reaches probe 12 at about 145 \( \mu \)s, and shortly after it reaches probe 1. This representation allows us to determine the spoke rotation direction and the spoke velocity.

The spoke mode number, which is defined as the number of spokes simultaneously observed over the target, is determined from the number of \( V_f \) maxima observed around the racetrack at a single time. Figure 5 shows examples of measurements for which spoke mode 1 (figure 5(a)) and spoke mode 2 (figure 5(b)) are observed. The black vertical lines are examples where a number of spokes can be clearly recognised. The right side of figure 5 shows an ICCD image observed at comparable discharge conditions showing spoke mode 1 and spoke mode 2.

3. Results

The spoke rotation direction, the spoke velocity and the spoke mode number are investigated for a range of discharge currents from 10 mA (current density 0.5 mA cm\(^{-2}\)) to 140 A (7 A cm\(^{-2}\)) for Al, Cr and Ti targets. For this purpose each result is represented by identical time series stacked for seven full cycles, each marked by multiplications of 2π. The presented results show typical observed behaviour of the spokes for three target materials, unless otherwise specified.

The observed trends are grouped in four phases. Phase I corresponds to the low current regime, where the spokes rotate in the direction opposite to the \( \mathbf{E} \times \mathbf{B} \) drift, i.e. with retrograde \( \mathbf{E} \times \mathbf{B} \) motion. Phase II corresponds to a reversal of the spoke rotation from retrograde \( \mathbf{E} \times \mathbf{B} \) motion to \( \mathbf{E} \times \mathbf{B} \) motion. Phase III corresponds to an \( \mathbf{E} \times \mathbf{B} \) motion with a decrease in the spoke velocity at high discharge currents. Phase IV corresponds to the disappearance of any observable self-organisation patterns with plasma becoming homogeneous.
3.1. Phase I—retrograde motion

At the lowest current density, between 10 mA and 20 mA, the spokes exhibit a periodic change of their rotation direction, as shown in figure 6. The change of rotation direction has a period of about 800 μs. During the retrograde $\mathbf{E} \times \mathbf{B}$ rotation, the angular speed is in the range 0.2–0.3 km s$^{-1}$ (angular frequency $\approx$ 3 kHz), and during rotation in the $\mathbf{E} \times \mathbf{B}$ direction the angular speed is in the range of 0.3–0.4 km s$^{-1}$ (angular frequency $\approx$4 kHz). Only spoke mode 1 has been observed.

For a discharge current between 20 mA and 130 mA, as shown in figure 7, the periodic change of direction stops, and the spoke rotates in the retrograde $\mathbf{E} \times \mathbf{B}$ direction with varying velocity, as shown in figure 7. Only spoke mode 1 has been observed with the angular speed in the range of 0.2–1.2 km s$^{-1}$ (angular frequency in the range 2–14 kHz) increasing with the discharge current as $\approx \log(I)$, shown in figure 16. Below 130 mA, no pulsing of the plasma is observed.

For discharge currents above 130 mA, the DC plasma appears to start pulsing as the floating potential minima and maxima appear simultaneously on all flat probes, as shown in figure 8(c). Comparison between a single floating potential signal (figure 8(a) and the plasma emission signal (figure 8(b)) shows simultaneous oscillations of plasma emission and floating potential at a frequency of about 250 Hz. The pulsing period can be divided into plasma ON times, characterised by high plasma emission and high floating potential oscillations, and plasma OFF times, characterised by low plasma emission and low floating potential oscillations, indicating low excitation levels of the plasma and low electron energy.

The spoke rotation during plasma ON times in the pulsed mode for all target materials, and in continuous mode for Cr and Ti targets for discharge currents above 500 mA, is in the retrograde $\mathbf{E} \times \mathbf{B}$ direction with a single spoke observed; see figure 9. The angular spoke speed is in the range of 1.4–2 km s$^{-1}$ (angular frequency $\approx$20 kHz). Between plasma ON times in the pulsed mode, retrograde $\mathbf{E} \times \mathbf{B}$ spoke rotation is observed, with the angular speed in the range of 0.13–0.34 km s$^{-1}$ comparable to the speeds at lower discharge currents.

