

1 **Abstract**

2 The localized target current density associated with quasi-periodic ionization zones (spokes)
3 have been measured in a High Power Impulse Magnetron Sputtering (HiPIMS) discharge using
4 an array of azimuthally separated and electrical isolated probes incorporated into a circular
5 aluminum target. For a particular range of operating conditions (pulse energies between 1 and 2 J
6 and argon pressures from 0.5 to 1.5 Pa), strong oscillations in the probe current density are seen
7 with amplitudes up to 60% above a base value. These perturbations, identified as spokes, travel
8 around the discharge above the target in the $\mathbf{E} \times \mathbf{B}$ direction. Using phase information from the
9 angularly separated probes the spoke drift speeds, angular frequencies and mode number have
10 been determined. During each HiPIMS pulse the spoke velocity is seen to increase, from
11 typically 6.5 km s^{-1} at a time when the total discharge current density approaches its peak value
12 to 10 km s^{-1} at the end of the pulse. Such an observation is consistent with the assertion of
13 Brenning and Lundin (2012 *Phys. Plasmas* **19** 093505) that spoke velocities correspond to the
14 critical ionization velocity (CIV), which changes as the plasma composition changes. The largest
15 fluctuations in the strip probe signal were observed at an argon pressure of 0.68 Pa and pulse
16 energy of 1.79 J. In this particular case, the temporal angular frequencies vary from 3.2×10^5 to
17 $4.5 \times 10^5 \text{ rad s}^{-1}$. From the shape of individual current density oscillations, it appears that the
18 leading edge of the spoke is associated with a slow increase in local current density to the target
19 and the rear with a more rapid decrease. The measurements show that the discharge current
20 density associated with individual spokes is broadly spread over a wide region of the target.

21

1 **1. Introduction**

2 High power impulse magnetron sputtering HiPIMS is gaining interest from the scientific,
3 engineering and industrial communities as an exciting and potentially valuable physical vapour
4 deposition technique for the production of high-quality functional thin films ¹. At its heart,
5 HiPIMS is a merger of pulse power and conventional magnetron sputtering technologies which
6 facilitates the production of extremely dense ($\sim 10^{19} \text{ m}^{-3}$), short lived ($\sim 100 \mu\text{s}$) plasmas with a
7 high fraction of the sputtered species arriving as ions at the substrate ². With target power
8 densities often up to 10 kWcm^{-2} during the main sputtering phase the typical duty cycles are kept
9 short ($\sim 1\%$) to prevent overheating of the device.

10

11 HiPIMS systems, as with all sputtering magnetrons, rely on electron trapping in mutually
12 perpendicular electric \mathbf{E} and magnetic fields \mathbf{B} to provide high ionisation rates at low operating
13 pressures ($\sim 0.1\text{-}10 \text{ Pa}$) ³. Typically, with vacuum magnetic fields up to $\sim 0.05 \text{ T}$ in the magnetic
14 trap, the bulk plasma electrons (of temperature a few eV) can be considered magnetized and
15 therefore execute Hall drifts while the heavy positive ions should be subject only to free fall in
16 the \mathbf{E} fields to the target. Any component of fast secondary electrons accelerated in the cathode
17 fall, will execute a cycloid motion with radius of curvature typically less than 10 mm and can
18 also be considered magnetised. This principle of intrinsic electron drift has been studied and
19 exploited in other technological plasmas such as homopolar plasmas ⁴, Hall thrusters ⁵, and Q-
20 machines ⁶ and other linear plasma devices ⁷.

21 Such configurations, in which considerable azimuthal (Hall) electron current flows against a
22 static background of ions in a closed loop $\mathbf{E}\times\mathbf{B}$ drift channel, are characterised by a great many
23 inherent plasma oscillations and instabilities over a wide range of frequencies, $10^4 - 10^{10} \text{ Hz}$ ⁸.
24 These can include, drift wave instabilities ⁶, modified two-stream instabilities (MTSI) ⁹ lower-
25 hybrid waves ¹⁰, as well as, in some discharge conditions, coherent plasma structures which
26 rotate around the discharge in the Hall $\mathbf{E}\times\mathbf{B}$ drift channel ^{11,12}.

27 In HiPIMS, such structures have been observed, and are usually referred to as either bunches ¹³,
28 spokes ¹⁴ or ionization zones ¹⁵. They are believed to be regions of higher local plasma density
29 that rotate along the drift channel in the $\mathbf{E}\times\mathbf{B}$ direction at speeds consistent with the critical
30 ionization velocity (CIV). This velocity depends on the ionisation potential and mass of the

1 species forming the majority of ions in the plasma ¹⁴. Experimental determined spoke velocities
2 (in the range 2 - 10 km s⁻¹) for a range of different HiPIMS target materials and sputtering gases
3 agree well with the predicted CIV speeds ¹⁶ and are always about an order of magnitude lower
4 than the $\mathbf{E} \times \mathbf{B}$ electron drift speed ¹⁵. With closed loop distances around the racetrack of several
5 tens of cm's and typical mode numbers m of the instabilities observed from m = 1 to 4, the
6 speeds of such structures correspond to frequencies of several 100's kHz ^{17,18}.

7 To study and characterize the detailed nature of spokes in HiPIMS discharges a number of
8 diagnostic techniques have been employed. These include the use of fast intensified cameras
9 (ICCD's) to determine their shape, mode number, wavelength, rotation speed, and the general
10 conditions of existence for such rotating instabilities ^{8,13,15,17-21}. Other non-perturbing optical
11 techniques including two fiber optical monitoring ¹⁸ and optical emission spectroscopy ¹⁹ have
12 been used to both observe spoke motion and learn more about the composition of emitting
13 species including Al I, Al II, Ar I and Ar II.

14

15 Electrical probes, operating in either the electron or ion saturation current regions of the
16 characteristic, positioned in the magnetic trap have been used to observe the modulation in the
17 plasma density as spokes intercept the probe ^{13,22}. Such probes, of varying geometry, have also
18 been used in more remote positions to detect long range signatures of the spokes ²³. Electrically
19 isolated ^{18,22} and emissive probes ¹⁵ have been used to determine the local fluctuation of the
20 floating and space potentials due to the presence of spokes. However, in some cases the probes
21 themselves can perturb the plasma under observation.

22

23 The existence of spokes has been linked to observations of enhanced ion energies in HiPIMS.
24 Using energy-resolved mass analyzers a number of studies have shown that when such
25 instabilities exist in the plasma (seen by probe detection), there are pronounced extensions in the
26 energy of post-ionized sputtered species arriving at the substrate ^{24,25} but also remarkably in
27 directions tangential the target ²³.

28

29 These observations have led to the idea that associated with the spokes there must be a region of
30 more positive plasma potential (i.e. a potential humps of 10–50 V) generated by regions of

1 differing space charge. This potential structure can accelerate ions (created in the spoke) to
2 higher energies in all directions away from the target ^{24,25} however enhanced in the direction of
3 spoke rotation ²³, with the positive potential structure of the spoke acting like an impenetrable
4 moving wall to on-coming ions ¹⁴.

5
6 Conditions for the existence of spokes has been investigated as a function of discharge current ²⁶,
7 applied power ²¹, target power density ²⁴, and pressure ¹⁸. It is found that the mode number m
8 decreases with increased power until a threshold is reached after which no spokes can be
9 identified ²¹. Increasing pressure slows the spokes velocity ¹⁸.

