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Propagation direction reversal of ionization zones in the transition between high and low current magnetron sputtering

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Abstract

Past research has revealed the propagation of dense, asymmetric ionization zones in both high and low current magnetron discharges. Here we report about the direction reversal of ionization zone propagation as observed with fast cameras. At high currents, zones move in the $\mathbf{E} \times \mathbf{B}$ direction with velocities of 10^3 to 10^4 m/s. However at lower currents, ionization zones are observed to move in the opposite, the $-\mathbf{E} \times \mathbf{B}$ direction, with velocities $\sim 10^3$ m/s. It is proposed that the direction reversal is associated with the local balance of ionization and supply of neutrals in the ionization zone.

Previous research on high current magnetron sputtering (MS) discharges, especially high power impulse magnetron sputtering (HiPIMS), has revealed a plasma that is rich in structure, featuring self-organized patterns^{1,2,3,4}, plasma flares^{3,5}, and azimuthally asymmetric particle jets^{6,7}. The high discharge currents suggest that waves and instabilities play a critical role in the formation of these phenomena^{8,9}. The most prominent features are drifting regions of enhanced excitation and ionization. Fast imaging of HiPIMS plasma has shown evidence of the presence of dense, asymmetrically shaped ionization zones that are more-or-less regularly spaced in the azimuthal direction. These zones have been observed to propagate between 10^3 m/s and 10^4 m/s, which is much slower than the $\mathbf{E} \times \mathbf{B}$ drift velocity¹⁰ of electrons, $\sim 10^5$ m/s, ref. 1,11. In contrast to high current MS, ultrahigh speed imaging of low current DC magnetized microplasmas¹² has revealed plasma waves propagating in the $-\mathbf{E} \times \mathbf{B}$ direction with phase velocities of approximately 10^3 m/s. Measurements of floating potential fluctuations in DC planar magnetron sputtering plasmas using a circular array of electric probes have shown the presence of several Fourier modes with azimuthal periodicity at 100 kHz, interpreted as drift waves propagating azimuthally¹³.

In this contribution, the transition regime between low and high current magnetron sputtering has been studied with fast imaging. A comprehensive set of streak and intensified charge-coupled device (ICCD) camera images has been acquired for magnetron plasmas operating at various currents and pressures. In the experiments, the discharge was powered by a high current pulse generator, model SPIK2000A (Melec GmbH), capable of delivering pulses up to 1 kV and up to 500 A. Typical pulse lengths were 20-200 μ s with repetition rates between 100 and 200 Hz, although we also investigated continuously operating (direct current or dc) discharges. An unbalanced planar magnetron (MeiVac Inc.) with a 76 mm (3 in.) diameter and 6.25 mm (1/4 in.) thick gold target was operated in argon. Gold was selected to exclude effects associated with target poisoning. The cylindrical anode was mounted flush with the target. The discharge current was recorded using a current transformer (Pearson model 101), and the discharge voltage was measured with a voltage probe (Tektronix P5100). All electrical signals were recorded with a National Instruments PXI-5105 oscilloscope. ICCD images were taken with a Princeton

Instruments PIMAX 4, equipped with an $f=80$ mm Nikon lens. The spectral response of the PIMAX is from 200 to 900 nm. Streak images were taken with a Hamamatsu C7700, equipped with an $f=35$ mm Nikon lens and detector module C4742-98 whose spectral response is in the range 300-1060 nm. All images are presented using the color scale “royal” of the image processing software IMAGEJ¹⁴.

Streak and ICCD images of pulsed magnetron discharges were systematically taken for pressures between 0.1 and 2.7 Pa and discharge voltages between 300 and 710 V. The streak camera’s horizontal slit was tangentially aligned to the sputtering target’s erosion racetrack as shown in Fig. 1 (c) to capture the movement of ionization zones in the azimuthal direction. In Figs. 1 to 3, the left sub-figure shows the streak camera image along with the discharge current-time profile, while the right sub-figures show ICCD images taken at various times into discharge pulses. Due to the limitations of the cameras, the sub-figures were not taken from the same pulse. However, due to the similarity between current profiles and images from one pulse to the next, these images can be associated with each other in this way. We imaged pulsed discharges with short and long pulse widths and with dc operation. The results showed that the longer pulses are like extensions of the shorter pulses when the transient switch-on is completed. Therefore, in terms of zone propagation direction, it does not really matter if the discharge is pulsed or continuous.

