Spoke transitions in HiPIMS discharges

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1. Introduction

High power impulse magnetron sputtering (HiPIMS) is a well-established deposition technique, where power is delivered in short pulses at low duty cycle. HiPIMS plasmas contain highly ionised sputtered material [1–4], resulting in dense functional films with superior properties [5]. Apart from a high ionisation degree of the sputtered metal, the high powers delivered in relatively short pulses result in highly dynamic non-stationary plasmas that have been extensively studied by various plasma diagnostics [6]. The highly ionised plasmas are commonly susceptible to oscillations and instabilities [7]. Periodic fluctuation of the electric field in the MHz range have been observed during the ‘on’-phase of the HiPIMS discharge, originating from a modified two stream instability [8]. Recently, we have reported fluctuations in the 100kHz range, represented by localised light emission rotating in the \( \mathbf{E} \times \mathbf{B} \) direction [9] [10]. Independently, Kozyrev et al [11] and Anders et al [12] have observed the same phenomena. Several decades earlier, similar rotating structures were discovered and have been well-documented in the literature on Hall thrusters, where they are commonly referred to as spokes [13]. Due to the similarity, we will adopt that name. The spokes exhibit a transition from stochastic to periodic behaviour depending on discharge conditions [10]. At high discharge powers, the spokes disappear and the plasma emission becomes homogeneous [14].

Spokes observed in cylindrical Hall thrusters [15] exhibit a wide range of fluctuations and instabilities [16]. Several publications pointed out that axial gradients of magnetic field and plasma density are the source of instabilities [17–19], including Rayleigh–Taylor instabilities [20]. Further publications indicate that the instabilities can explain an anomalous transport mechanism [13, 21–23]. An overview of high frequency instabilities and their correlation to the anomalous electron transport in Hall thrusters has been presented by Lazurenko et al [22]. Apart from Hall thrusters, fluctuations in the 100kHz range and anomalous transport have been found in several other systems, for example in closed drift accelerators [24] and in plasma gun experiments [25, 26]. The general understanding is that spokes originate from a charge separation between electrons, being magnetised and experiencing an \( \mathbf{E} \times \mathbf{B} \) force, and unmagnetised ions. The charge separation generates an azimuthal electric field \( \mathbf{E}_\phi \), resulting in an \( \mathbf{E}_\phi \times \mathbf{B} \)
drift towards the substrate. In DC magnetron sputtering discharges, fluctuations and localised plasma emission have been observed as well [27–30]. Waves propagating in the azimuthal direction behaving in a disorderly fashion have been reported [28], however, no indication of an enhanced cross field transport has been found [27, 29].

For a HiPIMS discharge, Kozyrev et al explained the spokes as an effect of a high density ion current driven along the \( \mathbf{E} \times \mathbf{B} \) drift, generated by the azimuthal electric field \( \mathbf{E}_\phi \) [11], attributed to azimuthal modulations of the plasma density. Anders et al presented a model of the spoke, where the spoke is an ionisation zone in which electrons are decelerated due to interaction with charged particles [12, 31] resulting in a region of locally enhanced potential [32]. Brenning et al [33, 34] presented a physical model to describe the spoke based on the Alfvén critical ionisation velocity (CIV). The ionisation within the structure creates an electric field as a result of a charge separation based on the CIV theory. This model postulates a modified two stream instability accelerating the electrons towards the target and generating a drift opposite to the \( \mathbf{E} \times \mathbf{B} \) direction explaining the rotation speed being one order of magnitude lower than the \( \mathbf{E} \times \mathbf{B} \) drift speed of electrons. Gallian et al [35] presented a phenomenological model where a steady-state configuration in the rotating frame is achieved for electron and neutral densities, assuming a balance of ionisation, electron loss, and constant neutral refilling. Based on the influence of the target material on the shape of the spoke Hecimovic et al have recently proposed a model based on a change of the secondary electron production due to localised transition from the Ar dominated to the metal dominated sputtering [36]. Our motivation for understanding the origin of spokes is not only of theoretical interest, but rather practical. Both experimental [37] and modelling [38, 39] results suggest a strong correlation between the spokes and the ion energy distribution arriving at the substrate, the latter being important for thin film deposition.

