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# Anomalous electron transport in high power impulse magnetron sputtering

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#### Abstract

Oscillating electric fields in the megahertz range have been studied in a high power impulse magnetron sputtering (HIPIMS) plasma with the use of electric field probe arrays. One possible reason for these oscillations to occur is due to charge perturbation – or so called modified two-stream instabilities (MTSI). It is known that MTSI give rise to acceleration of the charged plasma species, and can give a net transport of electrons across the magnetic field lines. Measurements of these oscillations confirm trends, specifically of the frequency dependence on ion mass and magnetic field strength as expected from the theory of MTSI waves. These results help to explain the previously reported anomalous fast electron transport in HIPIMS discharges, where classical theory of diffusion, using collisions to transport electrons has failed.

#### 1 Introduction

Magnetron sputtering is an important industrial process for many applications ranging from deposition of hard coatings to deposition of functional coatings for electronic applications. These processes are constantly being improved to meet ever increasing demands of thin film performance. One such process improvement, the increase of ionization of the sputtered material leading to improved film quality, was demonstrated by Rossnagel and Hopwood [1] in 1993 by using a radio frequency coil to increase the plasma density. Since this time, a number of alternative ways to increase the plasma density and thereby the ionization of the sputtered material have been investigated. One of the most promising such techniques is high power impulse magnetron sputtering (HIPIMS), which was introduced by Kouznetsov et al. [2] in 1999 and recently reviewed by Helmersson et al. [3]. HIPIMS utilizes a standard direct current (DC) magnetron sputtering system and requires only alteration of the power supplied to the target to generate enhanced metal ionization, and hence is readily scalable, unlike rfcoil or filtered arc systems. The power in HIPIMS is supplied in short very intense pulses, typically with a target voltage between 400 and 2000 V and currents densities between 1 and 10 A cm<sup>-2</sup>, and applied in pulses with lengths between 10 and 100  $\mu$ s to avoid target damage and/or melting. By using very high momentary power to the magnetron, one can easily increase the resulting charge carrier density (plasma density) from  $10^9$  cm<sup>-3</sup> for conventional DC magnetron sputtering to  $10^{13}$  cm<sup>-3</sup> for HIPIMS [4]. Generating a high plasma density increases the probability for ionizing collisions, and thereby produces a large fraction of ionization of the sputtering gas as well as sputtered material, the latter 30-90 % depending on material [5]. This increased ionization of the deposition flux has been shown to be an important factor in producing sputter deposited thin films of superior density and behavior, for example in deposition of complex shaped substrates [6, 7] or in obtaining dense microstructures [6, 8].

The high potential used in HIPIMS generates large discharge currents in the plasma between the sputtering cathode and the anode (often the chamber walls and the ground shield of the magnetron). Large Hall currents, circulating in the magnetic field trap close to the magnetron, also arise due to the presence of the magnetic field ( $\mathbf{E} \times \mathbf{B}$  drift of electrons). Measurements by Bohlmark *et al.* [9] show that the ratio between the Hall current density,  $j_{H}$  and the discharge current density,  $j_{D}$  for HIPIMS discharges is unexpectedly low, typically  $j_{H}/j_{D}$ ~2. Classical theory of diffusion and electrical conductivity using collisions to move electrons across the magnetic field, or Bohm diffusion, usually results in  $j_{H}/j_{D}$  values of about 16-35 [9, 10]. Thus, a low  $j_{H}/j_{D}$  value indicates that electron transport across the magnetic field lines is more efficient in the HIPIMS case than can be explained by standard theory. It should be noted here that in reality the Hall current is only one contribution out of three to the total azimuthal current density in HIPIMS. For a more detailed analysis of the  $j_{H}/j_{D}$  ratio in the experiment described here the reader is referred to Appendix A, where the value  $j_{H}/j_{D}$ ~2 is attained, in agreement with Bohlmark's investigation on anomalous electron transport in HIPIMS plasmas [9].

One possible explanation for this enhanced electron transport may be a phenomena previously observed in plasma gun experiments by Brenning *et al.* [11]. In this work a fast cross-magnetic field transport of electrons was measured, and it was shown that this phenomena could be quantitatively described as being related and mediated by highly nonlinear waves, likely due to the modified two-stream instability (MTSI), resulting in electric field oscillations.

Similar oscillations have been observed in DC magnetron plasmas [12, 13] as well as in HIPIMS plasmas [2, 14]. A few of these reports also indicate electric field fluctuations in the plasma in connection with fast electron transport [13, 15], but the mechanisms responsible for current flow across the magnetic field lines have not yet been fully characterized. The intention of this work is to expand existing DC magnetron studies and models of plasmas to include fast electron transport phenomena (often referred to as anomalous electron transport) in HIPIMS discharges, with the hypothesis that the high cross-field electron transport is mediated by the MTSI. To measure plasma instabilities, multiple position electric field probes

were used and the results are compared with known waves capable of traveling within magnetized plasmas, and the anomalous transport properties of these waves.

