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Afterglow dynamics of plasma potential in bipolar HiPIMS discharges

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Abstract

In bipolar magnetron sputtering, the plasma afterglow is initiated by switching the target bias from a negative to positive voltage. In the following, the plasma potential evolution in this configuration is characterized, being responsible for the ion acceleration at the substrate sheath potential fall, in particular in high power impulse magnetron sputtering (HiPIMS). A mass-energy analyzer and a Langmuir probe respectively measure the ion energies and the plasma/floating potential at different positions within HiPIMS discharges. A plasma potential drop and rise in the first 45 μs of the afterglow is observed, settling in the plasma bulk towards values below the applied positive bias. The measured ion energies agree with the plasma potential values before and after the drop-rise. To gain more comprehensive insights into the mechanisms responsible for such a potential evolution, particle-in-cell Monte Carlo 3D simulations of bipolar direct current magnetron sputtering discharges are explored in equivalent geometries. Despite their average power being orders of magnitude lower compared to the HiPIMS configuration, a similar afterglow behavior is observed. This indicates that the measured dynamics are not specific to HiPIMS, but rather a feature of bipolar magnetron sputtering. The responsible mechanisms are studied further: the effects of various system parameters are decoupled, with the magnetic field configuration emerging as crucial for the plasma potential drop-rise dynamics and the associated re-ionization close to the target.

Keywords: HiPIMS, positive pulse, afterglow, plasma, bipolar, magnetron sputtering

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

1. Introduction

Among the techniques currently implemented for the deposition of thin films, magnetron sputtering is one of the most commonly used [1]. Direct current magnetron sputtering (DCMS) has been exploited for several decades [2], up to the recent development of high power impulse magnetron sputtering (HiPIMS) [3]. In HiPIMS, the voltage at the target is modulated in negative pulses from a few to hundreds of μs with a duty cycle generally below 10%, but applying the same aver-

age power density than DCMS ($\sim 10 \text{ W cm}^{-2}$). This leads to higher peak power densities ($\sim 1 \text{ kW cm}^{-2}$) and plasma densities up to three orders of magnitude above those of DCMS, with the consequent ionization of the sputtered target atoms [4]. These ions can be accelerated onto the growing film either by biasing the substrate surface with a negative voltage [5–8], or by applying a positive pulse (PP) at the target during the afterglow between consecutive discharge pulses [9–11]. In comparison with DCMS, the higher flux of energetic metal particles onto the growing film allows for denser coatings, particularly in geometries featuring grazing angles of incidence [12–14]. During the afterglow in the PP configuration, the plasma potential V_{pl} rises to values comparable to the applied PP because of the higher electron mobility with respect to that of ions [15]. The V_{pl} evolution is crucial for determining at which energies the metal ions impinge on the substrate, as they

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are predominantly accelerated through the potential drop at the substrate sheath. Recent investigations with planar magnetrons have shown ion energies that do not match the positive voltage applied at the target [9], or even present double-peaked distributions [10, 16]. This requires a closer look at the V_{pl} dynamics during the afterglow. First proposed explanations suggest the establishment in a few μs of a V_{pl} spatial profile with at least two plateaus along the axial direction in the two regions inside and outside the magnetic trap [17, 18]. Recent results in HiPIMS show a temporal evolution of V_{pl} during the afterglow that is characterized by a drop and rise (D & R) occurring within 30 μs after the PP triggering, from a value close to the applied positive target voltage to a dip below half of that value, and to a final rise to a few tens of volts below the target voltage [19]. These two V_{pl} values before the drop and after the rise match the ion energies measured with mass and energy analyzers. A reverse discharge is proposed as the mechanism behind this dynamic, but a clear picture of the afterglow evolution in the presence of a PP is still missing, in particular with respect to the conditions that determine the observed D & R.