10
11 To prevent a discontinuity of electric potential around the racetrack the spoke itself must consist
12 of two adjacent double layers ²⁷ (actually a triple layer structure as seen in other types of plasmas
13 ²⁸). Such layers consist of three regions of alternating space charge polarity, with a reversal in the
14 internally generated azimuthal electric field components \mathbf{E}_θ . These E-fields cross with the
15 vacuum \mathbf{B} -field to produce electron drifts across the magnetic field lines, perpendicular to the
16 plane of the spoke motion ¹⁴. This phenomenon has been observed using streak cameras ^{15,29} as
17 the creation of electron flare or jets emanating from the center or possibly the leading edge of the
18 spoke, a region of the spoke where \mathbf{E}_θ points in the $\mathbf{E} \times \mathbf{B}$ direction ²⁷. It is conjectured that these
19 flares can carry away a substantial part of the total electron current eventually reaching the
20 grounded walls. Using segmented anodes in a Hall thruster device the electron return current
21 associated with a single spoke was found to be over 50% of the total discharge current ³⁰. Such
22 cross-field electron transport has been associated with the mechanisms behind anomalous
23 electron diffusion seen in Hall thrusters ³¹; appropriate to all $\mathbf{E} \times \mathbf{B}$ devices which can drive axial
24 two-stream instabilities ¹⁴.

25
26 In the triple layer model advocated in ^{24,25} and updated recently by ²⁷ the azimuthal electric field
27 must also reverse and so azimuthally drifting electrons in the $\mathbf{E} \times \mathbf{B}$ channel entering the rear of
28 the spoke will be directed towards the cathode ²⁷, however, overall the net electron drift from all
29 regions of the target must be to the anode of course. This drift was observed in a PIC-MCC
30 simulation model of a concentric cylindrical homopolar plasma by Boeuf and Chaudhury ¹².

1 Since it believed spokes are high density regions with higher associated ionization rates it is
2 logical to assume that in HiPIMS (as well as other similar discharges) they give rise to a
3 considerable fraction of the total measured discharge current. This was assumed in the
4 calculations of ion current to the target from spokes in ¹³ as well as the basis for the electron
5 current flowing in the opposite direction towards ground (vessel walls) through flare formation ²⁵
6 as discussed above.

7
8 Plasma parameters such electron temperature and density associated with the spokes have been
9 obtained in Hall Thrusters ³⁰ using flush mounted planar probes, however, such measurements
10 with electrical probes in HiPIMS are problematic, for instance they can potentially destroy the
11 phenomena to be measured, and the probes themselves can accumulate a considerable amount of
12 deposit on the tips, corrupting their operation. Here we do not attempt to measure the plasma
13 parameters within spokes themselves, however employ an array of electrical probes (strips)
14 forming a segmented target to measure directly the distribution of current sourced from
15 individual spokes as they rotate around the racetrack. The strip probes, made of the target
16 material, were mounted flush to the target surface, making them indistinguishable from the rest
17 of the target as far as the plasma is concerned. Using this arrangement, we also measure the
18 spoke velocities, mode number and azimuthal profile (shape) during the HiPIMS pulse.

19

20 **2. Experimental setup**

21 All experiments were conducted in a purpose-built cylindrical vacuum vessel, 300 mm in length
22 and 260 mm in diameter, pumped down to a base pressure of 6.5×10^{-4} Pa using a turbo molecular
23 pump backed by a rotary pump. Argon was introduced into the vessel with flow rates ranging
24 from 4 to 32 sccm (standard cubic centimeter per minute) to vary the operating pressure between
25 0.27 Pa – 2.4 Pa. The operating pressure and flow rates were determined and controlled using a
26 capacitance pressure gauge (MKS 628A) and a mass flow controller (MKS 1179A), respectively.
27 The general chamber configuration is shown in figure 1a.

28 The magnetron used in this study was a circular unbalanced type, equipped with a 75 mm
29 diameter aluminum target of 5.53 mm thickness and 99.995% purity. The magnetic field strength
30 on the target surface at the center of the racetrack (21.5 ± 0.5 mm radius) was ~ 80 mT.

1 To strike HiPIMS plasmas an in-house built power supply was used, providing a peak current of
2 60 A ³². In this study the supply was operated at a repetition rate of 3 Hz and a pulse width of ~
3 $70 \text{ }\mu\text{s}$ providing energies up to 2.15 J per pulse.

4 In order to measure the target current locally as the spokes rotate above it, the target was
5 segmented, with the introduction of three $2.14 \pm 0.02 \text{ mm}$ wide flush-mounted aluminum strip
6 probes (same purity as the target) placed in machined slots at 3 angular positions (45° from each
7 other) around the target, see figure 1b. The three strip probes filled the entire target radius and
8 extended past the target edge by 5 mm to allow electrical connections. The stripe probes were
9 electrically isolated from the rest of target by a thin layer of polyimide tape of thickness of 0.07
10 $\pm 0.01 \text{ mm}$, placed around 3 sides of the probe, it provided a tight fit. This insulation thickness
11 defined the gap w_{gap} between the strip probe and the target.

12 We argue that, from a plasma perspective, the stripe probes are indistinguishable from the rest of
13 the target provide w_{gap} is smaller than both the cathode sheath thickness s and the gyro-radius r_{eL}
14 of bulk electrons and secondary electrons emitted from the target. From the literature³³, we can
15 take the electron densities and temperatures close to the target to be $n_e \sim 10^{19} \text{ m}^{-3}$ and $(k_B T_e)/e \sim$
16 3 eV respectively. With a cathode fall voltage of $V_0 \sim 500 \text{ V}$ and magnetic field strength $B \sim 80$
17 mT we calculate $s \sim 0.15 \text{ mm}$ ³⁴ and $r_{eL} \sim 0.94 \text{ mm}$ ³⁵, satisfying the conditions $w_{\text{gap}} < s$ and w_{gap}
18 $< r_{eL}$. The gyro radii of high-energy electrons accelerated to a fraction of the cathode fall
19 potential ($\gg kT_e$) will strongly satisfy the condition $w_{\text{gap}} < r_{eL}$.

20 The three strip probes were connected directly the power supply as shown in figure 1b. In this
21 configuration they always remained at the same potential as the target, but with their contribution
22 to the total current measured separately using Pearson current probes (Model 2877 with 1 V/A).
23 The total target current, I_d , was measured using a larger Pearson probe (Model 3972 with
24 0.1 V/A). The four current waveforms were recorded using a digital oscilloscope (Tektronix DSP
25 3030D with 300 MHz bandwidth) in a single shot mode if not otherwise stated. The discharge
26 voltage waveforms V_d were measured separately using a high voltage probe (Tektronix P5100).

27 The area of each individual strip probes and whole target (including the probes) were calculated
28 to be $A_p \sim 0.61 \text{ cm}^2$ and $A_d \sim 44.18 \text{ cm}^2$ respectively. Together with the measured currents from

1 the strips I_p and the total target current I_d it is possible then to determine the respective current
2 densities j_p and j_d . However, to accurately compare j_p and j_d (as we do in the first part of section
3 2), it is important that these measurements agree in “spoke-free” conditions. Since the strip
4 probes are rectangular in shape and inserted into essentially circular geometry and that they
5 overhang the target at the edges to allow electrical connections, we cannot guarantee any “ridged
6 body” plasma structure moving above them will see exactly the same proportion of their entire
7 areas A_p and A_d . So, to effectively calibrate the strip probes, we ran the magnetron over a range
8 of DC conditions ($p_{Ar} = 0.78$ Pa, 100 W < power < 300 W) in which no spokes were observed,
9 and calculated the current densities j_p and j_d . This provided us with an effective area ratio of
10 $A_p/A_d \sim 0.0170$, 20 % higher than the geometric areas. One such note that although no spokes
11 were observed here in DC conditions, the signature of spokes have indeed been detected in DC
12 sputtering elsewhere³⁶. The conditions required to generate spokes in non-HiPIMS plasmas are
13 yet to be properly investigated.

14

15 Since the strip probes were isolated from the target, effective cooling of the probes over long
16 durations was problematic. To prevent overheating of the probes we used a very low frequency
17 of 3 Hz, while still applying high peak powers. With low associated duty cycles it was necessary
18 to use an auxiliary pre-ionizer source³² in conjunction with the HiPIMS unit to easy breakdown
19 on each new pulse. Measurements of j_p taken at the beginning and end of long periods of
20 operation showed no differences, indicating that if any overheating did occur, it did not affect the
21 results.