#### 2 Experimental procedure

A standard planar circular 0.15 m magnetron equipped with a Ti (99.9 %) target was mounted in a cylindrical vacuum chamber (height 0.70 m, diameter 0.44 m) pumped with a turbomolecular pump to a background pressure of about 10<sup>-6</sup> Torr after which a few mTorr of an inert gas (He, Ar, Kr or Ne with a minimum purity of 99.9997 %) was leaked into the chamber. The magnetic field configuration has previously been documented by Böhlmark et al. [9]. The degree of unbalance can be calculated by measuring the distance from the center of the target surface to the magnetic null point (where  $B_z = 0$ ) and compare that to the radius of the magnetor. In the case of the magnetorn described above the magnetic null point is roughly 0.10 m away from the target surface, and we therefore conclude that the magnetron is weakly unbalanced.

Figure 1 shows a schematic of the vacuum chamber with an electrical field probe inserted, as well as the direction of the magnetic field. High voltage pulses were applied between the cathode (target) and the chamber walls using a HIPIMS Sinex 1 (Chemfilt Ionsputtering AB, Sweden) power supply capable of delivering 2400 V and 1200 A peak values with pulse duration lengths of 100  $\mu$ s and a repetition frequency of 50 Hz. In this work pulses up to approximately 700 V and 100 A were used.

For the investigations of the electric fields and oscillations in the plasma, electrical probe arrays of three different geometries were used. These probes are designed for measuring: (1) the oscillating electric field vector in the  $(\rho - \varphi)$  plane (where  $\rho$  is the radial distance parallel to the target racetrack and  $\varphi$  the azimuthal angle) above the racetrack, (2) the azimuthal phase velocity of the oscillations, based on the time shift of the  $E_{\varphi}$  component, and (3) investigating the quasi-DC field component perpendicular to the cathode surface,  $E_z$ . The electric field is obtained from the difference in floating potential between pairs of cylindrical probe tips in the arrays. For accurate measurements, appropriate impedance matching is needed [16] and thus, two different impedance matching circuits, mounted inside the ceramic shaft of the probe, were used in these experiments. For frequencies above 300 kHz a 40:1



**Figure 1.** A schematic cross-section of the lid of the deposition chamber with magnetron and a probe feed through mounted. The  $E_{\varphi}$  probe is positioned above target racetrack, and radially ~0.04 m away from the center of the target. The magnetic field lines measured by Bohlmark *et al.* [9] are indicated as well as a cross section of the torus used for calculation of the discharge current density.

transformer, wound on a ferrite toroid which transforms the 50  $\Omega$  of the oscilloscope to 80 k $\Omega$  was connected to the probes. As seen in Figure 2 (solid curve) the calibration curve is flat between 300 kHz and 20 MHz, with only small phase shifts (not shown here), ensuring accurate measurements in this frequency region. For frequencies from tens of Hz up to a hundred kHz, this transformer has too low sensitivity and also too large phase shifts, so a conventional resistive divider of 10 k $\Omega$ /50  $\Omega$  was used instead. It has lower sensitivity at higher frequencies, but a flat low-frequency response (see the dashed calibration curve in Figure 2). All probe measurements were taken around the current peak of the HIPIMS pulse (40-50 µs after pulse ignition) when the highest cross-**B** currents are drawn and the expected anomalous transport is likely to be strongly developed.

For magnetron plasmas, where  $T_e$  varies over the geometry there will be errors introduced in the electric field measurements due to gradients in the floating potential (see for example [17]). This effect was minimized using two different strategies. For the high frequency wave measurements, **E** fields were only deduced from probe pairs at relative positions such that the difference in  $T_e$  is likely to be small: at the same distance z from the



**Figure 2.** Calibration curves measured for the two different impedance transforming circuits used. For the kHz to MHz region 40:1 transformers (standard probe shaft) were used. When measuring the low frequency oscillations a resistance divider of 10 k $\Omega$ :50 $\Omega$  was used, which has a damping factor of 200. target, and at a location ( $\rho = 0.04$  m, z = 0.04 m) where the **B** field is parallel to the target (*i.e.* perpendicular to the z axis). For the measurements of the  $E_z$  field, the situation is more problematic since there is a strong  $T_e$  gradient along z, probably on the order of 100 eV/m [18]. In this case, the error can be reduced by using cylindrical probes with length  $l \gg r_{ge}$ 

oriented along **B**, where  $r_{ge}$  is the electron gyration radius. This reduces the electron saturation current, but not the ion saturation current. For the optimal geometry, the electron and ion saturation currents are of similar strength (i.e.  $i_{es} \approx i_{is}$ ) for which case the floating potential can be very close to the plasma potential [19]. The probe tips have length  $l \approx 8r_{ge}$  and are oriented along the **B** field. It is uncertain how efficient this suppression of the varying  $T_e$  was, and therefore these probes were only used to obtain an upper limit to  $E_z$ .