In this work, we further the investigation of the plasma evolution during the afterglow of bipolar magnetron sputtering by pursuing two parallel approaches: (1) experimental measurements in HiPIMS with PP by means of a mass-energy analyzer (MEA) and a Langmuir probe (LP). (2) Simulations of a corresponding bipolar DCMS process with a particle-in-cell Monte Carlo (PICMC) 3D code [21] at lower discharge power. The experimental measurements confirm the observations presented in the previously discussed works [19], and provide a more refined radial and axial profile of V_{pl} , as well as of the ion energies. The simulated V_{pl} during the afterglow shows nonetheless a D & R as for the measurements, indicating that this feature is peculiar to bipolar magnetron sputtering in general rather than specific to HiPIMS. A deeper global understanding of the observed dynamics is obtained from the spatial and temporal evolution of other plasma parameters, including electron density and ionization collisions per unit of time and volume. Re-ionization close to the target is confirmed to occur because of the magnetic field geometry and consequent lensing of the electrons towards the target, similarly to the secondary discharge proposed in [19]. Also, physical parameters that can play a role during the afterglow are decoupled by including/removing them from the simulations to verify their effect. Ion-induced secondary electron emission (IISEE) from the grounded surfaces provides a source for electron collisions with the neutral gas, while the magnetic field plays the most crucial role in the observed D & R of the V_{pl} . Therefore, different magnetic field configurations are explored to confirm their impact on the afterglow plasma.

The manuscript is organized as follows. In section 2, the experimental setup of the measurements in HiPIMS is introduced, with an overview of the implemented numerical code. Experimental measurements are detailed in section 3, and the results of PICMC simulations are presented in section 4. Conclusions are given in section 5.

Figure 1. Magnetic configurations of the *CERN-balanced* (a) and *Gencoa-balanced* (b) planar magnetrons, as obtained with the FEMM software [20]. Half their cross-sections are detailed around their axis of symmetry (dashed red). Along these axes, the distance of the magnetic field null point ($B_z = 0$) from the target surface is indicated. All dimensions are given in mm.

2. Experimental and numerical tools

2.1. Experimental setups

An external DC-power supply (ADL, GS 20) is used to measure and control the operating average power that is fed to a dedicated 2 kW HiPIMS module (Starfire, SF-IMPULSE2KX-SH). The Starfire module allows to modulate the target voltage by inverting its polarity to positive values within 4 μs , while independently controlling the PP duration, delay, and amplitude. Target voltage and current are measured, respectively, with a high-voltage probe (4 kV, 400 MHz bandwidth) and a current probe (30 A, 100 MHz bandwidth).

Two different setups have been used in this work to investigate HiPIMS with Ar as the process gas at 8×10^{-3} mbar. Both systems include planar magnetrons featuring a 50 mm diameter Nb target, and an MEA-Pfeiffer plasma process monitor 422 that provides time-integrated ion energy distribution functions (IEDFs).

The first setup, from now on defined as *CERN-balanced* and described in [9], features a balanced planar magnetron with the custom design detailed in figure 1(a). It allows to modify the permanent magnet arrangement to experimentally explore alternative configurations, such as the unbalanced and strongly unbalanced assemblies that are discussed in section 4.2. The MEA aperture is placed on the target axis at an angle of 15° and at 145 mm from the target surface.

The second setup includes a commercial balanced planar magnetron, model Gencoa 3G-A, as described in figure 1(b) and defined as *Gencoa-balanced* in this manuscript. The *Gencoa-balanced* magnetron faces the MEA, which can be displaced along the magnetron central axis to spatially resolve

the IEDFs. A cylindrical LP with a 3 mm long tungsten tip of 1 mm diameter is aligned perpendicularly to the magnetron axis to perform radial or axial measurements. The fast dynamics of the HiPIMS discharge necessitate a temporal resolution on the μs scale for the LP measurements. The concurrent acquisition of time-resolved current–voltage (I – V) characteristics would thus require a voltage sweep between the probe’s electron and ion saturation regimes over even shorter times. The LP current is therefore instead measured for a fixed applied voltage, over 500 cycles of the discharge and afterglow. By repeating these measurements for a series of different probe voltages in sequence, the I – V characteristics for each time-step are reconstructed during post-processing, with a temporal resolution as high as that of the digital acquisition system. The plasma potential V_{pl} at which the second derivative of the curve changes sign (*knee* of the curve) can be qualitatively identified in the I – V curves, as described in section 3, despite the limited number of data points. Complementary floating potential V_{fl} measurements of the LP are performed to confirm the afterglow dynamics by directly connecting the LP tip to the digital oscilloscope, bypassing the LP amplification module used for the I – V curve measurements.