22

23 **3. Results and Discussion**

24 Figure 2 shows simultaneous measurements of the current density at a strip probe j_p and the
25 current density to the whole target (which includes the strip probe) j_d . We have restricted
26 ourselves to observations in the driven phase of a number of different HiPIMS discharges for a
27 variation of pulse energies E_p (0.15 to 2.2 J) and gas pressures p_{Ar} (0.18 to 1.59 Pa). For this
28 particular part of our investigations, it was necessary to use the effective area ratio calibration as
29 described in section 2.

1 The data was obtained using the probe marked at angle $\theta = 0$ in figure 1b. Under certain
2 conditions we observe strong oscillations in j_p which we attribute to the existence of coherent
3 spoke structures. At the low pressures ($p_{Ar} = 0.18$ Pa) and low pulse energies ($E_p < 0.5$ J),
4 oscillations appear somewhat chaotic with irregular and non-periodic peaks and valleys during
5 the pulse. However, as the discharge energy is increased more coherent periodic structures
6 develop with oscillation amplitudes growing monotonically with E_p . There appears to be a
7 threshold in E_p for coherent spokes to form that decreases with increases in pressure. For
8 example, in figure 2b and 2c we see at $p_{Ar} = 0.81$ Pa a threshold value of $E_p \sim 1$ J, however this
9 decreases to $E_p = 0.6$ J at $p_{Ar} = 1.59$ Pa.

10 The largest amplitude in the j_p oscillations (seen at $E_p = 1.8$ J, $p_{Ar} = 0.81$ Pa) is about 45% above
11 the base level (the j_p values of in the valleys) at 30 - 40 μ s into the pulse (figure 2b). In contrast
12 to the strip probe measurements, the total discharge current density j_d measurements at any
13 operating condition show little or no perturbations. This can be expected, since the current
14 contribution from any non-changing structure moving around the whole target will yield no
15 temporal dependency in the I_d measurement. The very small oscillations seen in j_d , discernable in
16 figure 2b and reported in the discharge current elsewhere²⁴, may be due to the temporal
17 evolution of the spoke plasma parameters in the frame of reference rotating with the spokes.
18 Although we have no optical imaging evidence for spokes in this study, the lack of oscillations in
19 j_d but strong oscillations in the j_p measurements gives us confidence we are observing moving
20 plasma structures passing over our strip probe and not any kind superimposed signal from the
21 plasma-power supply network.

22

23 To illustrate that the presence of spokes is conditional on the discharge energy E_p and to allow us
24 to see in detail the form of the j_p oscillations, measurements for three discharge conditions have
25 been made and are shown in figure 3. Here we choose pulse energies E_p of 0.11, 0.87 and 2.20 J
26 at a single operating pressure of $p_{Ar} \sim 1.30$ Pa.

27 During the early part of the pulse at low pulse energy, ($t < 30$ μ s, $E_p = 0.11$ J) we see no signature
28 of spokes in the j_p traces. This is in agreement with fast imaging evidence from other HiPIMS
29 plasmas operating in similar conditions²⁰. However, as the current increase even at this low pulse

1 energy, we can observe a fluctuation in the strip current waveform from about 40 μs to 65 μs ²⁰.
2 Increasing the pulse energy to $E_p = 0.87$ J reveals strong oscillations in j_p . It also seems clear that
3 in the early part of the pulse (i.e. at low discharge current) the plasma is homogeneous, which
4 has been seen elsewhere ²⁰. Here, strong oscillations in j_p start at $t \sim 15$ μs and remains
5 throughout the pulse. In figure 3 the dashed lines represent an effective oscillation envelop
6 within which the peak-to-valley amplitude of the oscillation Δj_p , can be determined. We define
7 the bottom of the valley as the base current j_b . For example, at $t \sim 40$ μs , $\Delta j_p \sim 0.39$ A cm^{-2} ,
8 which corresponds to about 32% of the base current density ($j_b \sim 1.20$ A cm^{-2}). The ratio $\Delta j_p/j_b$
9 varies during the pulse, rising to a plateau of ~ 0.32 at $t \sim 40$ μs , persisting up to the end of the
10 pulse. At the highest pulse energy condition, $E_p \sim 2.20$ J, oscillations are significantly attenuated
11 with $\Delta j_p \sim 0$, particularly during times of peak discharge current I_d ($t \sim 30$ – 40 μs).

12 The peak-to-valley strip probe current densities normalized to the base current (valley) $\Delta j_p/j_b$ are
13 shown in figure 4. The measurements were obtained at one particular time during the pulse ($t \sim$
14 $40\mu\text{s}$) and are mapped out as a function of pulse energy and operating pressure. The maximum
15 value of $\Delta j_p/j_b$ is ~ 0.6 at $E_p \sim 1.60$ J and $p_{\text{Ar}} \sim 0.7$ Pa. In figure 4, we represent an approximate
16 boundary which separates two regions, one where spokes appear to be chaotic and one where
17 they are coherent. It is at low E_p (typically < 1 J) where chaotic behavior is generally observed as
18 reported by Winter et al ⁸.

19 One should note that due to limitations in the power supply some operating parameters could not
20 be attained and these are represented by the shaded region in figure 4. Despite this experimental
21 limitation the contour plot does clearly reveal regions where spokes are chaotic, coherent and
22 non-existent. Our observations are in general agreement with those obtained using fast optical
23 imaging ²¹ to investigate spoke structural evolution.

24 The mechanism determining spoke formation is unknown but it appears that at low power only
25 chaotic structures are observed and their amplitude is largely independent of pressure. However,
26 once coherent spokes are created pressure plays a key role in determining the current oscillation
27 amplitude above its base value. It is argued that collective behavior such as spoke formation is a
28 result of insufficient ion production, with structures moving to seek regions of enhanced
29 ionization ¹⁵. At high powers, sufficient ionization takes place and spoke structure is lost.

1 Individual spokes may merge to form a single continuous structure ²¹, possibly maintaining a
2 reversal in axial electric field (potential fill) but with no azimuthal component. Increased
3 pressures may provide the necessary sustainment and hence reduced spoke current amplitudes.

4 To gain information on the speeds of the spokes and their mode number j_p measurements were
5 made simultaneously at three strip probes at $\theta = 0, \pi/4$ and $\pi/2$. An example taken for the case E_p
6 ~ 1.79 J and $p_{Ar} \sim 0.68$ Pa is shown in Figure 5. The j_p waveforms have been stacked (with a
7 vertical increment of $j_p = 1$ Acm⁻²) to allow the phase information to be seen clearly. The j_p
8 oscillations between probes are coherent in terms of shape, amplitude and period but phase
9 shifted (a time lag τ) with increasing θ . This demonstrates the existence of essentially coherent
10 (and rigidly-formed) current perturbations, (ionization zones) rotating in the anticlockwise $\mathbf{E} \times \mathbf{B}$
11 direction. In the particular case chosen, over the driven part of the discharge, we can identify
12 seven distinct oscillation packages as labelled I to VII in figure 5. To calculate the spoke angular
13 frequency, ω and tangential speed, v at the racetrack center (radius $r_c \sim 21.5 \pm 0.5$ mm) average
14 time delays τ_{avg} in the peaks have been used. We define $\tau_{avg} = (\tau_{AB} + \tau_{BC})/2$, where τ_{AB} and τ_{BC} is
15 the time delay between probes A and B and probes B and C, respectively. The spoke speed
16 calculations are summarized in table I.

17 From the average period of oscillation T_{avg} and the time lag τ_{avg} data the mode number m can be
18 determined as $m = (2\pi\tau_{avg})/(\Delta\theta T_{avg})$. In our chosen operating condition ($p_{Ar} \sim 0.68$, $E_p \sim 1.79$ J),
19 we find values of m between 2.8 and 3.1 giving us confidence that we have $m = 3$.