The systematic study of high frequency waves also requires a wide set of basic plasma parameters for the particular HIPIMS device used, which are necessary when comparing experimental results with theoretical estimates. This data is presented in Appendix A, and is primarily drawn from a study by Bohlmark *et al.* [9] that utilized the same deposition system and power supply described here. Other basic parameters such as the probe current, target voltage and target current were monitored and recorded on a Tektronix TDS

520C oscilloscope. A Tektronix P6015 high voltage probe measured the target voltage. The current was measured with a Tektronix CT-04 high current transformer together with a Tektronix TCP202 current probe which uses the magnetic field of the wire to determine the current flowing through that wire. In order to perform real-time fast Fourier transforms (FFTs) an HP54645D oscilloscope was used, capable of making time-averaged FFT of the electric field oscillations. By averaging over 256 measurements better statistics were obtained instead of having to rely on single snap shot data.

#### 3 Theory

In order to explain how it is possible to drive electrons across magnetic field lines through plasma oscillations, some of the classical linear theory of instabilities is needed. The twostream instability and the modified two-stream instability (MTSI) are driven by the relative drifts between electrons  $(u_e)$  and ions  $(u_i)$  in the plasma, *i.e.*  $u_{rel} = u_i - u_e$ , in the presence of a magnetic field component perpendicular to this relative drift. This is the case for the circulating Hall current in the magnetic field trap of a magnetron above the target in a HIPIMS discharge [9]. Because the ion gyro radii in magnetrons are typically larger than the spatial dimension of the plasma, only the electrons are magnetized and take part in this azimuthal drift. If the wave electric field has a small but finite component along the magnetic field the electrons will not only Hall drift transverse to the E and B fields, but also be accelerated along the magnetic field, as if they possessed a larger effective mass [20]. Previous investigations of the MTSI have shown that the result will be large oscillations in the electric field, which are often correlated with the plasma density resulting in a net transport of electrons perpendicular to both  $j_H$  and **B**. The ions are too heavy to follow this motion [21]. In the case of the HIPIMS plasma, where the conditions for the MTSI regarding a discharge in the presence of a magnetic field are fulfilled, we propose that a similar transport mechanism could operate and facilitate the discharge current conduction.

Waves (or turbulences) that can give rise to anomalous electron mobility are found in the frequency range for which the ions are unmagnetized and the electrons magnetized,  $f_{gi} \ll f$  $\ll f_{ge}$  [22], where  $f_{gi}$  is the ion gyro frequency and  $f_{ge}$  the electron gyro frequency. For the present HIPIMS device this is approximately the range  $10^4 < f < 10^8$  Hz. Following the work of Ott et al. [20] and later Hurtig et al. [22] on MTSI, the charge perturbation results in growing oscillations frequencies around hybrid with the lower frequency.  $f_{lh} = eB/(2\pi\sqrt{m_em_i})$ , which is about 0.5 – 5 MHz in the case of HIPIMS plasmas. The lower hybrid frequency is the geometric mean between the ion gyro frequency and the electron gyro frequency.

In an earlier experiment on a plasma gun arrangement by Brenning *et al.* [11] anomalous fast cross-field electron transport and magnetic field penetration were quantitatively shown to be closely related and mediated by highly nonlinear waves oscillating in the lower hybrid range. The primary difference between the HIPIMS and the plasma gun experiments are geometrical in nature, with a more complex drift of charged species for the HIPIMS case. The wave structures here are thought to move both in the *z* and  $\rho$  directions, which would lead to oscillations with the wave field  $\mathbf{E}_w$  along more than just the azimuthal direction. This has been verified and is discussed in the next section.

#### 4 Experimental results and discussion

To determine whether or not the MTSI can be responsible for electron cross-field transport, the HIPIMS plasma was observed for oscillations in the lower hybrid frequency range. Since the magnetic field is strongly inhomogeneous above the HIPIMS magnetron [9] there are also variations in the lower hybrid frequency ( $f_{lh} = eB/(2\pi\sqrt{m_em_i})$ ) with position above the magnetron cathode. Right above the racetrack it was found that the calculated  $f_{lh}$  increases from about 0.5 to 5 MHz as z decreases from 0.04 to 0.01 m, assuming a plasma consisting of Ar gas ions.