The spoke mode observed for discharge currents between 10 mA (0.5 mA cm$^{-2}$) and 600 mA (30 mA cm$^{-2}$) is always $m = 1$. The length of the racetrack, which is for $m = 1$ also a spoke wavelength, for a 2″ magnetron is $\sim$85 mm. For larger magnetrons, having a longer racetrack, it is reasonable to assume that the spoke mode number will be larger than one. Martines et al [11] used a 4″ magnetron with a racetrack length of $\sim$250 mm. At a pressure lower than 0.5 Pa and at a comparable discharge current of 600 mA (7.2 mA cm$^{-2}$) they observed dominating spoke mode number $m = 3$, which yields a spoke wavelength of $\sim$83 mm comparable to the spoke wavelength of $\sim$85 mm in our experiment.

3.2. Phase II—spoke rotation reversal

The cooling capacity limits the use of the DC power supply to a maximum current of 600 mA. For that reason a HiPIMS power supply was used, with a minimal achievable discharge current of 1 A. In a HiPIMS discharge, the discharge current is not constant in time, thus a discharge waveform is plotted in each figure. Figure 10 shows a HiPIMS discharge with an Al target, with discharge current increasing from 0 to 5 A. For a discharge current below the threshold the spokes propagate in the retrograde $\mathbf{E} \times \mathbf{B}$ direction.

Increasing the discharge current above a certain threshold the spoke rotation changes direction from a retrograde $\mathbf{E} \times \mathbf{B}$ direction to a $\mathbf{E} \times \mathbf{B}$ direction. Figure 11 shows a HiPIMS discharge with Al target, with peak currents of 10 A, and a transition at a discharge current of about 7 A. The discharge current at which the transition takes place depends on the
target material. For Cr and Ti it is at discharge currents of about 1–2 A.

In figure 12 it can be seen that the spoke angular velocity increases with increasing discharge current, as the tilt becomes steeper, for a HiPIMS discharge with Ti target. This is in good agreement with previous results, showing a linear increase of the spoke angular velocity with discharge current [13].

3.3. Phase III—$E \times B$ motion with spoke velocity decrease

From the discharge current of several amperes (exact value depends on target material), at which the spoke rotation changes direction, up to 140 A the spoke rotation is in $E \times B$ direction, as previously reported [14, 15].

Fast ICCD camera measurements showed that the spoke mode number is high for low discharge currents, but decreases to spoke mode 1 for high discharge currents [15]. The same behaviour was observed with the 12 FPA. Figure 13(a) shows spokes in spoke mode 2 and 3 in a medium-power HiPIMS discharge, Al target.
discharge. Figure 13(b) shows the signals for times between 60 μs and 80 μs ($I_d \sim 20 \text{ A}(1 \text{ A cm}^{-2})$), and between 130 μs and 150 μs ($I_d \sim 40 \text{ A}(2 \text{ A cm}^{-2})$) indicating spoke mode 3 and spoke mode 2, respectively. The spoke mode transition itself has been discussed in detail elsewhere [15].

3.4. Phase IV—homogeneous plasma

Increasing the discharge current further leads eventually to a homogeneous plasma [21]. For a homogeneous plasma, the measured $V_f$ is flat, without any detectable oscillations. Figure 14 shows the transition from a plasma with spoke mode 1 to a homogeneous plasma, for a HiPIMS discharge for a Cr target. At the lower currents, spoke mode 2 is visible, transitioning to spoke mode 1, and at a discharge current of about 110 A, the floating potential oscillations disappear, indicating a homogeneous plasma. ICCD images of the HiPIMS plasma for a Cr target are shown as examples in figure 14(a). For the discharge with Al and Ti targets, no transition has been observed up to discharge currents of 180 A, which is the highest current achievable, before the discharge starts to arc uncontrollably. Recently Poolcharuansin et al [22] showed that a transition to a homogeneous plasma in a HiPIMS discharge for an Al target takes place at pressures above 1.5 Pa.