2.2. Numerical code

In this work, a parallelized 3D PICMC simulation code [21] developed at the Fraunhofer IST is used to model the plasma discharge time evolution by self-consistently computing charged particle trajectories and their electric field distribution. For all presented numerical studies, the magnetic field generated by the permanent magnets of the planar magnetron is first computed in the simulation model corresponding to its experimental counterpart. Then, a PICMC simulation of the DCMS plasma discharge is performed with the target negative voltage is regulated to reach the power setpoint. Once the global discharge current and voltage reach a steady-state condition, the target voltage is switched to +50 V, and the free plasma evolution is computed for about 200 μs to simulate the plasma afterglow. However, due to numerical constraints inherent to the PICMC method in terms of cell spacing and time-step, the HiPIMS main pulse discharge cannot be simulated because of the high charged particle densities and currents. A power setpoint of 2 W is therefore used in the DCMS simulations, about two orders of magnitude lower than the experimental average values. The current density reached before the voltage inversion is $\sim 0.5 \text{ mA cm}^{-2}$, while a value in the range $1\text{--}3 \text{ A cm}^{-2}$ is achieved in the experiments. All the physical elements present in the experimental setups, namely the vacuum chamber, the MEA extraction hood, and the magnetron, are modelled as grounded surfaces, unless specified otherwise.

The three species of interest included in the simulations are neutral argon (Ar), singly ionised argon (Ar^+) and electrons. At the beginning of the DCMS discharge, a uniform Ar pressure of $8 \times 10^{-3} \text{ mbar}$ (as in the experiments) is set in the simulation volume, with about 20 Ar super-particles present in each Cartesian cell to ensure proper collision statistics, while Ar^+ and electrons are uniformly seeded with an initial density of $5 \times 10^{12} \text{ m}^{-3}$. Niobium particles in

the neutral (Nb) or ionised (Nb^+) state are not included in the simulations because the niobium ionisation is negligible in low power DCMS discharges, and their presence would have no impact on the plasma behaviour. Volume collision reactions have been detailed in [22], while surface reactions have been extensively described in [23]. In particular, electrons are assumed to be fully collected on any physical surface that they reach, without further re-emission. Ar^+ particles are also fully collected with the re-emission of one Ar, and of electrons according to the IISEE yield of 0.13, with a Gaussian energy distribution. This is equivalent to consider only potential (*Auger*) secondary emission, while neglecting kinetic emission and emission under bombardment by other types of particles (e.g. neutral, metastable or photons).

Typical simulation outputs include, non-exhaustively, the time-resolved charged particle densities, energies and electric potential at the scale of the Cartesian volume mesh, but also fluxes of charged particles on the physical surfaces.

3. Experimental results

3.1. Time-integrated Nb^+ IEDFs

In a previous work [9], the Nb^+ IEDFs were measured with the *CERN-balanced* setup in HiPIMS at main pulse duration of 30 μs , PP delay of 4 μs , PP duration of 100 and 250 μs , PP amplitude of +50 V, and 1 kHz repetition rate, reaching 1 A cm^{-2} peak current density averaged over the whole target surface. The IEDFs included a dominating energy peak at $\sim 26\text{--}28 \text{ eV}$ [9], which is herein further investigated in the same experimental conditions (1.5 A cm^{-2}) by reducing the PP duration. As shown in figure 2, the amplitude of the Nb^+ energy peak at $\sim 28 \text{ eV}$ progressively increases for longer PP: it is missing for a PP of 20 μs , indistinguishable from a broader peak in the $10\text{--}30 \text{ eV}$ range for 50 μs , and dominant for 100 μs , where a multi-peaked distribution can be observed. The mechanism responsible for these features will be further discussed in section 3.2.

Spatially resolved time-integrated Nb^+ IEDFs are obtained in the *Gencoa-balanced* setup where the target-MEA distance can be continuously varied (figure 3). The same operating parameters of [9] are used, except for the PP duration (200 μs). A population close to 40 eV is observed up to 110 mm from the target, where it starts to decrease, but the dominant peak is located at 28 eV for all the explored target-MEA distances up to 230 mm.

3.2. Plasma and floating potential evolution

The measured ion energies necessarily reflect the V_{pl} spatial profile and temporal evolution encountered by the ions along their trajectories, with most of the ion acceleration that should occur at the grounded wall sheath. An LP is therefore used to measure the IV curves at different distances from the target, from which the spatio-temporal evolution of V_{pl} can be extracted. In figure 4, the I – V curves measured during the afterglow at 15 mm and 115 mm from the target are shown for the same HiPIMS parameters of section 3.1, except for a peak current density of $\sim 3 \text{ A cm}^{-2}$: the HiPIMS discharge at 1 kHz

Figure 2. Time-integrated Nb^+ IEDFs for different PP durations at a target-MEA distance of 145 mm.