Figure 3 shows an example of electric field data with high time resolution, taken with the **E** vector ( $\rho$ - $\varphi$ ) probe at current maximum. Electric field oscillations are presented in the radial  $E_{\varphi}$  (Figure 3a) and azimuthal  $E_{\varphi}$  (Figure 3b) directions. In both components, the oscillations in the MHz range dominate the visual impression. With 7 periods during 3 µs, the frequency can be directly estimated to be 2 to 2.5 MHz, *i.e.* within the  $f_{lh}$ -range 0.5 – 5 MHz. It is a feature that is clear on all electric field measurements taken, but which has some shotto-shot variations in frequency by 0.5-1 MHz. A comparison between  $E_{\varphi}$  (Figure 3a) and  $E_{\varphi}$ (Figure 3b) shows that the amplitude of  $E_{\varphi}$  typically is a factor of five higher than  $E_{\rho}$ . Spectra of the whole pulse show the same result. Figure 3c shows an FFT-spectrum of the time series covering the whole discharge pulse from which Figure 3a and Figure 3b are extracted. The lower hybrid oscillations are here seen as a peak at 2.35 MHz. Other spectra (not shown here) show that the  $f_{lh}$ -range-peak is always present on both components, but is a factor of five higher on  $E_{\varphi}$ , meaning it will dominate the anomalous transport process. Thus, this component is therefore chosen for further systematic study in the  $f_{lh}$ -range-peak.

There are also oscillations below the  $f_{lh}$ -range, both on the externally measured discharge curves (Figure 4) and on the internally measured electric fields. The oscillations found on the discharge voltage curve in Figure 4 are in the kHz regime and are not considered here. Figure 5 shows internal oscillations, taken with the phase velocity  $E_{\varphi}$  probe



**Figure 3.** Electric field oscillations derived from the **E**-vector probe data at z = 0.04 m,  $\rho = 0.04$  m, and approximately at the HIPIMS peak current. (a)  $E_{\rho}$  oscillations (b)  $E_{\varphi}$  oscillations, and (c) FFT of the azimuthal oscillations in the MHz range displayed as a power spectrum (*i.e.* V<sup>2</sup><sub>rms</sub> [arb. units]) giving the intensities of the measured frequencies.



Figure 4. Discharge curves from the HIPIMS Sinex 1 power supply.



**Figure 5.** Azimuthal **E** field oscillations measured by two pairs of probe tips (separated 0.008 m) with the phase velocity probe array during the active phase of the pulse approximately 0.04 m above the target racetrack.

array around the current peak of the HIPIMS discharge. The two curves are from differential measurements between probe pairs 1-2 and 3-4, and represent  $E_{\varphi}$  measurements separated 0.008 m in azimuthal direction. The oscillations that are most obvious to the eye at this time resolution are below the  $f_{lh}$ -range (and with no obvious azimuthal phase motion that can be

resolved in this measurement). It is possible that this low frequency range of oscillations contributes to carrying the discharge current. One possible mechanism is through spoke formation, as studied in early rotating plasma devices (see section 6.1.2 in [23] and references therein). This however, may be the subject for future study. Here we concentrated only on the  $f_{th}$ -range oscillations.

In comparing the observed oscillations with the theoretical lower hybrid frequency, it is necessary to estimate the appropriate values to use both for the magnetic field and the ion mass. The uncertainty regarding the magnetic field arises because B has a strong variation with z. Suppose, for example, that the wave structures extend between z = 0.02 m and z =0.04 m. Over this distance the magnetic field strength varies by a factor of 3, and it is not obvious which value to choose. Also, it is likely that the wave structures are formed and move, with the electrons, in their drift away from the cathode. In that case they would generally have their origin in a stronger magnetic field than where they are measured in this study. The uncertainty in ion mass is due to the fact that the HIPIMS generates a highly metallic plasma, in some cases with more than 90% metal ions during the active phase of the discharge [24]. The inert gas ions and target ions mix, thus the effective ion mass depends on an unknown ion mixing ratio. These issues have been resolved in three steps. First, data was collected using Ar chamber gas ( $m_{Ar} = 39.9$  a.u.), which has a mass close to the target material  $(m_{Ti} = 47.9 \text{ a.u.})$ . Here, the mixing ratio should not be critical, and thus this case can be used to determine the B field strength (and thereby the z coordinate) for which the measured oscillations agree with the theoretical  $f_{lh}$ . Second, in order to determine the ion mixing ratio, the variation of the  $f_{lh}$ -range-peak with the chamber filling gas mass from 4 a.u. (He) to 83.8 a.u. (Kr) has been measured and analyzed. Third, an investigation of how the  $f_{lh}$ -range-peak varies with the magnetic field strength by varying the radial probe position has been conducted. The last measurement is intended to reflect the dependence of the  $f_{lh}$ -range-peak on magnetic field strength. The magnetic field strength decreases monotonically with the radius for all values of z.