4. Discussion

4.1. Spoke rotation models

The experiments revealed a very characteristic dependence of spoke rotation on discharge current and target material: for discharge currents in the range between 10 mA and several amperes, the spokes propagate in the retrograde $\mathbf{E} \times \mathbf{B}$ direction and above several amperes ($\sim 7 \text{ A} (0.35 \text{ A cm}^{-2})$ for Al, $\sim 2 \text{ A} (0.1 \text{ A cm}^{-2})$ for Cr, and $\sim 1.5 \text{ A} (0.08 \text{ A cm}^{-2})$ for Ti target) the rotation direction reverses and the spokes rotate in the $\mathbf{E} \times \mathbf{B}$ direction. This behaviour is summarised in figure 15 for the three materials under investigation.
The dynamics of spoke rotation are separated into four phases I–IV: phase I corresponds to the low-current regime, where the retrograde spoke velocity gradually increases before it reaches a steady-state small value. At the beginning of phase II, the direction of rotation reverses and the spoke velocity increases before it saturates at a large value. This large value of about 10 km s\(^{-1}\) may be reached rather quickly as in the case of Al (figures 11 and 15), or only at very high discharge currents as in the case of Ti (figures 12 and 15). The transition at the beginning of phase II coincidences with the onset of gas rarefaction and the transition from a noble gas plasma to a metal plasma. This is corroborated by the fact that the spoke velocity is independent of the target material in phase I where the plasma is argon-dominated, as shown in figure 16. In contrast, the spoke dynamics are dependent on the target material in phases II–IV where metal ions dominate, as shown in figure 15. Phase III corresponds to a \(\mathbf{E} \times \mathbf{B}\) motion where the spoke velocity decreases at very high discharge currents. This phase is only reached with Al and Cr as target materials. Phase IV corresponds to the disappearance of any observable self-organisation patterns with the plasma becoming homogeneous. This phase is only reached with Cr as the target material.

The description of the variation of spoke velocities is a very controversial topic with several different explanations at hand:

- **Critical ionisation velocity (CIV) model** Brenning et al [23] postulated a hypothesis that spoke velocities are correlated with the critical ionisation velocity \(v_{\text{CIV}} = \sqrt{\frac{2eU_i}{m_i}}\), comparing the kinetic energy of the drifting spoke with the ionisation energy \((U_i)\) of the background gas. Poolcharuansin et al [22] tried to explain the variation of spoke velocities with the target material by arguing that at low discharge current the spoke velocity should be close to the value of CIV of Ar, whereas at high discharge currents, the Ar rarefaction results in a sputtered metal-dominated plasma, resulting in a spoke velocity close to the value of CIV for a given metal species. The CIV of Ar \(v_{\text{CIV,Ar}}\) and CIV of metal ions \(v_{\text{CIV,M}}\) are plotted in figure 15. Such a model may only account for spoke velocities in phase II and III where rarefaction plays a role.

For the HiPIMS discharge with a Cr target, the results quite nicely fit to the CIV hypothesis, with the high spoke velocity at the beginning of phase III of \(\sim 10\) km s\(^{-1}\) being close to \(v_{\text{CIV,Ar}} = 8.71\) km s\(^{-1}\). At higher currents in phase III, the spoke velocity of \(\sim 5\) km s\(^{-1}\) is close to \(v_{\text{CIV,Cr}} = 5.01\) km s\(^{-1}\). For HiPIMS discharge with an Al target, the spoke velocity exhibits trends compatible with the CIV hypothesis, but the velocity ranges from \(\sim 9\)–\(14\) km s\(^{-1}\), which is higher than the expected CIV. For a HiPIMS discharge with a Ti target, the spoke velocity exhibits the opposite trend and it reaches higher velocities than predicted by the CIV hypothesis.

There are two issues that lead us to the conclusion that the CIV model cannot explain our data. First, the observed velocities and trends of the spoke velocity do not fit to the CIV model, except for the Cr target at high power. Second, a reversal in spoke rotation for low discharge currents is not predicted by the CIV model.

- **Ionisation rate model** Yang et al observed a propagation direction reversal similar to the trends in our experiments [9]. They explained this reversal with the difference in the rate of ionisation at the leading and the trailing edge of a
rotating spoke. Since the spoke rotates slower than the $E \times B$ velocity of the electrons, the electrons usually enter the spoke from the trailing edge of a previous rotating spoke. It is postulated that at low discharge currents, the noble gas depletion is small so that the entering electrons encounter a sufficient high neutral gas density. As a consequence, the ionisation front of the spoke moves in the opposite direction to the incident electrons (retrograde $E \times B$). At high discharge currents and high depletion, the incident electrons’ impact on a low noble gas density plasma may traverse the spoke and ionise at the leading edge of the spoke so that the ionisation front moves in the direction of the incident electrons ($E \times B$).