20 It is known from the literature that the mode number may vary with operating conditions, for
21 instance with higher m observed for increased pressure (for Ti) ¹⁸, increasing from $m = 1$ to a
22 more fragmented situation with $m > 4$. The mode number tends to decrease with increasing
23 discharge current as seen in the sputtering of Mo ²¹ and Al ²⁰ targets. Over all the conditions in
24 this study we find predominately $m = 3$. At certain conditions the $m = 4$ can be identified (for
25 instance at $p_{Ar} = 1.6$ Pa and $E_p = 0.8$ J). We do not observe the large $m = 1$ structure even at low
26 powers (when spokes exist) as observed with Al targets elsewhere ²².

27 It is interesting that in similar conditions to ours ($p_{Ar} = 0.27$ Pa and $j_d > 1.8$ A cm⁻²) with an Al
28 target the $m = 4$ mode rather than $m = 3$ is seen ²⁰. More work is needed to understand the form
29 of rotating spoke patterns and their link to discharge conditions.

1 The tangential speeds shown in Table I, lying between 6.95 and 9.79 km s⁻¹ agree well those seen
2 in other HiPIMS discharges with Al targets¹⁵. We observe that for conditions when spokes
3 actually occur, their speed increases during the HiPIMS pulse, for instance from ~6.95 km s⁻¹ (t ~
4 22–30 μs) to ~9.76 km s⁻¹ (t ~ 56–60 μs) as seen in figure 5 for E_p ~ 1.79 J and p_{Ar} ~ 0.68 Pa.

5 To determine any gross trends in the spoke speed v during the evolving HiPIMS pulse, the speed
6 of individual spokes has been calculated over ten identical HiPIMS pulses and plotted as a
7 function of time in figure 6. This has been done for only one discharge operating condition (E_p ~
8 1.79 J and p_{Ar} ~ 0.68 Pa) but the same trends are seen at different conditions for which spokes
9 exist.

10 We see that as the pulse progresses v increases from about 6.5 to 10 km s⁻¹. This increase can
11 partly be understood from a change in composition of the plasma. It is postulated that spoke
12 speeds adhere to the critical ionization velocity CIV hypothesis¹⁶, with the CIV speed given by

13
$$v_{CIV} = \sqrt{\frac{2eU_i}{m_n}}$$
, where U_i is the ionization potential and m_n is the mass of the species being ionized

14 with e the elementary charge. As the discharge builds up and certainly at times close to the peak
15 discharge current we expect the discharge to be dominated by Al giving a CIV velocity $v_{CIV,Al} =$
16 6.5 km s⁻¹. However towards the end of the pulse we conjecture that argon replaces the Al
17 particles and the CIV velocity will therefore increase to $v_{CIV,Ar} = 8.7$ km s⁻¹. The results in figure
18 6 suggest such a trend as the weighted mass m_n and effective U_i value change with composition.
19 The CIV limits for the two species are shown in figure 6. For times greater than 50 μs our
20 calculated spoke velocities exceed the CIV velocity for Ar. One possible explanation for this is
21 that as the spoke evolves in time its' average radial position (as well as shape) may change. If the
22 spokes average radius (correlated with the position of maximum current) were to reduce by about
23 3 mm our calculated velocities would always be limited to $v_{CIV,Ar}$. Our estimated spoke velocities
24 compare well to those obtained in other HiPIMS plasmas with Al targets, for instance a velocity
25 of 8.1 ± 0.3 km s⁻¹ determined by fast camera imaging¹⁵.

26 This argument of changing composition is consistent with measurements made using time-
27 resolved tunable diode-laser adsorption³⁷, where Ar depletion was found to occur at the peak of
28 the target current. In the early stages of the discharge it is difficult to identify and then calculate

1 spoke speeds, so we are unable to demonstrate that spoke speeds are consistent with the $v_{\text{ClV.Ar}}$
2 velocity as we would expect this stage in the discharge before Al becomes the dominant species.

3 From observation of the time profile in j_p (as a spoke passes over a probe) useful information on
4 the spoke structure can be obtained. From figure 5 taking a representative spoke (peak II at probe
5 position $\theta + \pi/2$) between 30 and 35 μs we see at the front of the spoke there is a relatively slow
6 two-stage rise in j_p (slew rates $\sim 7.8 \times 10^4$ and $\sim 3.5 \times 10^5 \text{ A cm}^{-2} \text{ s}^{-1}$) to a peak value followed a
7 more rapid fall ($\sim 6.7 \times 10^5 \text{ A cm}^{-2} \text{ s}^{-1}$). This profile is wholly consistent with an elongated
8 (triangular) shape as reported in the literature for spokes above an aluminum target^{20,22} in which
9 the apex of the spoke is pointed in the $\mathbf{E} \times \mathbf{B}$ direction, followed by a sharp trailing edge. This
10 edge is aligned with the direction of the radial magnetic field lines, connecting the inner and
11 outer magnetic pole pieces. This profile also fits with electrical probe data of the signatures of
12 spokes²² where the bulk of the evidence suggests that spokes are characterized by a gradual rise
13 (in density, potential, current) in the leading edge but a faster decrease at the rear.

14

15 Inspection of figure 5 shows very little gap (or time lags) between individual spokes. It was
16 expected that we would see a considerably lower current density between discrete spokes
17 representing a background of weaker homogeneous plasmas. It appears however that the spoke
18 structures are smeared out in the azimuthal direction almost merging nose-to tail but with each
19 spoke characterized by a region of high current densities (40-50% higher than at the periphery of
20 the spokes).

21

22 To demonstrate this we have attempted to display the current density data in 2D as done for
23 optical signals obtained from fast imaging. This can be achieved by transforming the temporal
24 distribution $j_p(t)$ to an angular distribution $j_p(\theta)$ and convolving it with a Guassian radial
25 distribution to artificially add radial information. We choose a Guassian function $G(r) = A \exp(-$
26 $(r-r_0)^2/L^2)$, where r_0 ($= 21.5 \text{ mm}$) is the radius of racetrack center, L ($= 10 \text{ mm}$) is an assumed
27 mean radial width of the spokes, and A is a scaling constant to be calculated. Although we have
28 no radial measurements of j_p in this study, the invoked Guassian representation of $j_p(r)$ centered
29 on the middle of the racetrack³⁸ is sufficient to produce a realistic 2-D distribution of target
30 current density $j(\theta, r)$, as shown in figure 7. Here we chose the current density data from figure 5

1 (probe at $\theta + \pi/2$) over one period T from 30 to 47 μs . To ensure our function $j(\theta, r)$ provides the
2 correct measured discharge current I_d it must satisfy the condition

3

$$\iint_{0,0}^{2\pi,R} j(\theta, r)r d\theta dr = I_d$$

4

5 Where $j(\theta, r) = G(r)j_p(\theta)$ and R is the target radius. Here the target current I_d is averaged over the
6 same time period ($30 \leq t \leq 47 \mu\text{s}$) as the $j_p(t)$ was collected. Performing the integral yielded, in
7 the particular case chosen, a scaling constant $A = 1.3$.

8

9 It is interesting that in figure 7 our constructed map of $j(\theta, r)$ is very much consistent in form to
10 optical emission fast imaging^{20,22}. However due to the lack of real radial information it cannot
11 reproduce for instance a triangular form to the spoke as often seen optically. It seems clear
12 however that spokes do carry a considerable fraction of the discharge current since their
13 elongated structure occupies a significant area of the target in the racetrack region. In the case
14 displayed the peak current density at the spokes center is highly localized and has a current
15 density $\sim 43\%$ greater than that in the racetrack minimum. Future studies using radial separated
16 target probes will yield accurate radial information allowing accurate 2D current density maps to
17 be constructed.

18

19 **4. Conclusions**

20 Using a segmented target the current densities associated with rotating spokes in a HiPIMS
21 discharge have been investigated. Spokes are identified as regular perturbations in local
22 discharge current density with peak values up to 50 % higher than the valley currents. Generally,
23 as the energy of the HiPIMS pulse is increased the spokes change their nature, moving from
24 chaotic behavior to form coherent regular structures. At our highest pulse energies spokes are
25 extinguished and may form a single continuous structure.