The time-averaged,  $f_{in}$ -range-peak when Ar is the chamber filling gas, is approximately 3.3 MHz, as displayed in Figure 6 (the data point at 40 a.u.). This is equal to the theoretical value of  $f_{ih}$  for Ar when B = 0.03 T, and corresponding in Figure 8c to  $z \approx 0.01$ m above the target racetrack. This means that the oscillations are consistent with lower hybrid oscillations originating around z = 0.01 m, which is where Bohlmark [9] located the azimuthal current maximum, and where one could expect the instability to be most strongly driven. However, a note of caution regarding this interpretation should be added. The exact value of the frequency that is observed depends on the frame of observation. The measurements were carried out in the lab- (equal to the ion-) rest frame simply because this is the only possible frame in which to make the measurements. An equally important frame to consider would be the azimuthally rotating electron rest frame, which would give another frequency due to the Doppler shift. It is also worth noting that the anomalous transport mechanism works in the same way over a broad frequency range around the lower hybrid frequency, as long as the inequality  $f_{gi} \ll f \ll f_{gg}$  holds. For the efficiency of the transport, the precise identification of the observed frequency peak with  $f_{th}$  at the location z = 0.01 m is therefore not a key issue.

The variation of the measured  $f_{lh}$ -range-peak with chamber gas mass  $m_G$  is presented in Figure 6. It decreases with increasing  $m_G$ , but more slowly than the proportionality  $\propto (m_G)^{-1/2}$  for a pure gas. On the other hand it is important to keep in mind that HIPIMS plasmas are highly metallic; thus chamber gas ions and target ions mix resulting in an effective ion mass. Let us define by p the fraction of target ions with mass  $m_T$ . An effective ion mass can then be calculated as  $\langle m_i \rangle = pm_T + (1-p)m_G$ , and the corresponding lower hybrid frequency becomes

$$f_{lh} = \frac{eB}{2\pi\sqrt{m_e(pm_T + (1-p)m_G)}}.$$
(4.1)

This relation is plotted in Figure 6 for p = 0.7, 0.8 and 0.9, and using the magnetic field B = 0.03 T at z = 0.01 m. The best fit to the experimental results is obtained for p = 0.8, in line with the high ionization for HIPIMS. It must be pointed out that the sputtering yield also

varies depending on chamber gas, which may affect the plasma composition. This has not been taken into account here.



**Figure 6.** The frequency shift in the lower hybrid range as a function of chamber gas mass, as well as theoretical fits of the lower hybrid frequency using effective ion masses with varying metal ion content. The chamber gases used were He (4 a.u.), Ne (20.2 a.u.), Ar (39.9 a.u.), and Kr (83.8 a.u.).

The dependence of the  $f_{lh}$ -range-peak on the radial position, at constant z = 0.04 m, is shown in Figure 7. There is a monotonic decrease by 15 % from  $\rho = 0.04$  m to  $\rho = 0.09$  m. The magnetic data in Figure 7 shows that the relative decrease in the *B* field strength at the probe location is larger. The conclusion is that a magnetic field dependence is there, but it is not fully proportional to the variation of *B* at z = 0.04 m. A tentative explanation is that the structures are formed closer to the racetrack and then move both in the *z* and in the radial direction to the location where they are meaured, and thus represent magnetic field strengths at smaller radii  $\rho$  and lower *z*.

In summary, the conclusions that can be drawn regarding the waves in the  $f_{lh}$ -range at  $(\rho = 0.04 \text{ m}, z = 0.04 \text{ m})$  are as follows: (1) There is always a significant peak in the lower hybrid range, with an azimuthal  $E_{\varphi}$  field amplitude of typically 50 V/m, which is a factor of 5 stronger than the radial component  $E_{\rho}$ . (2) The measured frequency does not correspond to lower hybrid at the position z = 0.04 m where the measurement is made, but closer to the target, at  $z \approx 0.01 \text{ m}$ . (3) The variation with ion mass is consistent with lower hybrid

oscillations and an assumed ion mixing ratio of 20 % gas ions (He, Ne, Ar, or Kr), and 80 % target ions (always Ti). (4) The variation with magnetic field strength (obtained by measuring at varying radial positions) is consistent with lower hybrid oscillations assuming that they originate closer to the target in the radial direction than where they are measured.



