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Evidence of ion energy distribution shift in HiPIMS plasmas with positive pulse

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Corrigendum: Evidence of ion energy distribution shift in HiPIMS plasmas with positive pulse (2019 *Plasma Sources Sci. Technol.* **28** 01LT03)

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Figure 3. Time-integrated EDFs of Nb^+ for the case without PP, +50 V PP of 100 μs , and 250 μs duration.

A problem in the analysis of the acquired MEA data-set has been identified for the Nb ions count-rate. The correct versions of figures 3 and 4(a) are the following: the main features of the previous versions of the figures are maintained. In figure 3, the IEDF without PP is still centered at 2 eV but it

shows a more visible tail in the range 3–10 eV. The IEDFs with the PP show a dominant energy peak centered at 28 eV, with a higher fraction of ions in the range 43–50 eV. A higher value of count-rate is generally observed, which is reflected in figure 4(a) for the Nb ion curves. The Nb^+ population at

Figure 4. (a) Ar^+ and Nb^+ ion fluxes as a function of the PP duration, evaluated on the EDF at the low energy maximum (1.75 eV) and at the high energy maximum.

1.75 eV drops below 10^2 cps for $120 \mu\text{s}$ PP, and the crossing point between low and high energy peaks occurs for a PP duration close to $70 \mu\text{s}$.

To conclude, the herein presented corrections do not significantly alter the results discussed in the letter. While the energies of the dominant peaks of the Nb^+ EDF are not changed, the count-rate is corrected to higher values, indicating an effective ion flux of metal ion much closer to that of the sputtering gas ions of Ar^+ .

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Letter

Evidence of ion energy distribution shift in HiPIMS plasmas with positive pulse

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Abstract

The deposition of a dense and void-free coating in complex geometries can be obtained with High Power Impulse Magnetron Sputtering (HiPIMS) by negatively biasing the substrate. This accelerates the incoming metal ions that consequently densify the film. However, a biased substrate can be a technical challenge for large objects that are generally kept grounded. HiPIMS with a positive voltage pulse after the main negative one is a potential alternative to be still fully investigated. In this letter, we report on the first measurements of a dominant population of energetic ions in HiPIMS with a positive pulse. Time-integrated energy distribution functions of the background gas ions (Ar^+) and target ions (Nb^+) are measured with and without the positive pulse. The peak of low energy ions at ~ 2 eV is shown to disappear by varying the duration of a +50 V pulse. For a sufficiently long duration of 100 μs , energy peaks in the range of ~ 30 eV are observed for both Ar^+ and Nb^+ . Negligible ion fluxes at the energies below 10 eV are measured for 250 μs , indicating an effective acceleration of ions towards the substrate. Time-resolved ion flux measurements provide insights on the positive pulse effect on the ion fluxes.

Keywords: HiPIMS, positive pulse, niobium, superconducting radio-frequency

The large fraction of ionized sputtered atoms in plasmas obtained with High Power Impulse Magnetron Sputtering (HiPIMS) is making this technique very attractive for a constantly increasing number of fields [1–6]. The scientific community is devoting a significant effort to investigate and unveil the physical mechanisms of HiPIMS plasmas [7–11], as well as to explore the advantages with respect to well known sputtering techniques such as Direct Current Magnetron Sputtering (DCMS) [12, 13]. Very recently, the effects of a positive voltage pulse (PP) applied to the target after the main negative pulse in the HiPIMS regime have started to be explored [14, 15]. Originally used with DCMS processes, one advantage of the PP is the avoidance of charge build-up on the substrate in reactive coatings producing insulating or poorly-conducting films [16–18]. The use of HiPIMS with PP

for the coating of complex geometries is a further potential application [19]. By implementing standard cylindrical cathodes with grounded substrates featuring concave and convex surfaces, particles necessarily impinge with small grazing angles at specific locations. This leads to a columnar growth of the films and the consequent presence of voids. To improve the densification and obtain a conformal coating, a negative voltage is generally applied to the substrate to have the metallic ions accelerated and collected with a normal angle of incidence [20, 21]. However, this solution is not optimal for applications with large objects, such as the Nb coating of superconducting radio-frequency (SRF) cavities [22]. As underlined in previous studies [23], obtaining an equivalent effect with the PP in HiPIMS can open new perspectives, with the simplification in the design of the cathode and of the coating setup. However, no evidence of ion energy distribution shift in HiPIMS plasmas with a positive pulse has been published so far.

In this manuscript, we provide experimental evidences of a significant ion acceleration in the presence of a +50 V PP

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Figure 1. (a) Geometry of the setup including the magnetic configuration (black lines) with the null-point (red dot). (b) In blue, the applied HiPIMS voltage pulse of 30 μs duration, without PP (continuous line) and with PP of +50 V and 250 μs duration (dashed line). In red, the corresponding discharge currents.

with HiPIMS. Time-integrated energy distribution functions (EDFs) of Ar and Nb (target) ions are measured. For a specific set of parameters, a dominant population of ions at ~ 30 eV is clearly observed and no ions are detected at energies below 10 eV. The results are complemented with time-resolved measurements, where the effect of the PP on the ion fluxes is shown.