Figure 3. Time-integrated, spatially-resolved, Nb^+ IEDFs.

repetition rate starts at 0 μs , ends at 30 μs , while the PP starts at 34 μs , and ends at 234 μs .

The knee of the I - V curves, which corresponds to V_{pl} , is located close to 40 V up to 58 μs . For the following 12 μs the curves are perturbed or shifted towards a lower voltage until 70 μs , when a knee close to 22 V can be identified for the data at 115 mm. Starting from 74 μs , V_{pl} stabilises at around 28 V at 115 mm, which is consistent with the dominant energy peak in figure 3. These results indicate a time evolution of the bulk V_{pl} featuring a first value close to 40 V until $\sim 58 \mu\text{s}$. This initial rise is followed by a D & R stabilizing at ~ 28 V, with the V_{pl} perturbation occurring along the magnetron axis within 10 μs at two locations that are 100 mm apart. Assuming a negligible variation of the electron temperature during the first phase of the afterglow, as discussed in [16], V_{fl} measurements can be used to confirm the V_{pl} amplitude variations. Examples of V_{fl} time-traces obtained with the LP are given in figure 5, at the same locations as those of the I - V curves.

Figure 4. I - V curves measured at different times from main pulse start at 15 mm (a) and 115 mm (b) from the target.

As the target potential is triggered to the PP set-point of 50 V at 34 μs , the V_{fl} increases accordingly between 35 μs and 55 μs . This occurs at first close to the target (15 mm from it), then 115 mm from it with a slower ramp-up. This first rise can be interpreted as a net flow of electrons out of the system, preferentially via the target, until a sufficient electric field establishes at the target sheath to maintain the plasma bulk quasi-neutrality. We note that the voltage ramp up characteristic time to the set-point of $\approx 20 \mu\text{s}$ is determined by the HiPIMS module dynamics rather than by possible capacitive couplings. The V_{fl} local maximum is reached at first close to the target at 55 μs , with a subsequent drop that is sharper at 115 mm from the target. A second slower rise of V_{fl} to an intermediate value takes place between 60 and 100 μs , with some oscillations for the time traces close to the target that can be similarly observed in the magnetron current and voltage. When PP lasts at least 50 μs , the broad energy peak of figure 2 centered at 20 eV, and in the range 10–30 eV, results from the ions accelerated by the MEA sheath electric field when the plasma bulk is experiencing the V_{pl} D & R. Therefore, the lower limit of this peak provides an indication of the minimum value of V_{pl} (~ 10 V) reached at the dip of the D & R. For a longer PP of 100 μs , the peak amplitude at 20 eV remains almost constant, while the peak at 28 eV increases by more

Figure 5. Time traces of the magnetron current and voltage. V_{II} measured with the LP at 15 and 115 mm from the target.

than one order of magnitude because of the time-integration of the ion flow for the successive $50 \mu\text{s}$, according to the accelerating fields in the MEA sheath during the bulk V_{pl} plateau after the D & R.

To explore more in detail the V_{II} time-evolution close to the magnetron, the floating LP is displaced radially at 15 mm from the target surface. The 2 mm spatial steps lead to the discontinuous features in the radial direction that can be observed in figure 6. Time oscillations originate at the center of the target after the V_{II} drop, and propagate radially outwards with a progressive damping. The associated frequency of 130 kHz is much slower than the electron and ion plasma frequencies in the GHz and MHz ranges, respectively, assuming a plasma electron density of $n_e \approx 10^{13} \text{ cm}^{-3}$. Breathing modes of the order of 100 kHz are discussed in previous investigations [24], where it is pointed out that relatively slow processes ($\leq 100 \text{ kHz}$) are an evidence for a strong role of neutral density (rarefaction). Floating potential oscillations during the electron sheath formation up to 50 kHz have been previously measured and associated to the repetitive formation and collapse of the double layer structure of anode spots [25], that will be further discussed in section 5. Further investigations on these fluctuations are beyond the scope of this work.

3.3. Comparison to simulations

Numerical simulations are performed firstly for the *Gencoa-balanced* configuration to compare the simulated V_{pl} evolution with the experimental one, using a cell resolution of $0.5 \times 0.5 \times 0.5 \text{ mm}^3$. In figure 7, the temporal evolution of the normalized experimental V_{II} and simulated V_{pl} in the *Gencoa-balanced* configuration is shown at 15 mm along the target axis, with the afterglow starting at $t = 0 \mu\text{s}$. This time axis is kept in section 4. Both time-traces feature a drop followed by a stabilization within the first $60 \mu\text{s}$. Considering that the simulated discharge power is two orders of

Figure 6. V_{II} as a function of time, at different radial positions, at 15 mm from the magnetron target.