**Figure 7.** Study of peak shift in the lower hybrid range as a function of radial distance from the target at  $z \sim 0.04$  m. The absolute value of the magnetic field strength z = 0.04 m has also been plotted to better illustrate the scaling of this frequency shift. The values of the magnetic field are taken from Bohlmark *et al.* [9].

The oscillations detected are in the lower hybrid range, which is known to be associated with anomalous mobility in plasmas driven by the modified two-stream instability. In order to excite the MTSI in the HIPIMS plasma it is necessary to have a sufficiently fast relative drift between electrons and ions. If the instability is driven by the azimuthal current (in analogy to Hurtig *et al.* [22]) then the total azimuthal electron drift should be higher than the threshold velocity for the MTSI instability,  $u_{e,\varphi} > u_{MTSI}$ , for which below there is strong Landau damping. As MTSI threshold the ion average speed is taken (which in this experiment is higher than the ion acoustic speed), using the ion velocity derived from time-resolved ion energy measurements made by Bohlmark *et al.* [5] on the same HIPIMS system. These measurements were

performed using an energy resolving quadrupole mass spectrometer mounted on the side wall of the chamber and thereby scanning the ions moving in the azimuthal direction (since the magnetron is top mounted). From these measurements we find an average ion energy of 40 eV in the case of Ti<sup>+</sup>. This will be our upper threshold, since the average ion energy of Ar<sup>+</sup> is considerably lower and therefore resulting in a much lower threshold value. Calculating the ion velocity using  $\sqrt{2W_i/m_i}$  gives a speed of  $1.3 \times 10^4$  m s<sup>-1</sup>. This is clearly below the azimuthal velocity,  $u_{e,\varphi} \approx 5 \times 10^4$  m s<sup>-1</sup>, obtained in Appendix A. Thus, the threshold for the MTSI is exceeded, further strengthening the case for the MTSI in HIPIMS plasmas.

#### 5 Conclusion

It has been shown that there are high frequency **E** field oscillations within the HIPIMS plasma, and proposed that these oscillations may play a key role for the high currents in HIPIMS sputtering. The anomalous fast electron transport previously observed in HIPIMS discharges can be explained by fluctuations of the electric field in the azimuthal direction. This results in a drift of electrons across the magnetic equipotential lines directed momentarily by the direction of the oscillating electric field. One of the driving mechanisms behind this drift is suggested to be the modified two-stream instability (MTSI), which would generate electric field oscillations in the lower hybrid regime. Experimental investigations of HIPIMS plasmas yielded resonant frequencies in the MHz range corresponding to the lower hybrid frequency. Further investigations of these frequencies confirmed trends predicted by the theory of lower hybrid waves. These frequencies are suggested to be a mechanism for the high currents developed in HIPIMS plasmas.

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### **Appendix A**

In this section the current density discharge ratio,  $j_H/j_D$ , is estimated based on the electron drifts found in HIPIMS plasmas. The discharge current - voltage characteristics,  $I_D(t) - U_D(t)$ , of single HIPIMS pulses were analyzed by triggering on the voltage rise flank. A typical discharge pulse is shown in Figure 4.  $U_D$  shows a strong increase at the beginning of the pulse, and dropping as the plasma is ignited. The work focused on the time around the current peak of the HIPIMS pulse (40-50 µs after pulse ignition) where the highest cross-B currents are drawn and the anomalous transport is expected to be the most strongly developed. At this time, a high density doughnut shaped plasma has formed above the target racetrack. At z =0.04 m the electron density is about  $1 \times 10^{18} \text{ m}^{-3}$  as illustrated in Figure 8a. Around the time of maximum current, a dense plasma expands away from the target, indicated by the decreasing electron pressure in Figure 8b. The basic plasma parameters estimated here are scaled from two studies by Bohlmark et al. [9, 25] using the same deposition system and power supply described herein. This scaling is based on the fact that the shapes, as functions of time, of Bohlmark's  $U_D(t)$  and  $I_D(t)$  curves are very similar to what is reported here in Figure 4, particularly around the current maximum. The main difference is that Bohlmark's ID maximum was a factor 4.5 higher than the present measurements;  $u_D$  at the same time was only a factor 1.1 higher. Scaling of the data from their study is therefore made such that the current densities, the plasma density, and the plasma (electron) pressure are all reduced by a factor 4.5, while the electron temperature and the particles' drift speeds are taken to be the same.