The velocity and direction of propagation of ionization zones can be deduced from the streak images by relating the spatial displacement of light emission (horizontal axis) to the elapsed time (vertical axis). For this experimental setup, bright streaks travelling from the upper right to the bottom left indicate motion in the $\mathbf{E} \times \mathbf{B}$ direction, and vice versa, streaks from the upper left to the bottom right correspond to the $-\mathbf{E} \times \mathbf{B}$ direction. Figs. 1 and 2 show images from discharges with similar current-time profiles, but with pressures of 2.7 Pa in Fig. 1 and 0.8 Pa in Fig. 2. Early on in these pulses, when discharge currents are higher than approximately 6 A, zones move in the $\mathbf{E} \times \mathbf{B}$ direction at velocities of approximately 5×10^3 m/s. However, later in the pulses, when discharge currents have fallen below approximately 6 A, the bright streaks start to become indistinguishable and their tilt undefined. As current fluctuates around 6 A or somewhat lower, the zone propagation changes between the $-\mathbf{E} \times \mathbf{B}$ and $\mathbf{E} \times \mathbf{B}$ directions many times, which suggests that there is a current regime in which the zones are quasi-stationary. At the very end of the pulses, when the discharge currents have fallen significantly, the streak tilts clearly indicate a reversal of the zones’ direction of travel into the $-\mathbf{E} \times \mathbf{B}$ direction.

Pressure and discharge current [together](#) determine the number of the ionization zones. Consider for example the frame images taken at approximately the same current, at 80 μ s in Fig. 1 (d) and at 60 μ s in Fig. 2 (d). We find that the higher pressure results in a greater number of zones. In the extreme case shown in the frame image at 90 μ s in Fig. 3 (c), the zones merge and smooth out into a uniform ring. In Figs. 1 and 2, as the pulse evolves and the discharge current diminishes, the inter-zone spacing in the streak images increases significantly, consistent with the corresponding frame images in which the number of zones decreases.

In the many streak images taken with a gold target, direction reversal was observed between 0.5 and 6 A. When the argon gas pressure was increased, the discharge current density needed for direction reversal decreases, and the range of discharge current density for direction reversal narrows. For example, at 0.1, 0.8 and 2.7 Pa, the discharge current density range for reversal is approximately 0.62 to 7.40, 0.21 to 2.7 and 0.08 to 1.48 A/cm², respectively. We use here current density as this is likely to most relevant physical quantity. Since the discharge current is localized in ionization zones, current densities were determined by dividing the discharge current by the observed dense plasma areas as seen in the images. While this bears some uncertainty due to way of determining the “area” of ionization zones, the so-determined current density has certainly greater physical meaning than the commonly cited (nominal) current density where the entire target area is applied because most of the target area is actually not facing dense plasma.

In Fig. 2 (a), the 20 μ s long streak image clearly shows ionization zones “hovering”, i.e., staying relatively stationary over the racetrack. To further illustrate the very slow motion of ionization zones, Fig. 4 (b) shows the corresponding ICCD image with a long, 30 μ s exposure time: we see slightly blurred but still distinct zones. Images of zones taken with even longer exposure times of 100 μ s, Fig. 4 (c), are still not showing plasma rings that one would expect from blurring due to motion. In contrast, images of high current magnetron sputtering with 25 μ s exposure time, like the HiPIMS images in Fig. 3 of Ref. 1, show azimuthally uniform plasma rings due to blurring of fast moving ionization zones. We conclude that the ionization zones move quite slowly, if at all, in the transition regime, i.e. slower than at high current and slower than at low current.

Images of ionization zones can and should be interpreted as distributions of electric potential and related electron energy¹⁵, and there is evidence that an ionization zone is a location of an asymmetric potential hump of several 10 V, see e.g. ref.16. Therefore, when drifting electrons stream through an ionization zone, they have a tendency to gain energy from the region’s potential hump, enabling them to cause enhanced excitation and ionization of the ions and neutrals present¹⁷. The region of greatest ionization is clearly indicated by the region of most intense light emission, indicated in Fig. 2 (d), as atom and ion excitation and ionization are mostly caused by energetic electrons. To interpret the observation of zone drift reversal, we consider the interaction of electrons with all particles and take into account both electron energies and the densities of these particles.

When the discharge current is high, namely higher than approximately 6 A for our magnetron with a gold target, a large fraction of neutral atoms within the region of greatest ionization are ionized. Therefore, the neutral density is reduced, followed by a reduction of the ion densities as ions move away following the local electric field. When new drifting electrons arrive what used to be the region of greatest ionization, they encounter a depletion of neutrals and hence reduced interaction, and they will drift a bit further in the $\mathbf{E} \times \mathbf{B}$ direction before encountering a neutral density sufficient for high excitation and ionization rates. Therefore, at high currents, the location of the ionization zone is observed to shift or propagate in the $\mathbf{E} \times \mathbf{B}$ direction.