The shortcoming of this model is that it compares ionisation rates at the leading and trailing edge, which depend on the actual electron density, the local neutral density and the fraction of the electron energy distribution function of the streaming electrons responsible for ionisation. Despite strong noble gas rarefaction, an intense metal sputtering occurs, which again modifies the ionisation rates at the leading and trailing edge of the spoke. In some cases the local density of neutrals can exceed the density of noble gas in the target vicinity [24]. During a HiPIMS pulse the neutral gas depletion is replaced by the high metal density and based on this model one would expect a further spoke reversal at very high metal atom densities, which is not observed.

Drift wave models Drift wave models have been postulated for various $E \times B$ discharges to explain low-frequency oscillations such as the spoke rotation. Drift waves are driven by density gradients perpendicular to the magnetic fields, which may become unstable for certain critical gradients. Such density gradients connect the high electron density in the spoke region with the cathode and the anode. Good agreement between spoke velocities and drift wave velocities can be found by assuming reasonable electric fields and density gradients surrounding the spokes [25]. The influence of gas rarefaction and depletion is usually not regarded. Ito et al. recently observed a retrograde spoke rotation for small discharge currents in a recent micro-sized $E \times B$ configuration and connected it to a drift wave model by including also the ambipolar diffusion along the $B$-field lines parallel to the cathode. Their main postulate was that the retrograde motion with respect to the electric field at the cathode can be explained by invoking an electric field towards the anode which connects a potential hump in the spoke region to the anode.

### 4.2. Potential hump model for spoke rotation

Based on the hypothesis of a potential hump in the spoke region [26, 27], we develop in the following a consistent model for the observed variation of the spoke velocities with discharge currents. Figure 17 is an illustration of the variation of the axial magnetic field $B_z$, the electrical potential between cathode and anode $\Phi_{pot}$, the sheath thickness and the region of plasma emission between cathode and anode. Figure 17(a) depicts the situation for low discharge currents corresponding to phase I and figure 17(b) for high discharge currents corresponding to phases II and III.

The spoke itself corresponds to a region of high electron density and therefore to a hump in the electrical potential. This hump is connected by a gradient in electron density and an ambipolar electric field parallel to the $B$-field line to the anode. This hump is also connected with a gradient in electron density to the cathode. In addition, an electric field towards the cathode in the magnetic pre-sheath has to exist to confine the unmagnetised heavy ions to assure quasi-neutrality with respect to the magnetised electrons. The height of the potential hump and thus the density gradients depend on the discharge currents. The strengths of the electric fields depend on the dynamic of the dominant ions either streaming to the anode or being confined in the magnetic trap above the cathode. ICCD camera measurements from the side (not shown here) reveal that the plasma emission zone at low power is spread away from the target, while at high power it is confined near the target, as sketched in figure 17.

- **Phase I**: retrograde spoke motion: spokes are regarded as drift waves driven by the density gradients and $E$-fields connecting the potential hump to the anode. At low discharge currents, the target sheath is of the order of a few mm, shifting the plasma further away from the cathode. One may postulate that the region of plasma emission is apparently dominated by the density gradient and $E$-field to the anode. These density gradients are rather small and thus also the ambipolar electric field gives rise to rather small spoke velocities. This explanation is identical to the hypothesis of Ito et al [25]. The spoke velocity saturates to a certain value, when the density gradient and thus the ambipolar $E$-field as the driver for the drift waves reach a critical value. Any steeper density gradient may give rise to turbulence or to flares. In this phase the dominant ions are argon ions, so the velocities are independent of the target material, as shown in figure 16.

- **Phase II**: spoke rotation reversal: above a certain discharge current the transition to the metallic mode occurs. As a consequence, the ions are created from sputtered species at the target. With metal atom ionisation energy in range of 6–6.8 eV, compared to 15.7 eV for the Ar atom, ionisation is enhanced. These sputtered atoms exhibit kinetic energies corresponding to the Thompson energy distribution with most neutrals having energies in the range of a few eV depending on the surface binding energy of the material. The electric field in the magnetic pre-sheath (see figure 17) has to be stronger than in the case of the argon-dominated discharge in phase I, because the ions are generated by ionisation of fast neutrals travelling away from the cathode. The potential distribution changes to a configuration with a strong $E$-field in the magnetic pre-sheath, which then dominates the direction of spoke rotation. With a further increase in discharge currents, the density gradient might further change, which leads to a further increase in spoke velocity. This behaviour...
is apparently dominant for Ti, where the spoke velocity continuously increases in phase II before it saturates at very high discharge currents. This saturation at high velocities is reached at much lower discharge currents in the case of Al and Cr.