The experimental setup is composed of a 43 cm long vacuum chamber, with Ar as sputtering gas at an operating pressure of 8×10^{-3} mbar. In figure 1(a) a schematic of the experimental setup is presented. A magnetron source at 15° with respect to the horizontal plane is installed on the bottom. It houses a 2 mm thick, 50 mm diameter Nb target. A grounded guarding ring (anode) of 46 mm inner diameter is placed on the top of the target, with a gap of 1 mm. The magnetron features a balanced magnetic configuration with the null-point at ~ 3.5 cm from the target surface, as indicated in figure 1(a) with a red dot, together with the field lines computed with FEMM [24]. A DC power supply powers the system through a 2 kW HiPIMS module. The implemented HiPIMS setup provides the possibility of varying the delay, duration and amplitude of the PP after the end of the main pulse. An ion mass and energy analyzer (MEA) is used to discriminate the species and the energy of the collected ions. The model is a Pfeiffer Plasma Process Monitor 422 and it is mainly composed of a cylindrical energy analyzer and a quadrupole mass analyzer. The MEA is vertically aligned and installed on the top of the vacuum chamber in order to have the extraction hood (EH) intercepting the axis of the target within 5 mm accuracy. The EH features a 100 μm entrance orifice and it can be externally biased or kept electrically floating. With the implemented ion optics parameters, the acceptance angle resulting from SIMION 8.1 simulations [25] varies between 20° for energies below 10 eV, to less than 3° for energies above 20 eV. For the herein presented measurements, the EH is kept grounded to reproduce the coating conditions of a grounded SRF cavity. The distance between the EH and the target surface is 145 mm. Time-resolved measurements can be obtained from the MEA ion counter

preamplifier by extracting two signals based on the emitter-coupled logic (ECL) from the secondary electron multiplier output. These signals are acquired by a multichannel scaler (MCS) after passing through an ECL-to-TTL converter. The MCS allows to integrate the detected events in each channel with a dwell-time down to 50 ns. By using this lower value and 16 384 channels, the incoming ions are measured over the ~ 820 μs following the trigger of the HiPIMS voltage pulse by accumulation over 20 000 periods. The pulsed voltage is measured by a high-voltage probe (4 kV) with 400 MHz bandwidth. Current measurements are obtained with a current probe measuring up to 30 A with 100 MHz bandwidth. For the investigated discharges, voltage pulses of -550 V with a repetition rate of 1 kHz and duration of 30 μs (3% duty cycle) are applied, as shown in figure 1. The presented time-traces have been averaged over 50 pulses to improve the signal-to-noise ratio.

In all the explored scenarios with PP, the positive voltage ramp is triggered 4 μs after the end of the main negative pulse. This is the shortest accessible delay with the implemented HiPIMS system. The applied PP voltage is of +50 V and the durations range from 20 to 250 μs . A peak current of 20 A for the main pulse is measured, as visible in figure 1 for the 0 and 250 μs PP. The associated current density is 1 A cm^{-2} . For the configuration with 0 μs PP, this corresponds to an average power of 230 W. In figure 1, the reference configuration without PP is presented (continuous line), together with the longest explored PP of 250 μs (dashed lines). We note that by increasing the duration of the PP, a progressive delay in the current ramp of the main pulse occurs. In particular, a PP of 250 μs corresponds to a delay of ~ 10 μs with respect to the trigger of the voltage pulse, as can be observed in figure 1. This leads to variable power, from 230 W for the configuration without PP, to 110 W for that with 250 μs PP. The appearance of this delay with the application of the reversed polarity has been observed [14] and discussed [26] in previous works. It has been ascribed to the suppression of the plasma ignition by helping the diffusion loss of charged particles in the after-glow discharge. In the framework of the present work, it has been verified that the EDFs do not show significant changes, either by increasing the duration of the voltage pulse to obtain the same effective 30 μs current pulse duration, or by increasing using the same applied average power of 230 W.

The time-integrated EDFs of Ar^+ and Nb^+ are measured for the explored operating parameters. In figure 2, the most relevant spectra for Ar^+ are shown, with an energy resolution of 1 eV. The spectrum without PP features a peak at 2 eV with no detectable ions at higher energies. Progressively increasing the PP duration, for constant amplitudes of +50 V, the spectrum undergoes significant changes. With a PP lasting 100 μs , the peak at 2 eV is reduced by almost one order of magnitude, while the ion population in the energy range 2–40 eV significantly increases. In particular, a second peak appears at 28 eV with a count rate comparable to that of the 2 eV peak. The spectrum with a PP of 250 μs presents a negligible portion of Ar^+ for energies below 10 eV ($\sim 10^2$ cps), while a second peak at 32 eV can be observed in

Figure 2. Time-integrated EDFs of Ar^+ for the case without PP, +50 V PP of 100 μs , and 250 μs duration.