When the current is low, namely lower than approximately 0.5 A for our magnetron with a gold target, we have a relatively low ionization rate. Even at locations of enhanced ion density, neutrals are abundant at all times: they are never depleted by ionization processes. Electrons arriving at the zone of higher potential approach this zone from the $-\mathbf{E} \times \mathbf{B}$ direction and thus are more likely to deposit their energy on the $-\mathbf{E} \times \mathbf{B}$ side of that zone, making the zone to shift or propagate in the $-\mathbf{E} \times \mathbf{B}$ direction.

In the intermediate transition regime, namely at currents between approximately 0.5 A to 6 A for our magnetron with a gold target, there seems to be a balance of these two counteracting phenomena. The ionization rate and arrival rate of neutrals are approximately balanced, making the location of highest electron energy deposition approximately quasi-stationary.

In summary, the exploration of the transition regime between MS operation at low currents and power densities and high currents and power densities has revealed an interesting reversal in the direction of ionization zone propagation. We find that the discharge current (or power) density plays a vital role in determining the ionization zones' direction of propagation. This suggests that the direction reversal is associated with localized heating of electrons, local neutral ionization rate and local electron energy deposition in relation to a supply of neutrals, which is pressure dependent.

Our findings of reversal in the direction of ionization zone propagation are not limited to a Au target, as they are also observed with many other target materials such as Al, Ti, Cu, Nb and W. With respect to industrial applications, one could expect that plasma phenomena such as these may have an influence on the quality of the deposited thin films. Research with other target materials and effects on film microstructure is ongoing and results will be published elsewhere.

Figure Captions

FIG. 1: Images of the zone's propagation direction change in 2.7 Pa of argon and discharge voltage of 650 V. (a) and (b) were taken with a streak camera with a sweep time of 50 μ s. (c) to (e) were taken at different times within the pulse by an ICCD camera with exposure time 20 ns. Each image was recorded using light emitted from a different pulse. The faint circle visible in the center of each frame image is a reflection from a hollow gold cylinder covering a nut used for target clamping.

Fig. 2: Images of the direction change of ionization zone propagation at argon pressure of 0.8 Pa and discharge voltages of 700 V. (a) and (b) are streak images with a sweep time of 50 μ s. (c) to (e) are frame images with an exposure time of 20 ns.

Fig. 3: Images of the zone's propagation direction change in 2.7 Pa of argon and discharge voltage of 350 V. (a) contains a stitch of two streak images, each with a sweep time of 50 μ s. (b) to (d) are frame images with 20 ns exposure times.

Fig. 4: Images taken in 0.1 Pa argon, discharge current and voltage are 5 A and 700 V. The streak image (a) was taken 60 μ s after the beginning of the pulse when the discharge current has reached a relatively stable value. Frame images were also taken at approximately 60 μ s into the pulse, with long exposure times (b): 30 μ s, (c): 100 μ s.