Figure 7. Experimental and simulated time traces of, respectively, V_{II} (blue circles) and V_{pl} (black dashed, red continuous line) at the center of the target at 15 mm from it.

magnitude lower than in the experiment, the observed dynamics is likely not related to the higher peak powers and densities reached in HiPIMS. This is a strong indication that the V_{pl} D & R is a general afterglow feature in bipolar magnetron sputtering discharges. A second set of numerical simulations is performed for the *CERN-balanced* assembly, at a cell spacing of $1 \times 1 \times 1 \text{ mm}^3$.

The two simulations feature the same physical parameters, except for the magnetic assembly and the anode/cathode geometrical configuration, as can be observed in figure 1. These produce a balanced configuration in both, with a similar D & R, and thus justifies dedicated investigations based on the *CERN-balanced* assembly for a deeper understanding of the physical mechanisms responsible for the afterglow dynamics in section 4.

4. Numerical simulations

In this section, the bipolar discharge afterglow of the *CERN-balanced* configuration is simulated and analysed at first

in section 4.1. Alternative configuration are explored in section 4.2 to highlight the role of the magnetic field in the afterglow dynamics.

4.1. CERN-balanced magnetron afterglow

To gain a global overview of the temporal dynamics of both ions and electrons, the flow of both species is shown in figure 8(a) and analysed hereafter. The total net flow is calculated as $\Phi_{\text{tot}} = \Phi_{\text{e-ground}} + \Phi_{\text{e-target}} - \Phi_{\text{i-ground}} - \Phi_{\text{i-target}}$. The V_{pl} time-traces are included in figure 8(b) at six representative spatial positions in the plasma: at the center (C, $r = 0$ mm), racetrack (R, $r = 14.5$ mm), and edge (E, $r = 21.5$ mm) of the target, at perpendicular distances of both 2.5 and 115 mm from it. These hence reflect the dynamics close to the target surface, as well as that in the plasma bulk. The corresponding 2D profiles of V_{pl} are shown in figure 9(a) at 25, 30, 40, and 45 μs from the triggering of the PP. The evolution of the corresponding electron density, charge imbalance, electric field, and ionization rate are reported in figures 9(b)–(e), respectively, and analysed at each time window in the following.

4.1.1. Initial V_{pl} (0–25 μs). As the target is set to the PP voltage, $\Phi_{\text{i-target}}$ quickly becomes negligible after $\approx 25 \mu\text{s}$, with the large majority of the ions leaving the system via the grounded chamber. Their outflow is mostly balanced by the electron flow across the target, reflecting the intrinsic drive of the bulk plasma to maintain quasi-neutrality. We note that this situation with the electrons and ions leaving the plasma via two distinct surfaces corresponds to the *global non-ambipolar flow* discussed in [26, 27]. However, the model proposed in those articles based on the estimates of the target-to-wall surface ratio cannot be directly applied to our configuration due to the magnetic field. It alters the effective charge collection area on the target surface and localizes the plasma far from most of the grounded walls. Furthermore, there is no active plasma source in our system during the afterglow.

$\Phi_{\text{e-ground}}$ is slightly negative, implying a greater electron inflow than outflow due to the secondary electron emission induced by the ion bombardment on the grounded surfaces. The positive total flow of charge is consistent with the negative slope of the V_{pl} on all the presented time-traces.

4.1.2. V_{pl} drop (25–30 μs). At $\approx 25 \mu\text{s}$, the inflection in the target electron flow observed in figure 8(a) begins in the highlighted green region. This leads to a local maximum of Φ_{tot} at 30 μs , which is a consequence of the electron density evolution near the target shown in figure 9(b), and likewise with the magenta contours in figure 9(d): due to the large volume of magnetic field lines closing onto the target in this balanced configuration, the electron population is repelled from the guarding ring at the edges and drained to the centre following the magnetic field lines and target positive bias potential. Keeping in mind the 3D nature of the set-up, electrons are subject to a lensing effect towards the target centre. This electron flow is sufficient to lead to a deviation from quasi-neutrality here, as quantified in figure 9(c), and therefore causes the local