1 The spokes have been found to have an elongated leading edge and sharper trailing edge, and
2 travel with velocities increasing from 6.5 to 10 km s⁻¹ as the HiPIMS pulse develops. These
3 velocities are consistent CIV predictions.

4 The time signature of spoke current densities show that spokes are effectively smeared out to
5 cover a larger area of the racetrack leaving no discrete gaps in between them. Assuming a
6 realistic radial distribution of current density, a 2D map of the target current density $j(\theta,r)$ has
7 been constructed, showing a general distribution consistent with fast optical imaging of spokes in
8 other studies.

9 Further investigations using our method is planned to include the use of radially distributed
10 segments to provide information of the radial current density distribution. In addition, the
11 combined use of fast optical imaging and target probes will allow advances in the understanding
12 of spoke creation and sustainment for a variety of different target materials.

13

14 **Acknowledgements**

15 Phitsanu Poolcharuansin acknowledges support by Mahasarakham University, Thailand and
16 University of Liverpool, UK for his short-term postdoctoral fellowships. Francis Lockwood
17 Estrin would like to thank the University of Liverpool for his studentship funding, as part of the
18 Engineering and Physical Sciences Research Council (EPSRC) Fusion Doctoral Training
19 Network EP/K504178/1. He also wishes to thank the STFC, AsTEC Group, Daresbury, UK for
20 their funding support and Dr. Reza Valizadeh and Dr. Oleg Malyshev of the same for their
21 advice and encouragement.

22

23

24

25

26

1 **Figure Captions**

2 **Fig. 1:** Schematic diagrams of (a) the magnetron and chamber set-up and (b) the strip probes and
3 current measuring arrangements.

4 **Fig. 2:** (a), (b) and (c) time traces of the current density, j_p , measured by a strip probe, (d), (e)
5 and (f) time traces of the total target current density, j_d , for a number of pulse energies E_p and
6 argon pressures, p_{Ar} of 0.18, 0.81 and 1.59 Pa.

7 **Fig. 3:** The strip probe current densities j_p for a number of pulse energies at $p_{Ar} = 1.30$ Pa. The
8 marked envelop (dashed lines) is used as a guide to calculate the amplitude (Δj_p) of the
9 oscillation in j_p above the running minimum value j_b .

10 **Fig. 4:** A contour plot of $\Delta j_p/j_b$, the ratio of Δj_p normalized to the base current density j_b , at time
11 $t = 40 \mu s$ for a range of pulse energies E_p and argon pressures p_{Ar} . The hatched region represents
12 an area inaccessible to experiment due to power supply limitations. The dotted line represents the
13 approximate boundary between chaotic and coherent spoke behavior.

14 **Fig. 5:** The strip probe current density time traces for $E_p = 1.79$ J and $p_{Ar} = 0.68$ Pa. The traces
15 are stacked (incremented by 1 A cm^{-2} to allow the phase relationship to be seen). The rapid,
16 short-lived, negative current observed at the end the pulse is thought to be the result of finite
17 inductance of strip probe assembly and connecting lines.

18 **Fig. 6:** A plot of spoke velocities through a HiPIMS pulse, calculated from the time lags in the
19 waveforms between the azimuthally separated strips. The data was recorded over ten HiPIMS
20 pulses for $E_p = 1.79$ J and $p_{Ar} = 0.68$ Pa. The target current and voltage for this discharge are
21 also shown.

22 **Fig. 7:** A 2D reconstruction of the target current density using measured azimuthal data
23 combined with an assumed Gaussian radial current density distribution, of the form $G = A \exp(-$
24 $(r-r_0)^2/L^2)$. The azimuthal data was obtained for $p_{Ar} \sim 0.68$ Pa, $E_p \sim 1.79$ J, and for spokes passing
25 a single strip in the time period 30 - 47 μs .

26

1 **Table I:** The time lags between neighboring strips τ_{AB} , τ_{BC} and their average value τ_{avg} , angular
2 frequency ω and the tangential speed v at the racetrack center for each ionization zone identified
3 in figure 5.