ICCD camera measurement [15, 16] revealed that the spokes in the case of Ti are rather diffuse, indicating small density gradients surrounding the spoke. An increase in discharge current is apparently converted only into a gradual change of spoke size. Also here, a finite gradient is apparently reached for high discharge currents. This saturation is identical to phase I, because any steeper gradient would induce turbulence and flares. The absolute value is much higher for phase II, because here, the plasma is in its metallic mode with a very different ion dynamic to the argon plasma in phase I.

**Phase III: \( \mathbf{E} \times \mathbf{B} \) motion with spoke velocity decrease:** beyond phase II, a decrease in spoke rotation is observed. This is only visible for Al and Cr. In contrast to Ti, Al and Cr exhibit rather triangular spokes with sharp density gradients. Based on this observation, one may speculate that the transition from phase II and III for Al and Cr is reached at rather small discharge currents, because the critical gradient in the plasma density and thus maximum spoke velocity is established already at small discharge currents at the end of phase II.

If the discharge current is increased in phase III, the spoke velocity decreases and saturates at a lower velocity. This behaviour may be explained by the general dependence of spoke velocity on background pressure. Since the absolute spoke velocity at high discharge currents is dominated by the dynamic of local gas depletion in front of the leading edge of the spoke, a general decrease in spoke velocity with increasing background pressure is observed [13]. Such an interplay may also dominate phase III where the intense metal sputtering causes a very high local neutral density which slows down spoke movement. The self-sputtering yield of Al at a typical ion energy of 500 eV is 1.07, of Cr 1.09 and of Ti only 0.51. In particular, the high self-sputtering yields for Al and Cr above unity support the runaway regime and a very high metal atom density in front of the target.

### 4.3. Plasma oscillations

Besides the general behaviour of spoke rotation in phase I, II, III and IV, one also observes peculiar plasma oscillations such as a periodic spoke rotation reversal at the beginning of phase I and a plasma pulsing at the end of phase II at the onset of the metallic mode.

#### 4.3.1. Periodic spoke rotation reversal at very low currents

As shown in figure 6, the spoke rotation is changing periodically at very low discharge currents with a period of the order of 1 ms, and the measured floating potential is higher for the retrograde period compared to the normal period of spoke rotation. If one assumes that the floating potential is a measure for the proximity of the plasma region to the probe location, one may conclude that the plasma is in its retrograde mode further away from the target and thereby close to the flat probes compared to the normal motion. Based on the potential hump hypothesis, the direction of spoke rotation depends on the direction of the density gradient and the electric field in the region of plasma emission. An oscillation period of 1 ms may indicate an influence of a periodic movement of the local neutral gas density leading to a plasma emission zone either in the density gradient close to the target and the normal \( \mathbf{E} \times \mathbf{B} \) motion or away from the target leading to the retrograde motion. This description remains rather speculative and requires more investigation in the future.

#### 4.3.2. Plasma pulsing at the end of phase I

Change in the plasma dynamics due to gas rarefaction could lead to a pulsing plasma. Gas rarefaction [28–32] and sputtering wind [33] are
well-known and well-investigated phenomena. In principle, gas rarefaction occurs as a gas displacement process due to momentum transfer by sputtered atoms. The gas rarefaction is enhanced for target materials with high sputter yield [29, 34], and for target materials with atomic mass comparable to the working gas, yielding larger efficiency in energy and momentum transfer [35].

In a simplified picture, the gas rarefaction becomes substantial when the sputtered metal particle pressure $p_m$ exceeds the Ar pressure $p_{Ar}$. The calculated value (complete calculations are presented in the appendix) stems from the most probable values of sputter yield, electron temperature and atom binding energy, and gives an approximate current at which the sputtered particle pressure is comparable to the background Ar pressure. The calculated 120 mA fits well with the observed threshold current of 130 mA, above which the DC plasma exhibits a pulsing behaviour.

The presented simple calculation shows that discharge current above 120 mA (average current density of 6 mA cm$^{-2}$) could result in sputter wind displacing Ar and causing disruptions in continuous DC discharge. A similar result has been presented by Rossnagel [32] and Kobayashi [30]. Rossnagel observed a trend in which an Ar gas density reduces when increasing the magnetron discharge current, exhibiting a considerably reduced gas density starting at a discharge current of 700 mA (on 6th target, average current density of 4 mA cm$^{-2}$).