Figure 8. In panel (a), the electron (Φ_{e}) and ion (Φ_{i}) flows drawn out of (positive values), and entering (negative values) the system are shown. The particle flows across the positively biased target (Φ_{target}) are distinguished from those across the grounded chamber (Φ_{ground}). The total net flow (Φ_{tot}) is calculated including the electron flow as a negative contribution. For readability, Φ_{target} and Φ_{tot} are up-scaled by a factor of $\times 10$. Panel (b) shows the evolution of V_{pl} at the center, racetrack, and edge of the target surface, at the two indicated normal distances. The regions shaded in green, red, and blue are discussed in more detail in this section.

depression in V_{pl} observed in figure 8(a). The increasing build-up of negative space charge thus inhibits some of the electron flow from being drawn in the target centre, until the total flow of charge in the system reaches its maximum at 30 μs . Near the target racetrack and edges, the electron density has been mostly depleted along field lines of shorter connection lengths towards the target. With no noticeable deviations from quasi-neutrality, this region can be considered as increasingly depleted of ions as well. The overall plasma potential drop thus seems dominantly set by the bias and space-charge build-up near C. With quasi-neutrality holding throughout most of the plasma away from the target center, the likewise falling evolution of V_{pl} even at distant locations is thus not unexpected. However, the specific strength or delay of the potential drop (see e.g. figure 8(b), dashed) relies on the specific charge dynamics in the system. For instance, the delays near R and E

Figure 9. 2D plasma parameters at 25 μs , 30 μs , 40 μs , and 45 μs from the start of the PP. (a) V_{pl} profiles with the dots corresponding to the positions of the time-traces shown in figure 8(b) with the same color. (b) Electron density profiles. (c) Profiles of the ion–electron density difference. Local statistical fluctuations between the electron and ion density are present in the raw data. Each profile was therefore time-averaged with two more at $\pm 0.5 \mu\text{s}$, which is much shorter than the time scale of all D & R dynamics. Furthermore, the profile was smoothed with a circular Gaussian kernel with $\sigma = 1 \text{ mm}$, corresponding to the raw data resolution. All physical spatial structures are therefore still resolved. (d) Electric field profiles calculated as 2D gradients of the V_{pl} in (a), with contours of the electron density shown at $n_e = \{2, 4, 6\} \times 10^{14} \text{ (m}^{-3}\text{)}$ in darkening shades of magenta. (e) Ionization rate profiles on a *logarithmic* color scale. Note the stringent localization of ionizing events at locations where significant electron densities meet strong electric fields in (d).

close to the target are likely the result of the transition between residual plasma shielding effects at lower densities, and the electric field taking its free-space structure as the local Debye length increases.

4.1.3. Propagation and rise (30–40 μs). During this time-window (figure 8(b), red band), the V_{pl} time-traces at **R** and **E** still gradually decrease, albeit more slowly. After having caused the minimum of V_{pl} at 30 μs , the negative space-charge close to the target centre slowly begins to dissipate nonetheless. This is consistent with a gradual recovery of V_{pl} from its original drop, as well as the diminished inflection of the target electron flow (see figure 8(a)). However, this flow even

becomes stationary, resulting in a sharp decline of the total current. Moreover, the target ion flow begins to re-emerge at $\approx 38 \mu\text{s}$.

These observations beckon the investigation of new ionization events near the target center, requiring an analysis of when electrons reach the energy threshold for ionization collisions. Throughout much of our system, electron–neutral collision-times of up to a few hundred nanoseconds are estimated, with mean free paths of the order of 10 cm. However, as the electrons approach the target, the effective mean free path decreases due to their Larmor motion and azimuthal drifts. This leads to an increasing number of collisions in this region. Electrons can therefore only acquire a limited

Figure 10. Simulated V_{pl} time evolution at the center of the target at different axial distances in a configuration with the magnetron guarding ring at the PP potential at the triggering of the PP. The same features are verified on the racetrack and at the edge of the target.

Figure 11. Simulated V_{pl} profiles at 2.5 mm from the target on its axis, with and without the SEE-g, with and without the balanced magnetic field $\mathbf{B}$.

amount of energy over their trajectories between collisions. This holds especially during the initial drop in V_{pl} , when spatially smooth potential gradients and low electric fields permeate most of the region of high electron density, as specifically illustrated in figure 9(d). Nonetheless, ionization never completely ceases within the volume, but it is only temporarily diminished as most free electrons converge towards the center.