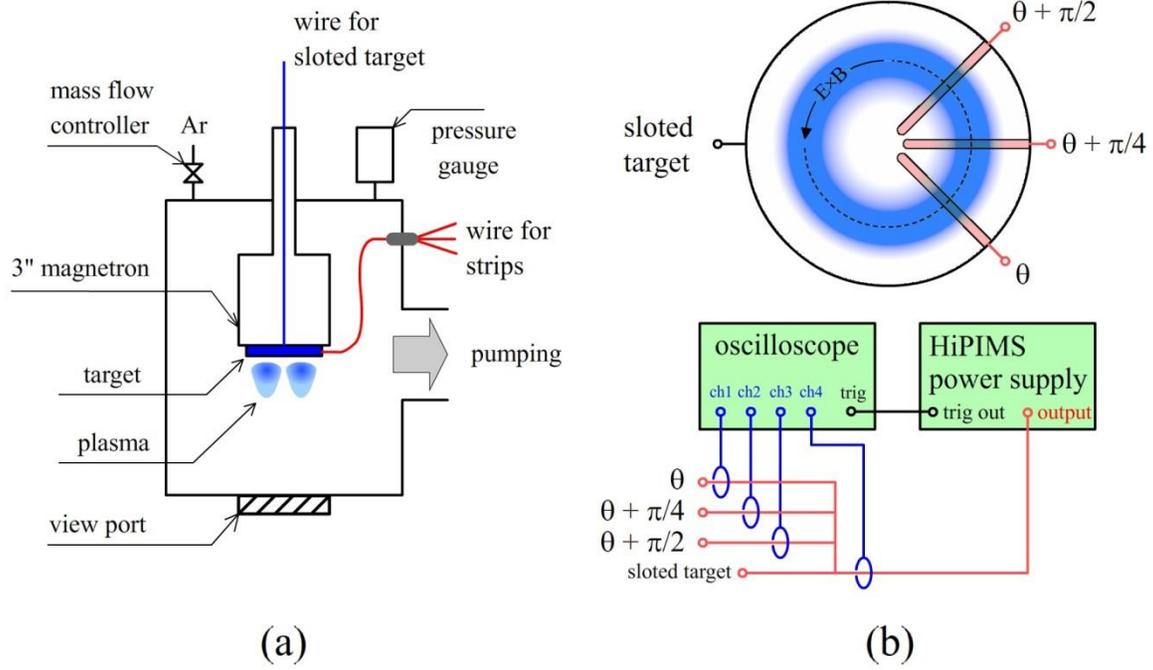
4

1 **References**

- 2 ¹ D. Lundin and K. Sarakinos, *J. Mater. Res.* **27**, 780 (2012).
- 3 ² J.T. Gudmundsson, N. Brenning, D. Lundin, and U. Helmersson, *J. Vac. Sci. Technol. A*
4 *Vacuum, Surfaces, Film.* **30**, 030801 (2012).
- 5 ³ A. Anders, *Surf. Coatings Technol.* **205**, S1 (2011).
- 6 ⁴ P.B. Barber, D. a Swift, and B. a Tozer, *J. Phys. D. Appl. Phys.* **5**, 693 (1972).
- 7 ⁵ V. V Zhurin, H.R. Kaufman, and R.S. Robinson, *Plasma Sources Sci. Technol.* **8**, R1 (1999).
- 8 ⁶ H.L. Pecseli, T. Mikkelsen, and S.E. Larsen, *Plasma Phys.* **25**, 1173 (1983).
- 9 ⁷ A.E. Shumack, H.J. de Blank, J. Westerhout, and G.J. van Rooij, *Plasma Phys. Control. Fusion*
10 **54**, 125006 (2012).
- 11 ⁸ J. Winter, A. Hecimovic, T. de los Arcos, M. Böke, and V. Schulz-von der Gathen, *J. Phys. D.*
12 *Appl. Phys.* **46**, 084007 (2013).
- 13 ⁹ D. Lundin, P. Larsson, E. Wallin, M. Lattemann, N. Brenning, and U. Helmersson, *Plasma*
14 *Sources Sci. Technol.* **17**, 035021 (2008).
- 15 ¹⁰ D. Lundin, U. Helmersson, S. Kirkpatrick, S. Rohde, and N. Brenning, *Plasma Sources Sci.*
16 *Technol.* **17**, 025007 (2008).
- 17 ¹¹ E.Y. Choueiri, *Phys. Plasmas* **8**, 1411 (2001).
- 18 ¹² J.P. Boeuf and B. Chaudhury, *Phys. Rev. Lett.* **111**, 1 (2013).
- 19 ¹³ A. V. Kozyrev, N.S. Sochugov, K. V. Oskomov, a. N. Zakharov, and a. N. Odivanova, *Plasma*
20 *Phys. Reports* **37**, 621 (2011).
- 21 ¹⁴ N. Brenning, D. Lundin, T. Minea, C. Costin, and C. Vitelaru, *J. Phys. D. Appl. Phys.* **46**,
22 084005 (2013).
- 23 ¹⁵ A. Anders, P. Ni, and A. Rauch, *J. Appl. Phys.* **111**, 053304 (2012).
- 24 ¹⁶ N. Brenning and D. Lundin, *Phys. Plasmas* **19**, 093505 (2012).
- 25 ¹⁷ A.P. Ehiasarian, A. Hecimovic, J. Winter, T.D.L. Arcos, R. New, V.S. Der Gathen, and M.
26 Böke, *IOP Conf. Ser. Mater. Sci. Eng.* **39**, 012012 (2012).
- 27 ¹⁸ A.P. Ehiasarian, A. Hecimovic, T. de los Arcos, R. New, V. Schulz-von der Gathen, M. Böke,
28 and J. Winter, *Appl. Phys. Lett.* **100**, 114101 (2012).

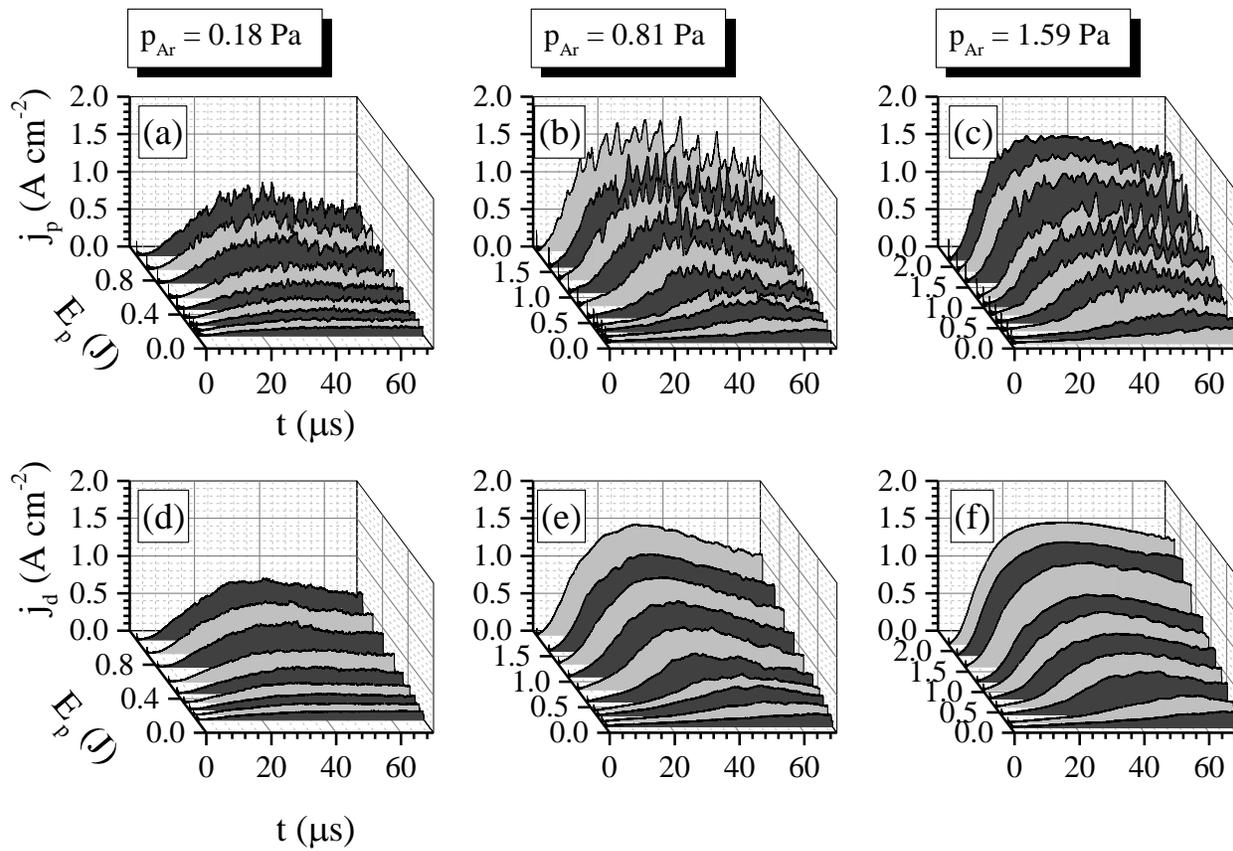
- 1 ¹⁹ J. Andersson, P. Ni, and A. Anders, *Appl. Phys. Lett.* **103**, 054104 (2013).
- 2 ²⁰ A. Anders, *Appl. Phys. Lett.* **100**, 224104 (2012).
- 3 ²¹ T.D.L. Arcos, V. Layes, Y.A. Gonzalvo, V.S. Der Gathen, a Hecimovic, and J. Winter, *J.*
4 *Phys. D. Appl. Phys.* **46**, 335201 (2013).
- 5 ²² a Hecimovic, M. Böke, and J. Winter, *J. Phys. D. Appl. Phys.* **47**, 102003 (2014).
- 6 ²³ M. Panjan, R. Franz, and A. Anders, *Plasma Sources Sci. Technol.* **23**, 025007 (2014).
- 7 ²⁴ C. Maszl, W. Breilmann, J. Benedikt, and a von Keudell, *J. Phys. D. Appl. Phys.* **47**, 224002
8 (2014).
- 9 ²⁵ A. Anders, M. Panjan, R. Franz, J. Andersson, and P. Ni, *Appl. Phys. Lett.* **103**, 144103
10 (2013).
- 11 ²⁶ T.D.L. Arcos, R. Schröder, Y.A. Gonzalvo, V.S. Der Gathen, and J. Winter, *Plasma Sources*
12 *Sci. Technol.* **23**, 054008 (2014).
- 13 ²⁷ A. Anders, *Appl. Phys. Lett.* **105**, 244104 (2014).
- 14 ²⁸ G. Petraconi and H.S. Maciel, *J. Phys. D. Appl. Phys.* **36**, 2798 (2003).
- 15 ²⁹ P. a. Ni, C. Hornschuch, M. Panjan, and A. Anders, *Appl. Phys. Lett.* **101**, 224102 (2012).
- 16 ³⁰ C.L. Ellison, Y. Raitses, and N.J. Fisch, *Phys. Plasmas* **19**, 013503 (2012).
- 17 ³¹ G.S. Janes, *Phys. Fluids* **9**, 1115 (1966).
- 18 ³² P. Poolcharuansin, B. Liebig, and J. Bradley, in *IEEE Trans. Plasma Sci.* (2010), pp. 3007–
19 3015.
- 20 ³³ A. Anders, J. Andersson, and A. Ehiasarian, *J. Appl. Phys.* **102**, 113303 (2007).
- 21 ³⁴ A.J.L. Michael A. Lieberman, *Principles of Plasma Discharges and Materials Processing*
22 (John Wiley & Sons, Inc., 2005).
- 23 ³⁵ A. Rauch and A. Anders, *Vacuum* **89**, 53 (2013).
- 24 ³⁶ A. Anders, P. Ni, and J. Andersson, *IEEE Trans. Plasma Sci.* **42**, 2578 (2014).
- 25 ³⁷ C. Vitelaru, D. Lundin, G.D. Stancu, N. Brenning, J. Bretagne, and T. Minea, *Plasma Sources*
26 *Sci. Technol.* **21**, 025010 (2012).
- 27 ³⁸ G. Clarke, A. Mishra, P.J. Kelly, and J.W. Bradley, *Plasma Process. Polym.* **6**, S548 (2009).