In most plasmas, exhibiting self-organisation patterns, such as dielectric barrier discharge (DBD), gas-discharge semiconductor system (GDSS) [40], cathode boundary layer discharge (CBLD) [41], Hall thrusters [42], planar gas discharges [43], the mode number, i.e. the number of spokes or filaments, increases with increasing the current. The source of filamentation in a glow-like discharge is a space charge induced field redistribution [40]. Benilov [44] argued that two mechanisms are competing: electrostatic forces, favouring the appearance of modes with multiple spots, and charged particle diffusion favouring the appearance of modes with one spot. The competition of these two mechanisms leads to the appearance of multiple spots in an abnormal mode at low currents, and to the existence of a single spot in the normal mode. Due to their electrostatic nature, the multiple spots exhibit a repulsive interaction [43] and the number of spots scales proportional to the voltage supplied.

In HiPIMS, the number of spokes decreases by increasing the discharge current, which seems to be a unique behaviour of plasma self-organisation. In comparison to the other glow-like plasma discharges, in magnetron plasmas the particle density and the temperature are greatly influenced by plasma-target interaction. The influence of plasma-target interaction on a balance between repulsion and diffusion, and on the spoke mode transition is of central interests in this paper. Four fast ICCD cameras are used to present the evolution from stochastic to self-organised spoke patterns. However, understanding the spoke mode transition requires a better temporal resolution than four sequential images from ICCD cameras. Therefore, 12 flat probes placed azimuthally around the race-track are used, providing a sufficient spatial resolution with a temporal resolution of 10 ns. The flat probes measure floating potential oscillations corresponding to the spoke rotation [36].

2. Experimental setup

The experiments have been performed in a cylindrical chamber of 35 cm in diameter and 45 cm 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.

Figure 1 shows the applied setups of optical and electrical plasma diagnostics. In figure 1(a), a four ICCD cameras mount facing the target surface is shown. The setup is adjusted so that the light passing through a set of beam splitters arrives simultaneously at the photo-cathode of each camera. The four cameras were used in a sequential mode, where the cameras were triggered in sequence with a 1 µs delay. For all measurements the acquisition time was set to 100 ns. For a typical spoke angular velocity of 10 km s\(^{-1}\) [9, 12], in 100 ns the spoke moves 1 mm, which is 1.2% of the racetrack length.

Figure 1(b) shows the setup with a photomultiplier tube (PMT). The plasma light emission was measured by PMT. 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.

Figures 1(c) and (d) show a detailed scheme of the setup with 12 flat probes placed azimuthally around the magnetron, separated by 30°, covering the complete circle around the target. The probes are placed at a distance of 17 mm from the racetrack and 5 mm above the target surface. Each probe measures the floating potential \( V_f \) oscillations during the pulse independently. The correlation between the periodicity of floating potential oscillations and the spoke rotation has been established previously [36]. The 12 probes measure 12 \( V_f \) simultaneously within a single HiPIMS pulse. 12 \( V_f \) are measured using twelve 14-bit AD converters, with a rate of 100 MS s\(^{-1}\). Figure 2 shows an example of the signal plotted in a 2D plot with colour coded intensity, from 50 µs to 210 µs. The \( x \)-axis shows the time, and the \( y \)-axis shows the probe number and the corresponding angle. The floating potential oscillations are in range from 2 to 4 V, with green colour representing higher values and blue colour lower values. The tilt of the fluctuations corresponds to the spoke rotation.

The repetition frequency of the discharge was set to 10 Hz, the working gas was Ar and the pressure was 0.17 Pa in all experiments presented here unless otherwise specified. The peak discharge current was varied between 7 A (current density 0.35 A cm\(^{-2}\)) and 150 A (7.5 A cm\(^{-2}\)). In a previous publication, we demonstrated that the shape of the spoke depends on target material, exhibiting a ‘diffuse’ or a ‘triangular’ shape.
Here, we present results of the discharges with a Ti and an Al target, as representative examples of a 'diffuse' and a triangular spoke. Typical current and voltage waveforms of a HiPIMS discharge with a Ti target are shown in figure 3(a) and with an Al target in figure 3(b). After some delay, the current is rising until it reaches a relatively constant current plateau. The current plateau is of great significance, since it ensures a steady discharge during which the spokes are observed.