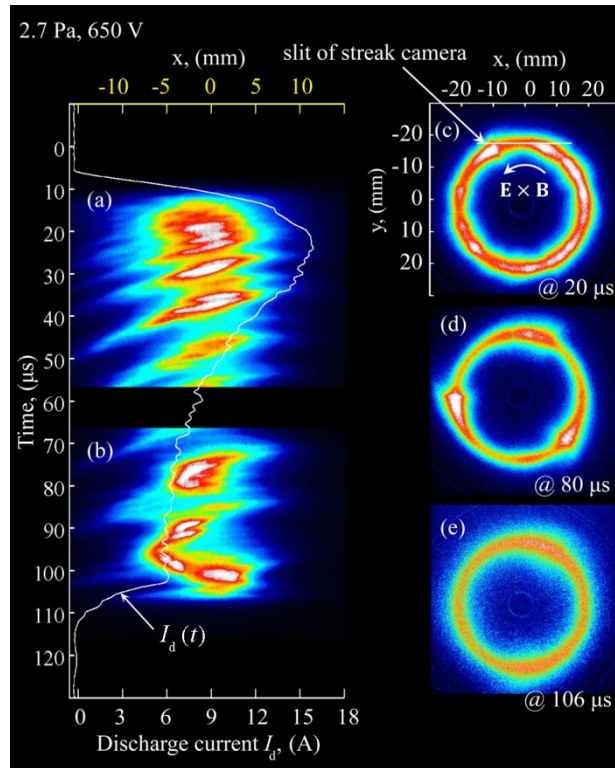


Fig. 1

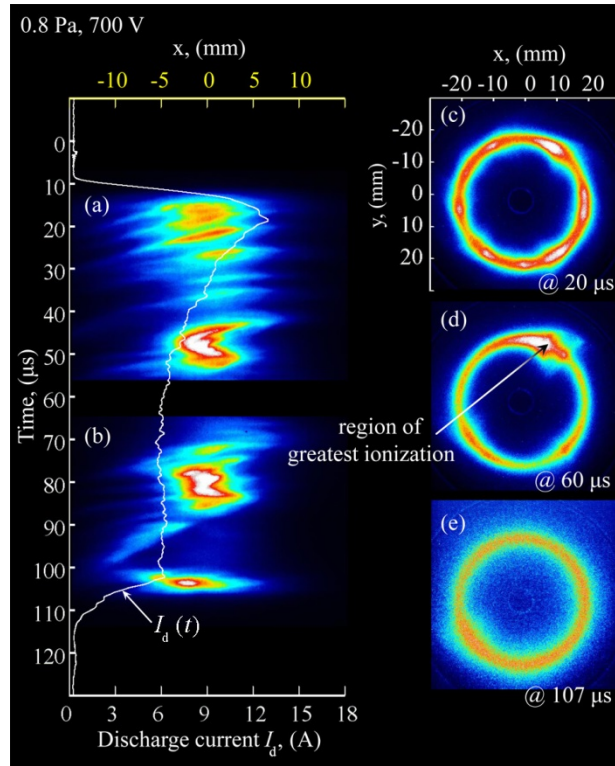


Fig. 2

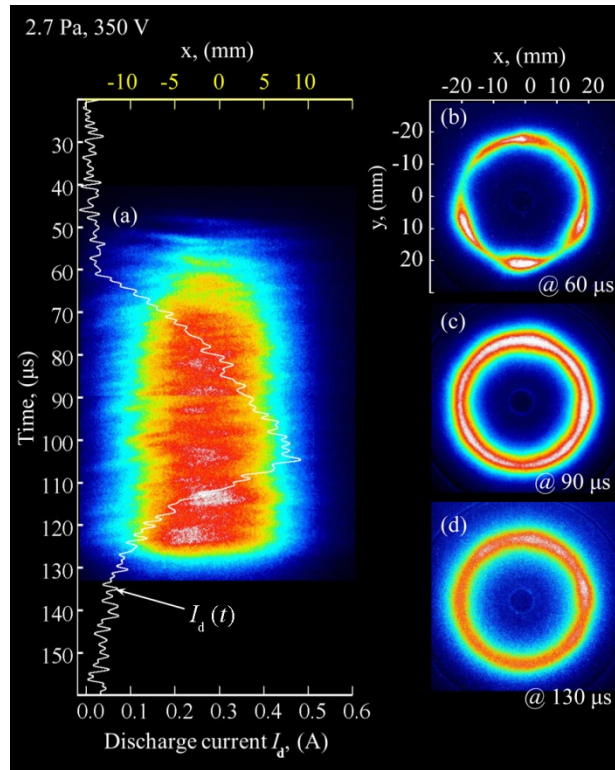


Fig. 3

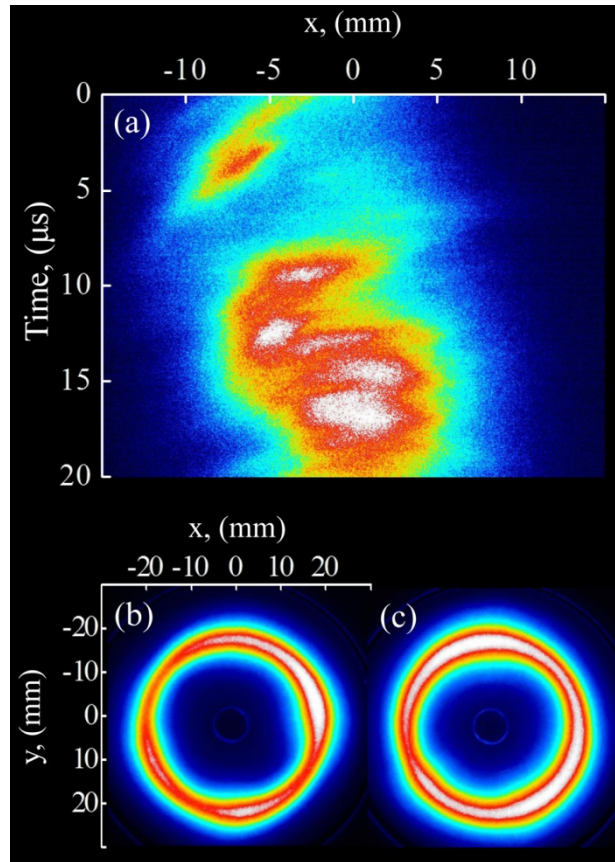


Fig. 4

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