While V_{pl} takes its lowest values near the target centre, the local potential gradients consequently steepen, implying higher local electric fields. At the same time, the peak density of the electron population can still be found here (figure 9(b)). It is this combination that drives a local rise in ionization rate by more than a factor of $\times 10$ between 30 and 40 μs ,

Figure 12. Average e-Ar ionization collisions in the plasma (a) and target electron emission (b) for the possible permutations of $\mathbf{B}$ and SEE-g.

with 2D profiles shown in figure 9(e). Although numerous electron–ion couples are thus generated near $\mathbf{C}$, electrons are quickly drained at the target at first. This hastens the dissipation of the negative space-charge until it even inverts towards positive values, reinforcing the rise of V_{pl} .

4.1.4. Stabilization (40–45 μs). Spatially, the dynamics close to the target culminate in the creation of a hat-shaped region between 40 and 45 μs (blue band in figure 8(a)), inside which the plasma potential is very close to the target bias, as shown in figure 9(a). This region of ion production thus features very low electric fields within. With electrons being drained very efficiently, the total current even becomes negative during this time, while a positive slope of V_{pl} is observed on all the curves in figure 8(b).

At $\approx 45 \mu\text{s}$, the central potential of the hat-shaped region even exceeds the target bias, so that some ions might be accelerated towards the target. Emitted electrons can thus likewise traverse the hat-shaped region, and even slightly accelerate away from the target. Conversely, electrons are confined by the potential drop at the edge of the hat-shaped region, leading eventually to a local stabilization of both electron and ion-densities, with the n_e near the target increased by a factor of two with respect to that at 40 μs (figure 9(b)). The residual space charge is smaller compared to before, and mostly positive.

4.1.5. Gradual dissipation ($\geq 45 \mu\text{s}$). Overall, a sustained rise of the plasma potential is reached, and the drawn total flow of charge is again slightly positive from 45 μs on, with most electrons being drawn through the target centre. It is reasonable to

Figure 13. Magnetic assembly with the corresponding magnetic field lines computed with the FEMM software [20] for the two simulated unbalanced configurations (a) and (b). The indicated dimensions are in mm. In red, the distance of the magnetic null-point ($B_z = 0$) from the target surface. The V_{pl} evolution at 2.5 mm from the target center is shown in (c).

expect that renewed electron lensing would compete with the rise of V_{pl} to form a local second drop. This could ultimately result in an oscillatory behaviour, as shown experimentally in figure 5, or in figure 6., similar to the oscillations in the electron sheath reported in a previous work [25]. While e.g. figure 9(c) points to the formation of a slightly negative space-charge at the top of the ionization region already at 40 μs, the investigation of any eventual oscillations in V_{pl} are clearly at the limit of our numerical capacity, as seen in figure 7. Either way, the density of successful ionizations continues to gradually diminish and the plasma in the hat-shaped region is slowly extinguished.

4.2. Alternative configurations

The model discussed so far hinges on the presence of a magnetic field geometry dominated by field-lines closing on the target centre that cause the discussed electron lensing. To further illustrate the role of the magnetic field, the afterglow is initiated with the magnetron guarding ring set to the same PP voltage as the target, instead of grounding it, but from the same plasma simulation described in section 4.1. This modified configuration of the *CERN-balanced* assembly artificially increases the size of the surface at positive potential, providing alternative paths of collection for the electrons, effectively preventing the accumulation of space-charge at the target centre. Hence, the V_{pl} evolution does not include a D & R anymore, as can be observed in figure 10.

Furthermore, the effect of the following parameters on the afterglow have been explored for the reference *CERN-balanced* case with grounded guarding ring on both the plasma discharge and the afterglow: the IISEE from the grounded surfaces including chamber wall, anode ring and MEA (SEE-g), the IISEE from the target (SEE-t), and the magnetic field (**B**). The removal of SEE-t from the simulation yields no significant differences in the V_{pl} evolution (not shown herein), while removing SEE-g leads to a 5 μs delay in the V_{pl} D & R, as

observed in figure 11. Given that the system is deprived of a large net-source of electrons, the accumulation of space charge near C is delayed, and so are the drops in V_{pl} and SEE-t, along with their eventual rise. An overview of the total ionization rate and SEE-t between the different afterglow simulation scenarios is given in figure 12. The data for the baseline scenario is shown with a continuous black line. In the scenarios without **B**, electron flows are drawn throughout the whole target area, no space-charge is accumulated at the centre, and the regions near **R** and **E** remain as populated as at C. Therefore, V_{pl} stays unperturbed and features mostly smooth gradients while the plasma extinguishes itself due to the continuous decrease of ionizations and lack of confinement. The e-Ar ionization collisions increase by two orders of magnitude within the entire plasma volume by including the SEE-g, as illustrated in figure 12(a). The SEE-t in figure 12(b) likewise features a D & R, as observed for the V_{pl} in the presence of **B**, with the same anticipation of it by 5 μs in the presence of the SEE-g.