For the discharge current density,  $j_D$ , and the corresponding drift,  $u_{eD}$ , a geometrical model has been used. The electron discharge current  $I_{D,max} = 100$  A is taken to flow across a torus-shaped surface, with a half-circle shape cross section. The half-circle centre is placed at the target surface at  $\rho = 0.04$  m with a circle radius of 0.04 m (see





**Figure 8.** Plasma parametric variation above target racetrack during the active phase of the pulse. The parameters studied are: (a) Plasma density,  $n_e$ ; (b) electron pressure,  $P_e$ .  $P_e$  values for the current peak and ±15 µs from the peak have been plotted; (c) absolute values of the magnetic field strength, *B*, and magnetic field curvature radius,  $R_c$ , as functions of distance from the target surface (z) at  $\rho \sim 0.04$ -0.05 m, and (d) absolute values of the magnetic field strength, *B*, as a function of  $\rho$  taken at the standard z position, z = 0.04 m. The data presented are taken from Bohlmark *et al.* [9, 25].

Figure 1). The area of this torus is about 0.03 m<sup>2</sup>, giving an average current density of  $j_D = 3 \times 10^3$  A m<sup>-2</sup>, and an average drift speed  $j_D(n_e e)^{-1}$  of  $u_{eD} = 2 \times 10^4$  m s<sup>-1</sup>, when putting in the estimated plasma parameters for this position.

Three mechanisms can give rise to an azimuthal electron velocity,  $u_{e,\varphi}$ , in sputtering magnetrons: (1) "curved magnetic vacuum field" gyro centre drift  $u_{e,R}$ , (2) pressure gradient drift  $u_{e,\nabla p}$ , and (3) Hall drift  $u_{e,E/B}$ . Below it is estimated to what degree each of these three mechanisms contributes to the total azimuthal current. The position z = 0.04 m above the racetrack is here considered, where the **B** field is perpendicular to the *z* axis (parallel to the target). The gyro center drift can, for an isotropic Maxwellian distribution, be written as [26, page 30]:

$$\mathbf{u}_{R} = \frac{2k_{B}T_{e}}{e} \frac{\mathbf{R}_{c} \times \mathbf{B}}{R_{c}^{2}B^{2}} = -2 \times 1 \frac{1}{0.027 \times 0.005} \hat{\varphi} \approx -1.5 \times 10^{4} \,\mathrm{m \, s^{-1}},\tag{A.1}$$

where the numerical values correspond to ( $\rho = 0.04$  m, z = 0.04 m), with  $R_c$  and B and taken from Figure 8c, and the electron temperature is approximated to be 1 eV based on Bohlmark's data [9].

The pressure gradient (or diamagnetic) drift [26, page 69] is:

$$\mathbf{u}_{P} = \frac{\nabla p \times \mathbf{B}}{en_{e}B^{2}} = -\frac{15}{1.602 \times 10^{-19} \times 1 \times 10^{18} \times 0.005} \hat{\varphi} \approx -1.9 \times 10^{4} \text{ m s}^{-1}.$$
 (A.2)

The density  $n_e$  is here taken from Figure 8a, and the pressure gradient  $\nabla p_e \approx -15$  J m<sup>-3</sup> is approximated from the electron pressure difference between z = 0.04 m and z = 0.06 m in Figure 8b, for the curve corresponding to the time of current maximum.

The Hall drift finally is somewhat uncertain since the  $E_z$  measurement performed might be corrupted by the strong spatial variation in electron temperature, as discussed in section 2. However, it is known that the temperature gradient  $\nabla T_e$  is directed towards the cathode, and therefore would give a "false  $E_z$  field" that would add, with the same sign, to a discharge  $E_z$  field in the  $-\mathbf{z}$  direction. An upper limit to the real  $E_z$  strength can therefore be calculated, corresponding to an assumed zero sensitivity to the temperature gradient. Due to the negative sign of  $E_z$ , the inequality takes the form  $E_z >$ -64 V m<sup>-1</sup>, and the corresponding limit for the azimuthal Hall drift [26, page 23] becomes:

$$\mathbf{u}_{e,E/B} = \frac{\mathbf{E} \times \mathbf{B}}{B^2} = \frac{E_z}{B_{\rho}} \varphi > -\frac{64}{0.005} \hat{\varphi} \approx -1.3 \times 10^4 \text{ m s}^{-1}.$$
(A.3)

Thus, all three contributions to  $u_{e\varphi}$  are in the same direction, and are of significant amplitude. The agreement in sign between the various drift terms is consistent with the fact that all three forces act on the electrons in the same direction, that is away from the cathode. These electron drifts add up to the total azimuthal electron drift, which is on the order of  $u_{e,\varphi} \approx$  $5 \times 10^4 \text{ m s}^{-1}$ .