1 **Figures**



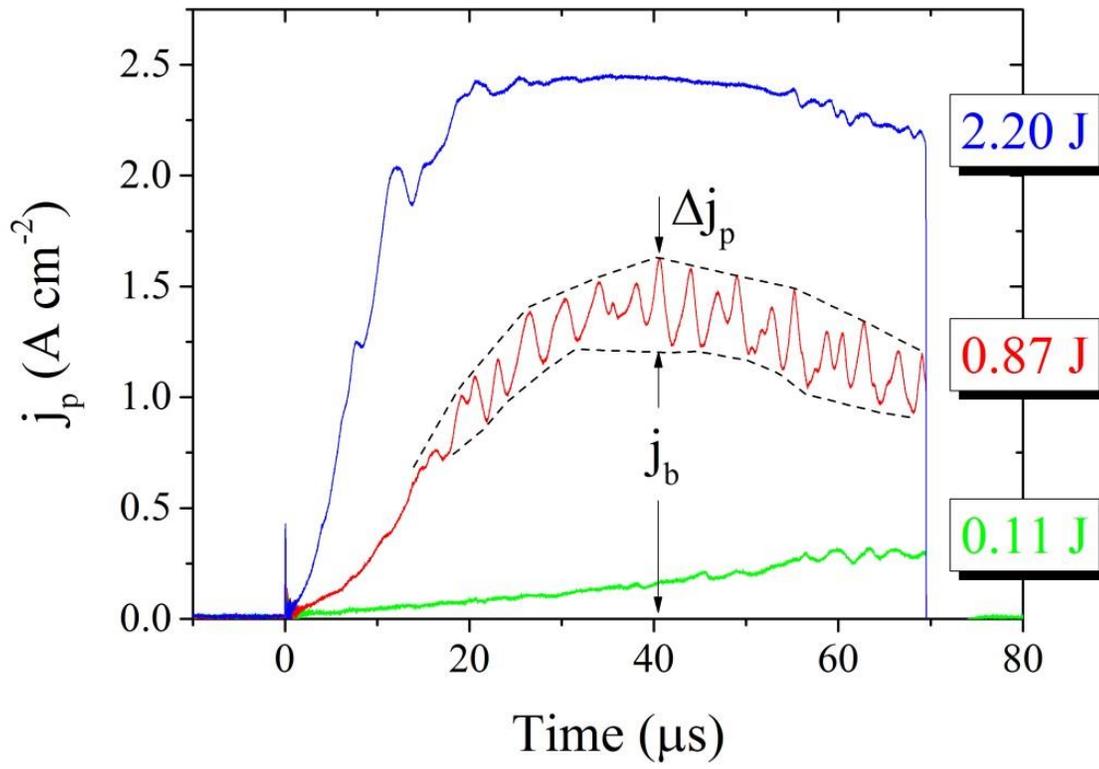
2
3
4
5

Figure 1.