Figure 3. Time-integrated EDFs of Nb^+ for the case without PP, +50 V PP of 100 μs , and 250 μs duration.

addition to that at 28 eV. A similar evolution of spectra is obtained for Nb^+ , as presented in figure 3, with two main differences. First, with the 100 μs PP the peak at 28 eV is already dominant with respect to that at 2 eV. Second, for the last configuration with 250 μs PP, the peak at 32 eV starts only to appear. The discrepancy between the applied +50 V and the measured energy peak close to 30 eV can be explained with a partial raise in the plasma potential during the after-glow discharge, as measured in previous works [23, 27]. Ion-neutral collisions could also contribute to the measured values of energy. A mean-free-path of approximately 2.7 cm and 6.3 cm is estimated, respectively, for Nb^+-Ar and Ar^+-Ar collisions. We remark that the spectra of Ar^{++} and Nb^{++} have been measured as well, showing features similar to the presented results, except for the peak intensity being one order of magnitude lower.

To make more evident the ion flux evolution at different energies with respect to the PP duration, we show in figure 4(a) Ar^+ and Nb^+ count rates associated to the energies of the dominant EDF peaks as a function of the explored PP durations. Dedicated refined energy scans were performed with an energy resolution of 0.25 eV. We have verified that

the low energy peak, identified at 1.75 eV, does not drift in energy in the range of operating parameters. However, the peak at ~ 28 eV has been revealed to slightly drift from 26.25 to 28.75 eV by increasing the PP duration, as clearly visible in figure 4(b) for Ar^+ . A similar behavior is observed for Nb^+ . In figure 4(a), the count rate corresponding to the maxima of the spectra has been considered. For Ar^+ , the drop of ion flux at 1.75 eV starts to be significant for PP durations higher than 100 μs and can be considered negligible (below 10^2 cps) for PPs longer than 200 μs . On the other hand, the high energy ion flux undergoes a steep rise in the range 50–100 μs , and then stabilizes. The crossing point between the two curves takes place close to 100 μs . Similar features can be observed for the Nb^+ , where the 1.75 eV ion population drops below the 10^2 cps threshold for 100 μs PP, even faster than for Ar^+ at 1.75 eV, while the crossing point between the low and high energy peaks occurs for a PP duration close to 70 μs .

Further insights in the dynamics of ion fluxes of the different species can be obtained by real-time measurements with the MCS. The results presented in the following are complementary to the time-integrated EDFs. A detailed investigation of the dependence of ion flux dynamics with

Figure 4. (a) Ar^+ and Nb^+ ion fluxes as a function of the PP duration, evaluated on the EDF at the low energy maximum (1.75 eV) and at the high energy maximum. (b) Refined EDF of Ar^+ for different PP durations.

respect to the PP parameters is of great interest, but it goes beyond the scope of this letter. In figures 5(a) and (b), an example of Nb^+ ion fluxes at 1.75 eV and at the energies corresponding to the peak of the spectra close to 28 eV, respectively, are presented. We can see in figure 5(a) for the Nb^+ ions at 1.75 eV that for an applied PP of 20 μ s, the shape of the incoming ion flux is modified. For higher PP durations the count rate progressively decreases together with a shift in time, as can be observed for the shown profiles for PP of 70 and 150 μ s. On the contrary, for the peak at 28 eV we can observe in figure 5(b) a higher Nb^+ count rate by increasing the PP duration from 60 to 80 μ s, where the count rate slope showed in figure 4(a) is largest, without a significant shift in time. Also, a comparison of the fluxes for PP of 80 and 250 μ s indicates that the peak count rate saturates. We underline that the zero of the time-axis corresponds to the trigger of the negative voltage pulse, as in figure 1. To assess the exact time of arrival of ions at the EH, and in particular to compare between different species and energies, the time-of-flight inside the MEA should be evaluated and subtracted, as included in previous works [28]. However, in the herein presented analysis, this correction is not necessary to get a

Figure 5. Time-resolved ion fluxes of Nb^+ at 1.75 eV (a), and at the energy corresponding to the maxima of the spectra close to 28 eV (b).

relative comparison between fluxes corresponding to different PP durations for a fix species and energy.

To conclude, we can assess that in the given experimental configuration, the implementation of a +50 V PP in the range 70–100 μ s makes the ions at ~ 30 eV dominating with respect to the ions at 1.75 eV measured without the PP. For a sufficiently long PP of 200 μ s, a negligible ion flux is measured at 1.75 eV for both Ar^+ and Nb^+ . The whole population of ions is shifted to higher energies, namely 28.75 and 32 eV for Ar^+ and mainly 28 eV for Nb^+ . The authors believe that these results are of utmost importance for future studies of HiPIMS, as they constitutes the proof of the effectiveness of the PP in accelerating the ions. Moreover, we have also shown the possibility of shaping the EDFs of a given species by varying the PP duration. This opens the way to future studies of time-resolved ion fluxes with PP, aiming at the optimization of the collected ions for a selected species and energy to precisely tune the desired thin-film properties (adhesion, morphology, contamination). To verify the effects of energetic ions on the film growth from the thin film point of view, coatings on samples at different grazing angles need to be investigated.

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