3. Results

In analogy to an early work in Hall thruster [13], the spoke is identified as a region of increased light emission propagating in the azimuthal direction, exhibiting a self-organisation pattern. On a circular magnetron, the spoke self-organisation is perceived as a specific number of spokes over the race-track. The number of spokes on the target, or the spoke mode number, depends on several discharge parameters. Increasing the pressure will result in an increased number of spokes [9]. Increasing the discharge current, the spoke appearance evolves from a predominantly stochastic to a periodic behaviour [9, 10, 31]. Further increase in the power results in a transition to a homogeneous plasma torus [14]. For a given pressure and discharge current, the number of spokes depends also on the target material, which will be discussed in the following.

3.1. Evolution of spoke during a HiPIMS pulse

Figure 4 shows the progression of spokes, for a HiPIMS discharge with Ti target, as the discharge current is increased. The HiPIMS discharge is very dynamic and the images on the right side show the most probable spoke appearance for an associated discharge current marked with an index. The plasma emission evolves from an almost homogeneous plasma (H), observed at low discharge currents, over a predominantly stochastic plasma (S) to a periodic spoke patterns exhibiting a self-organisation pattern (I–IV). By increasing the discharge current the spoke mode number decreases from spoke mode 4 to spoke mode 1. For each range of discharge current, several images from different discharges are shown. Entirely self-organised patterns, such as one shown in the first column, are observed in 50% of total recorded images. Fluctuations of the mode number by ±1 within one current region were observed, as shown in other columns in figure 4.

Titanium target—For a HiPIMS discharge with Ti target, the current range for spoke mode 4 (IV) is 40–60 A (2–3 A cm⁻²), for spoke mode 3 (III) 65–80 A (3.2–4 A cm⁻²), for spoke mode 2 (II) 85–110 A (4.2–5.5 A cm⁻²), and above 120 A (6 A cm⁻²) only a single spoke (I) is observed. Up to the maximum discharge current allowed by our power supply of 180 A (9 A cm⁻²), the plasma emission exhibits a single spoke and no transition to homogeneous plasma can be resolved. It is worth noting that the radial width of the plasma emission broadens as the discharge current increases. This is consistent with optical [45] and electrical measurements [46], reporting a broader target erosion profile during a HiPIMS discharge compared to a rather narrow target erosion profile during DC discharge.

Aluminium target—For a HiPIMS discharge with an Al target, the spoke modes appear at lower current levels compared to Ti. Unlike the diffuse spoke in the Ti case, the spoke shape for Al exhibits a triangular shape [36]. Figure 5 shows images of the spoke with mode number 3, 2, and 1, at discharge...
currents of 30 A (1.5 A cm⁻²), 50 A (2.5 A cm⁻²), and 100 A (5 A cm⁻²). Above 100 A, the discharge becomes very unstable and the pulse usually ends with an arc. By increasing the discharge current, the spoke mode number decreases from spoke mode 3 to spoke mode 1. The examples of spoke images for the Ti and the Al target show that for a given pressure and discharge current, the spoke mode will be different for different target materials. For example, at a pressure of 0.17 Pa and a peak discharge current of 50 A, a discharge with a Ti target exhibits spoke mode 4, while a discharge with an Al target exhibits spoke mode 2. The dynamics and the shape of the spoke is influenced by both the properties of the target material, e.g. sputter yield, secondary electron yield, and the properties of the sputtered vapour, e.g. ionisation potentials, mass, atom, and ion radius [36].

3.2. Stability of the spoke mode

In order to understand the stability of a spoke pattern in one mode, measurements with the PMT were performed. The correlation between the PMT signal and the spoke image taken by the ICCD camera has been established previously [36]. Figure 6 shows oscillations in light intensity for a time period of 80 μs, from 130 μs to 210 μs, with the pulse ending at 200 μs. The discharge currents in figure 6 demonstrate that during this time the change in current is relatively small, and within the current region described in section 3.1. It is between 130 A and 144 A for Ti (figure 4) and between 90 A and 100 A for Al (figure 5). During this period the single spoke appears to be steadily rotating without changing the mode number. This implies that the shape of the spoke is invariant with respect to time and space. We can speculate that keeping the current constant within one region, the spoke pattern would continue to rotate steadily around the racetrack without changing its mode number.