Finally, the setup dimensions and magnet materials of figure 1(a) have been modified to obtain the unbalanced magnetic assemblies depicted in figures 13(a) and (b). For each of these cases, the PP afterglow modelling follows a new DCMS plasma simulation with its corresponding magnetic field computation. The others simulation parameters remain unchanged, when compared to the *CERN-balanced* baseline. As the magnetic null-point is situated increasingly closer to the target, more and more magnetic field-lines open up towards the chamber volume, and no longer close near the target centre. Again, this diminishes the electron lensing effect and hence the accumulation of space charge there. While a smaller deviation from quasi-neutrality is still observed in the intermediate case in figure 13(a), the overall plasma density diminishes much faster than in the baseline case. Since the plasma is almost extinguished after the potential drop, only a slight rise occurs as this minor space charge dissipates, and no new ionization region is formed. In the strongly unbalanced

scenario displayed in figure 13(b), no noticeable space charges are accumulated and the D & R is hence prevented, as shown in figure 13(c).

This confirms that the electron dynamics under the influence of a balanced magnetic field are the key factors responsible for the observed D & R of V_{pl} . The onset of these dynamics appears gradual, and strongly dependent on the specific conditions required to initiate an accumulation of negative space-charge in front of the target centre. If a sufficient electron density remains during the dissipation of this space-charge, the steepened plasma potential gradients can lead to the development of a new, concentrated ionization region, stabilized by a reversal to positive space-charge.

5. Conclusion

The dominant energy peaks of the experimental IEDFs measured in bipolar HiPIMS are consistent with the plasma potential time-evolution measured during the afterglow. A V_{pl} D & R is observed over the whole plasma volume. To obtain more insights into the physical mechanisms behind the afterglow evolution, 3D PICMC simulations have been investigated, showing a similar D & R in the V_{pl} . The simulated discharge average power is two orders of magnitude lower than in experiments, which indicates that the high power and plasma density peaks reached in the HiPIMS regime are not necessary for the observed V_{pl} behavior. The numerical decoupling of the discharge parameters allows for the identification of the magnetic field configuration as a key element in the V_{pl} evolution during the afterglow. With a balanced magnetic field, the V_{pl} is not stationary, but features a D & R to a value lower than the PP set-point throughout most of the volume. By analyzing the plasma parameters during the first 45 μ s of the afterglow, a lensing of the electrons towards the center of the target is observed. This is caused by the magnetic field lines geometry, and leads to a local negative space-charge and consequent drop of the plasma potential. After passing its peak, the local electric fields enhance the ionizing collisions close to the target by one order of magnitude, allowing for a space-charge inversion to a net positive charge and corresponding local rise of the plasma potential. Such features are likewise observed in double-layers and anode spots in the absence of magnetic fields [27, 28], where oscillations similar to those herein have been measured and explained as the double layer stabilization in the electron sheath. We also note that the double layer potential drop between the region close to the target and the bulk plasma is slightly higher than the ionization potential of Ar [29].

The picture outlined here is confirmed by simulating modified magnetron assemblies to reduce the electron lensing towards the center of the target such that the D & R is no longer observed. First, by setting the magnetron guarding ring at the same PP as the target at the beginning of the afterglow, providing a second path for the electrons to flow outside the system, then by removing the magnetic field during the afterglow, and finally by simulating a plasma discharge and afterglow in unbalanced magnetic configurations. The

results obtained in the simulated unbalanced magnetic configurations are envisaged to be confirmed experimentally. The presented mechanism characterizing the afterglow dynamics in the presence of a target potential reversal is relevant in general for bipolar magnetron operation, and more specifically for bipolar HiPIMS with PP. The associated spatio-temporal evolution of V_{pl} is the main cause for the acceleration of the sputtered ions in the substrate sheath. The resulting impact energy on the growing film can be tailored to modify the film morphology.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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