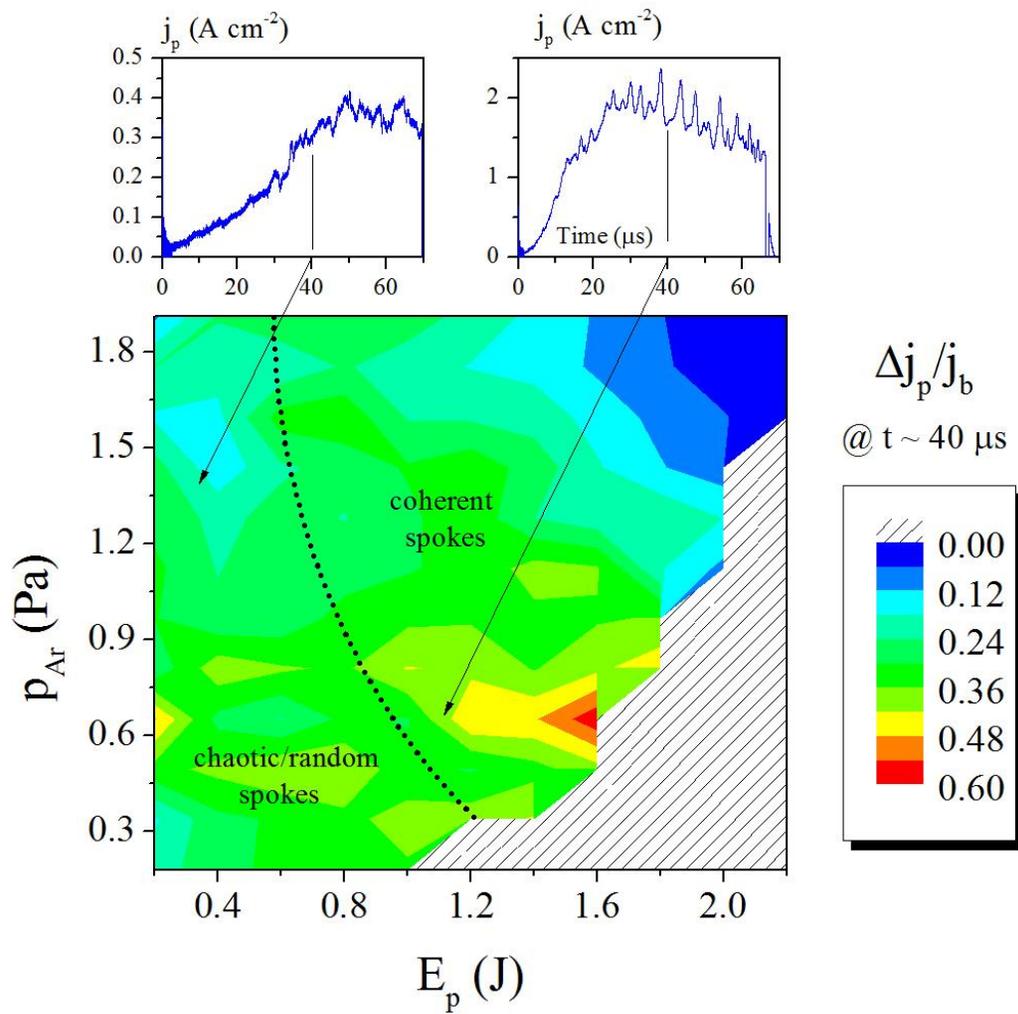
1

2 Figure 2.



1

2 Figure 3.

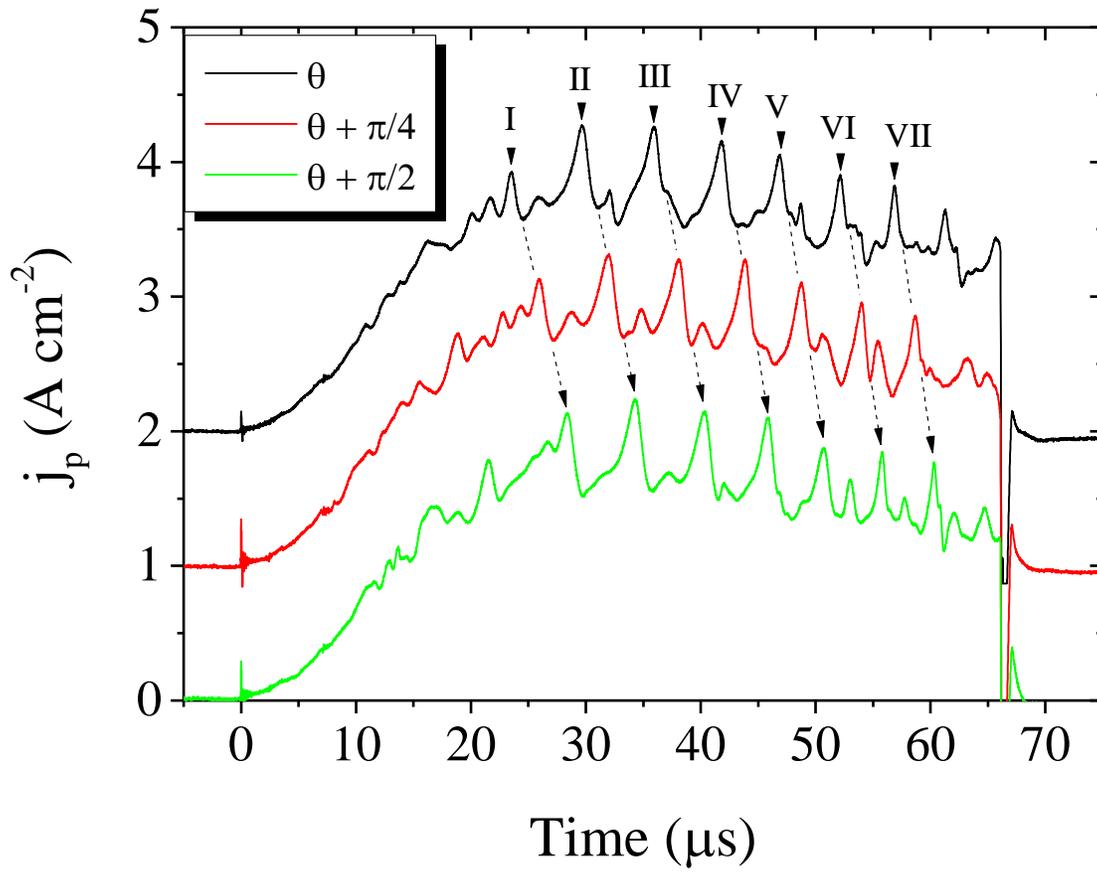


1

2

3

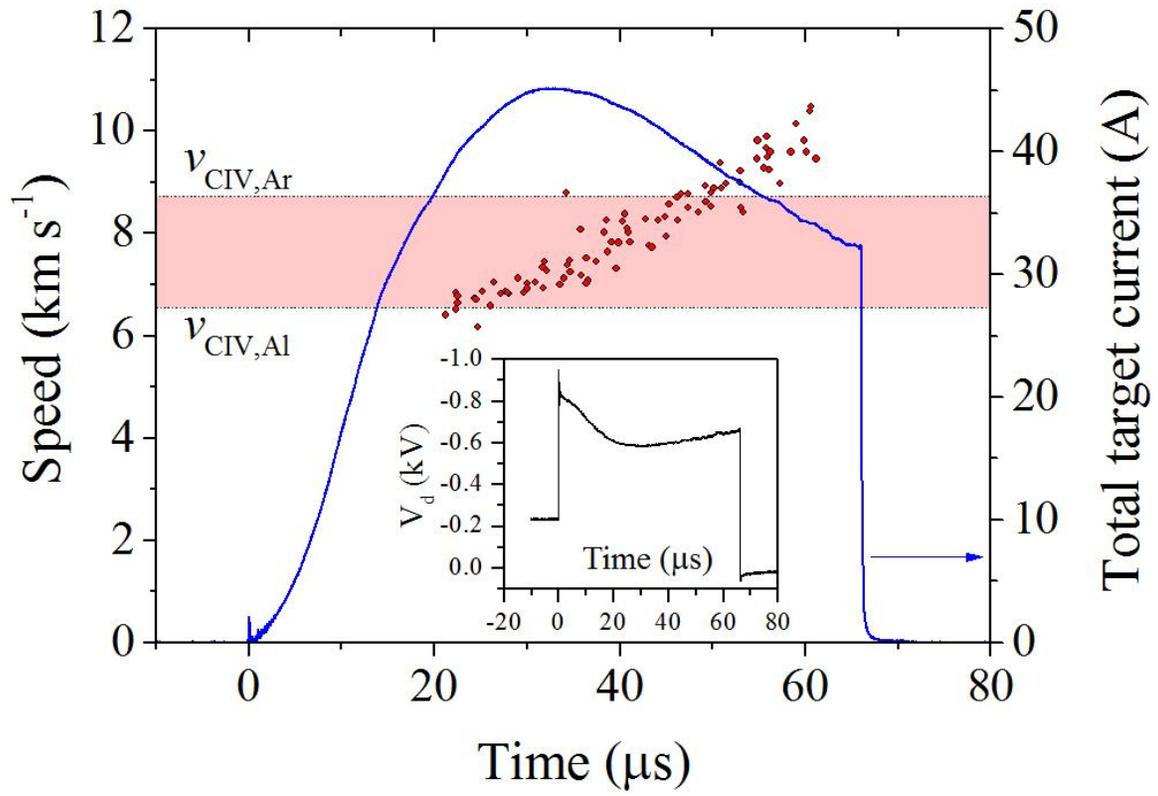
4 Figure 4.



1

2 Figure 5.

3

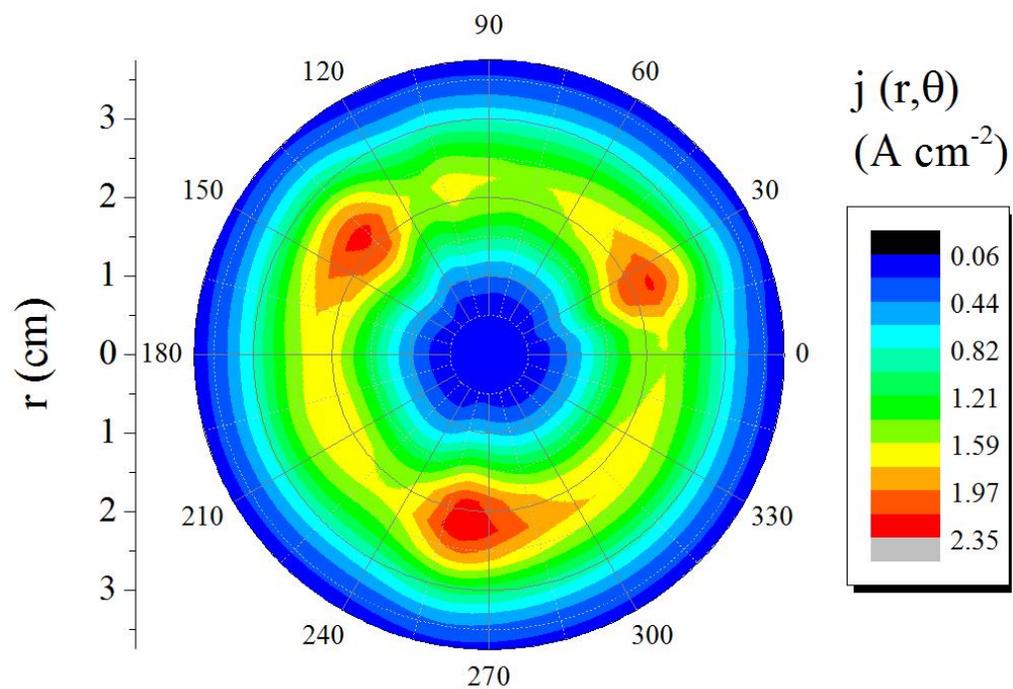


1

2 Figure 6.

3

4



1

2 Figure 7.

3

4

5

6

7

8

9

10

11

12

13

1

Peak	τ_{AB} (μs)	τ_{BC} (μs)	τ_{avg} (μs)	ω (rad s^{-1})	v (km s^{-1})
I	2.45	2.41	2.43	3.23×10^5	6.95
II	2.29	2.36	2.33	3.37×10^5	7.26
III	2.13	2.27	2.20	3.57×10^5	7.68
IV	2.04	2.02	2.03	3.87×10^5	8.32
V	1.96	1.91	1.94	4.05×10^5	8.73
VI	1.80	1.86	1.83	4.29×10^5	9.23
VII	1.67	1.78	1.73	4.54×10^5	9.79

2

3 **Table 1**

4