3.3. Spoke mode transitions

Transitions between spoke modes were observed using spatially and temporally resolved measurements. The 12 flat probes experiment was designed to allow a simultaneous measurement of the floating potential oscillations at 12 positions along the magnetron circumference. Figure 7(a) shows the floating potential oscillations during a single discharge. Figure 7(b) shows the current discharge waveform. The y axis corresponds to the azimuthal angle around the magnetron. One set of signals from 12 probes is plotted from 0 to 2π as shown in figure 2. The oscillations propagating in the E x B direction detected on probe 12, will next be detected on probe 1, 2 etc. Therefore, adding the time series on top allows us to observe propagation of the spoke in azimuthal direction. In figure 7(a) a time series over six periods is added, marked by multiplications of 2π. A transition between the spoke mode 2 and spoke mode 1 has been emphasized in figure 7(a).
It shows two spokes merging into a single spoke as the discharge current increases. Before 105 $\mu$s two spokes rotate with constant velocity. At approximately 105 $\mu$s the spoke merger starts (first vertical line in figure 7). The upper spoke appears to grow in size, and the bottom spokes appears to reduce in size and to speed up. At about 120 $\mu$s (second vertical line in

Figure 4. Images of the spoke showing evolution of the spoke during a HiPIMS pulse with Ti target. The grey rectangular shows the most probable shape of the spoke for a given range of discharge currents. Several images from different discharges are shown. The Roman capitals indicated the spoke mode number, $S$ denotes the stochastic regime, $H$ denotes homogeneous regime.

Figure 5. (a) The discharge current waveforms correspond to the discharge conditions at which the images were recorded, for Al target at 0.17 Pa. (b) Rotation of the spoke recorded by four cameras triggered consequently with $1 \mu$s delay. Three discharge conditions are shown, with the peak currents of 30 A, 50 A, and 100 A, showing spoke mode 3, 2, and 1, respectively. The speed of spokes is about 7 km s$^{-1}$. The ICCD camera acquisition time is 100 ns.
6. From 120 s until the end of the pulse a single spoke rotates steadily around the racetrack, as discussed in previous section.

3.4. Current density in the spoke

Recently we showed that the discharge current is driven through the spoke [36]. The current density through the sheath is determined by the Child–Langmuir law:

$$ j = \frac{4\varepsilon_0}{9} \sqrt{2e/m_i} \frac{V_d^{3/2}}{s^2}. $$

(1)

Figure 7) the bottom spoke catches up with the upper spoke, merging into a single spoke. From 120 s until the end of the pulse a single spoke rotates steadily around the racetrack, as discussed in previous section.

Table 1. Calculated current density in the spoke for Ti and Al, and for different spoke mode numbers.

<table>
<thead>
<tr>
<th>Spoke mode</th>
<th>Ti</th>
<th>V_d (V)</th>
<th>Al</th>
<th>V_d (V)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>43</td>
<td>600</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>3</td>
<td>32</td>
<td>650</td>
<td>42</td>
<td>690</td>
</tr>
<tr>
<td>2</td>
<td>29</td>
<td>730</td>
<td>48</td>
<td>750</td>
</tr>
<tr>
<td>1</td>
<td>49</td>
<td>870</td>
<td>60</td>
<td>870</td>
</tr>
</tbody>
</table>

Figure 8. Images of different spoke modes for with lines denoting the assumed effective spoke surface. (a) Spoke modes IV, III, II and I for the Ti target (from figure 4). (b) Spoke modes III, II and I for the Al target (from figure 5).
A linear fit of the calculated current density as a function of discharge voltage \( V_d \) for Al target yields \( j \approx V_d^{1.6} \), which is very close to \( j \approx V_d^{1.5} \) dependence predicted by equation (1). In case of Ti there is no clear trend, probably due to uncertainties in determining the spoke size. Based on equation (1) the sheath thickness can be estimated to 20 \( \mu \)m for all spoke modes.