5. Conclusions

The plasma oscillations in a round planar magnetron have been investigated by a 12 FPA mounted azimuthally around the magnetron, measuring floating potential oscillations with a high temporal resolution. The 2D spatio-temporal maps of the floating potential oscillations allowed us to investigate the rotation direction, mode number and angular velocity of the oscillations, i.e. spokes. The presence of spokes throughout the complete investigated discharge current range indicates that the spokes are an intrinsic property of a magnetron sputtering plasma discharge. The observed properties of the spokes exhibit a strong dependence on the discharge power level and on the target material.

Regarding the discharge power level dependence, the observed trends are grouped in four phases. Phase I corresponds to the low current regime, where the spokes rotate in the opposite direction to the $\mathbf{E} \times \mathbf{B}$ drift, i.e. retrograde $\mathbf{E} \times \mathbf{B}$ motion. Phase II corresponds to a reversal of the spoke rotation from retrograde $\mathbf{E} \times \mathbf{B}$ motion to $\mathbf{E} \times \mathbf{B}$ motion. Phase III corresponds to an $\mathbf{E} \times \mathbf{B}$ motion with a decrease in the spoke velocity at high discharge currents. Phase IV corresponds to a disappearance of any observable self-organisation patterns with the plasma becoming homogeneous.

Regarding the target material dependence, in phase I there is no dependence on the target material, indicating that the spoke dynamics are driven by Ar gas. In phase II the transition takes place due to a transition from Ar-dominated to metal-dominated sputtering. The transition takes place within several amperes for the Al target, within 15 amperes for the Cr target, and more than 140 A for the Ti target. In phase III the spoke angular velocity reduces for the Al and Cr target. Phase IV, homogeneous plasma, is observed only during HiPIMS discharge with a Cr target.

Based on the hypothesis of a potential hump in the spoke region, we present a consistent model for the observed variation of the spoke velocities with discharge currents. The spoke rotation reversal is explained by the transition from Ar-dominated to metal-dominated sputtering that shifts the plasma emission zone closer to the target. The spoke itself corresponds to a region of high electron density and therefore to a hump in the electrical potential. At low power, the plasma is further away from the target and it is dominated by the electric field to the anode, thus retrograde $\mathbf{E} \times \mathbf{B}$ rotation. At high power, the plasma is closer to the target and it is dominated by the electric field to the target, thus $\mathbf{E} \times \mathbf{B}$ rotation.

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Appendix

In a simplified picture, the gas rarefaction becomes substantial when the sputtered metal particle pressure $p_m$ exceeds the Ar pressure $p_{Ar}$. A similar approach was suggested by Rossnagel [32], considering the gas rarefaction. The discharge current at which the transition takes place can be calculated from $p_m/p_{Ar} = 1$. The sputtered metal particle pressure $p_m$ can be calculated from a simple gas equation, $p_m = n_m \times T_{m,eff} (eV) \times e$. Similar to the method used by Lieberman [36], the sputtered metal density $n_m$ can be calculated from the density of Ar ions impinging on the target, entering the sheath with Bohm velocity $v_B$, and by assuming sputter yield being equal to 1:

$$n_m = n_{Ar,i} = \frac{I_d}{v_B \times e \times A} \quad (A.1)$$

$I_d$ is a discharge current, and $A$ is an effective target area conducting discharge current. Assuming that the discharge is driven through a single spoke, covering half of the racetrack length of $\sim 4$ cm, with a racetrack width of $\sim 0.7$ cm, we get $A \sim 3$ cm$^2$. Bohm velocity is calculated for electron temperature $T_e = 3$ eV:

$$v_B = \sqrt{T_e (eV) \times e/m_{Ar}} \approx 2.7 \text{ km s}^{-1} \quad (A.2)$$

Combining equations (A.1) and (A.2), and taking the mean energy of Thompson distribution (2/3 of binding energy) as an effective temperature of sputtered species $T_{m,eff} = 2/3E_B \sim 3.33$ eV, we get:

$$p_m = n_m \times T_{m,eff} (eV) \times e = \frac{I_d}{v_B \times e \times A} \times \frac{2}{3} E_B \times e = p_{Ar} \quad (A.3)$$
From equation (A.3) a threshold discharge current can be calculated:

$$I_d = \frac{p_{\text{th}} \cdot v_B \cdot e \cdot A}{2/3 E_0 \cdot e} \approx 120 \text{ mA} \quad (A.4)$$

References