In magnetron plasmas the ions are almost unmagnetized, and the azimuthal current is carried mainly by electrons. The sum of these three drifts therefore gives the azimuthal current density  $j_{e,\varphi} = j_{e,R} + j_{e,\nabla p} + j_{e,E/B}$ . (The notation  $j_{e,\varphi}$  is here preferred instead of  $j_H$ that is more common in magnetron literature, and only describes the Hall current density.) Finally the azimuthal current density is obtained as  $j_{e,\varphi} \approx 7 \times 10^3$  A m<sup>-2</sup> and the current density ratio is found to be  $j_{e,\varphi}/j_D \approx 7 \times 10^3 / 3 \times 10^3 \approx 2$ .

The conclusions that can be drawn regarding the drift speeds and current densities at  $(\rho = 0.04 \text{ m}, z = 0.04 \text{ m})$  are: (1) The current ratio is consistent with Bohlmark's result at higher power,  $j_{H}/j_D \approx 2$ . (2) While Bohlmark concluded that the Hall drift probably is the major contribution to the azimuthal electron drift at z = 0.01 m during the discharge, it might be a minor contribution at z = 0.04 m even at current maximum. (3) The drift speeds are higher than those obtained by Bohlmark at z = 0.01 m, probably because the plasma density decreases faster with z than the discharge current density. At z = 0.04 m the resulting velocities are  $u_{e,\varphi} \approx 5 \times 10^4$  m s<sup>-1</sup> and  $u_{eD} = 2 \times 10^4$  m s<sup>-1</sup>. However, in view of the uncertainties in the estimates, these values might well be wrong by a factor of perhaps two or three.

The ratio  $j_{e,\phi}/j_D$  is an extremely important parameter since it reflects the degree of anomalous transport of electrons across the **B** field. For the case when the electrons carry the current, and collisions dominate the electric conductivity (as in the bulk plasma of magnetrons), the ratio is [27]:

$$j_H / j_D = \omega \tau , \qquad (A.4)$$

where  $\omega$  is the electrons' (angular) gyro frequency ( $\omega = eB/m_e$ ) and  $\tau$  is the electron momentum-exchange collision time. In the case where the current is driven by an electric field, this follows directly from the ratio between the Hall and Pedersen conductivities [28],  $\sigma_H$  and  $\sigma_P$  respectively (note that it does not require  $\omega_{ge}\tau_c >> 1$  as stated in [27]):

$$\frac{j_H}{j_D} = \frac{E\sigma_H}{E\sigma_P} = \frac{E\frac{en_e}{B}\frac{\omega_{ge}^2\tau_c^2}{1+\omega_{ge}^2\tau_c^2}}{E\frac{en_e}{B}\frac{\omega_{ge}\tau_c}{1+\omega_{ge}^2\tau_c^2}} = \omega_{ge}\tau_c.$$
(A.5)

This classical collision rate was in the HIPIMS case estimated by Bohlmark [9] to give  $\omega \tau \approx 35$  close to the target and  $\tau$  dominated by electron-neutral collisions. Where we measure the waves, at  $\rho = 0.04$  m, z = 0.04 m, the electron temperature is lower with the result that coulomb collisions dominate and give a somewhat lower value  $\omega \tau \approx 15$ . Still the conclusion is that, to explain the high discharge current density, some mechanism is needed that can give an effective collision rate such that  $(\omega \tau)_{eff} \approx 2$ , almost an order of magnitude lower than the classically predicted value. Recent studies of the current-density distribution in a pulsed DC magnetron discharge have given similar values [29]. This anomalous collision rate is also an order of magnitude lower than that given by Bohm diffusion, which has been determined experimentally to give an effective diffusion coefficient of  $D_B = 1/16[k_BT_e/(eB)]$  [26, page 190] (which is equivalent to  $(\omega \tau)_{eff} \approx 16$ ) in a wide parameter range. It is therefore very probable that for strongly driven pulsed discharges, such as the HIPIMS discharge, instabilities can rise to higher amplitudes, resulting in faster-than-Bohm anomalous transport. It is also likely that one and the same discharge can have different transport types in different parts of the plasma, and also varying in time during the discharge pulse.

The value  $\omega \tau \approx 2$  obtained in the HIPIMS discharge also strengthens the analogy to the plasma gun experiment on anomalous electron transport by Hurtig *et al.* [11], where they measured the anomalous transverse resistivity  $\eta_{\perp}$ . This can be used to obtain an approximate value  $2 < (\omega \tau)_{eff} < 4$ , that is consistent the magnetron data presented here, and their conclusion is, like ours, that an anomalous transport in excess of Bohm diffusion is needed [30].

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