4. Discussion

Figures 4 and 5 show that in HiPIMS increase in discharge current leads to decrease in the spoke mode number. This seems to be a unique behaviour of plasma self-organisation. In most plasmas exhibiting self-organisation patterns, (DBD), (GDSS), (CBLD), the mode number, i.e. the number of the spokes or filaments increase by increasing the current.

However, the key difference between the DBD, GDSS, CBLD and the magnetron sputtering is the plasma-surface interaction and resulting continuous influx of sputtered material to the plasma. This effectively changes the plasma chemistry, introducing particles of different mass, ionisation energy, and initial kinetic energy, simultaneously displacing the Ar gas. The importance of the plasma chemistry lies in the fact that it determines the generation of secondary electrons, and the secondary electrons are considered to be crucial for ionisation in the plasma.

There are three possible scenarios of spoke mode transition, shown in figure 9. (I) one spoke speeds up and merges with the other spoke, (II) one spoke fades away, and only one spoke remains, (III) one spoke slows down and merges with the other spoke. Experimentally, only examples of scenario I have been observed, figure 7 shows an example, with one spoke reducing in size, and accelerating until it merges with the other spoke.

We postulate a spoke transition model based on coupling between the current densities in the spoke, determined by the Child–Langmuir law, and Ar replenishment rates, which gives an explanation for the spoke mode transition scenario I.

Current density in the spoke—The current through the spoke increases as the discharge current increases. Higher current through the spoke results is higher plasma density and enhanced sputtering. Since the current density in the spoke is limited by the Child–Langmuir law, higher currents result in a larger spoke (figures 4 and 5), displacing Ar further away from the racetrack due to momentum transfer from the sputtered particles. Stronger displacement of Ar, i.e. stronger Ar rarefaction, results in reduced Ar replenishment rates at the race-track position.

**Ar replenishment rate**—The diminishing trailing edge (diffuse spoke) or the sharp trailing edge (triangular spoke) of the spoke was associated with a depletion of energetic electrons [36], due to a transition from Ar sputtering to self-sputtering in the spoke, and the inability of singly charged metal ions to generate secondary electrons. Displacement of Ar will be less pronounced for small spokes, and more pronounced for larger spokes. For a large spoke, characteristic displacement length in radial direction is about 1 cm, and the time between two spokes is about 10 \( \mu \)s. Thus, for a replenishment velocity of about \( 10^3 \) m s\(^{-1}\) Ar gas is replenished. The thermal velocity of Ar at temperature of 1200 K [47] can be estimated to 7.1 \( \times \) 10\(^2\) m s\(^{-1}\). For smaller spokes, the radial displacement length is shorter, and the Ar replenishment rate is higher.

This model is consistent with a phenomenological model of the spoke by Gallian et al [35]. The phenomenological model solves analytically a system of nonlinear coupled partial differential equations. Its solution gives the neutral density in the frame co-moving with the spoke. A steady-state solution for electron and neutral densities is achieved from the balance of ionisation, electron loss and constant neutral refilling. Figure 10(a) shows result of the model, where the Gaussian shaped electron distributions represents plasma density in the spokes. Steady state solution of the model shows electron density rotating with constant angular velocity, with the neutral density outside the spoke increased due to diffusion of Ar gas and Al sputtering at constant rate, dropping sharply in the spine [35].

Figure 10(b) shows a drawing representing a steady state with two spokes based on the phenomenological model of the spoke by Gallian et al [35]. Position of the spokes is maintained by electrostatic repulsion that dominates over charge diffusion, keeping the two spokes at symmetric positions. The symmetry is violated when discharge current increases beyond a certain threshold leading to a merger of two spokes, as shown in figure 7. In the process of spoke mode transition, one spoke increases in size and the other one reduces in size and accelerates (spoke mode transition scenario I).

According to the spoke transition model, the discharge current increase results in a larger spokes. Figure 10(c) shows two spokes, one with increased electron density, representing higher current through that spoke. Omnipresent
density fluctuations result in higher current through one of the spokes. In wake of the larger spoke the Ar replenishment rate is reduced. The following spoke encounters reduced neutral density which leads to a smaller spoke propagating with higher velocity. The spoke gets smaller since fewer Ar neutrals (ionised in the spoke) are available for generation of secondary electrons, responsible for excitation and ionisation in the plasma. Higher spoke velocity in the $\mathbf{E} \times \mathbf{B}$ drift direction can be explained by reduced friction due to reduced neutral density, in analogy to the spokes rotating faster at low pressures, compared to high pressures [9]. Eventually two spokes merge into one spoke.

Asymmetry induced by increase of the discharge current results in enhanced charge diffusion that prevails over the electrostatic repulsion, and leads to a spoke mode transition. The plasma dynamic described in the spoke transition model explains the unique behaviour of spoke mode reduction for higher discharge currents.

An example of the scenario when the charge diffusion is not sufficient to prevail over the repulsive force between the spokes is shown in figure 11. Figure 11 shows the transition from asymmetric to symmetric spoke during a period of 3 $\mu$s. One of the spokes breaks into two parts with different velocities, the faster and the slower part. The intensity of the slower part increases, particularly when it reaches the symmetric position with respect to the undivided spoke. In the meantime the faster part disappears. At the end of transition two symmetrically distributed spokes remain stabilised via electrostatic repulsion.

5. Conclusions

In this paper the temporal evolution of the spokes during a HiPIMS discharge is presented. The results show that the number of spokes, i.e. the spoke mode number, depends on the discharge current, and target material. By increasing the discharge current the spoke mode number decreases. This seems to be unique behaviour, since in a similar glow-like discharges the number of spokes increase with increasing current. Keeping discharge constant, the spoke mode rotates steadily around the racetrack until the end of the pulse. This implies that the spoke is invariant with respect to time and space. The current density through the spoke is determined by the Child–Langmuir law. For a discharge with Al target the scaling $j \approx V_d^{1.5}$ was found, which is comparable to $j \approx V_d^{1.5}$ dependence predicted by the Child–Langmuir law.

The transition between the spoke modes was observed using 12 flat probes placed azimuthally around the magnetron, measuring floating potential oscillations. The observed transition between spoke mode 2 and spoke mode 1 shows that two spokes merge after one spoke accelerates merging with the other spoke. We postulate the spoke transition model based on coupling between the current densities in the spoke, determined by the Child–Langmuir law, and Ar replenishment rates.

The transition takes place as the discharge current is increased, increasing the current through the spoke. Higher current through the spoke results in larger spoke and enhanced

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**Figure 10.** (a) Phenomenological model of spoke, from Gallian *et al.* [35]. The Gaussian shaped electron distribution $n_e$ represents the plasma density in the spokes. Neutral density $n_n$ profile represents diffusion of Ar gas between the spokes. (b) Drawing of azimuthal $n_e$ and $n_n$ distribution in case of two spokes, based on the phenomenological model of spoke. (c) Drawing of azimuthal $n_e$ and $n_n$ distribution, with one spokes having increased electron density, representing higher current through the spoke, and reduced $n_n$ in the wake of the larger spoke, representing reduced Ar replenishment rate due to enhanced Ar rarefaction.
Ar rarefraction. In the wake of the larger spoke the Ar replenishment rate is reduced. The following spoke encounters reduced neutral density which leads to a smaller spoke propagating with higher velocity. The spoke gets smaller since fewer Ar neutrals (ionised in the spoke) are available for generation of secondary electrons, responsible for excitation and ionisation in the plasma. Higher spoke velocity in the $\mathbf{E} \times \mathbf{B}$ drift direction can be explained by reduced friction due to reduced neutral density, in analogy to the spokes rotating faster at low pressures, compared to high pressures [9].

In the present study we have managed to demonstrate that the symmetry and self-organisation of the spokes is determined by electrostatic repulsion of the spokes. The symmetry is violated when enhanced charge diffusion prevails over the electrostatic repulsion. We postulate the spoke transition model where charge diffusion is enhanced due to asymmetry induced by increase of the discharge current. Increase of discharge current leads to a larger spoke, reduced neutral density in the wake of spoke, and merger of two spokes into one.

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**References**


Figure 11. Example of plasma self-organisation recorded by four cameras triggered consequently with 1 $\mu$s delay. The dashed line denotes a magnetron symmetry with respect to the unaltered spoke. HiPIMS discharge with Al target, 50 A, 0.5 Pa.


