Fast Track Communication

The characteristic shape of emission profiles of plasma spokes in HiPIMS: the role of secondary electrons

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Abstract
A time-resolved analysis of the emission of high power impulse magnetron sputtering (HiPIMS) plasmas reveals inhomogeneities in the form of rotating spokes. The shape of these spokes is very characteristic depending on the target material. The localized enhanced light emission has been correlated with the ion production. Based on these data, the peculiar shape of the emission profiles can be explained by the localized generation of secondary electrons, resulting in an enhanced outward diffusion. This general picture is able to explain the observed emission profile for different target materials including gas rarefaction and second ionization potential of the sputtered elements.

Keywords: HiPIMS/HPPMS, plasma spokes, secondary electron generation, gas rarefaction, electron and ion dynamics

(Some figures may appear in colour only in the online journal)
resulting in different shapes of the spokes depending on the target material [1, 4], and the appearance of the ‘jets’ reported by Ni et al [11]. The model presented here is based on the data measured outside the closed magnetic field (CMF) region. In order to provide a complete understanding of the plasma dynamics of the spoke within the CMF region, additional non-invasive experiments are planned.

The experiment consists of two setups. One setup comprise a fast ICCD camera facing the target, with an acquisition time of 100 ns, as described in Winter et al [2]. The other setup is shown in figure 1 comprising a photomultiplier tube (PMT) and a double flat probe (2FP). The aim of the setup is to correlate optical and electrical attributes of the spoke, in a time-resolved mode. The light emitted from the racetrack, closest to the 2FP, was collected using two apertures and the PMT. The 2FP was mounted 10 mm from the racetrack, see figure 1. It consists of two flat concentric surfaces, one rectangular with a surface of 0.5 cm² and a surrounding one with a surface of 1 cm². The inner surface was biased, either to +30 V or to −30 V, to measure the electron or ion saturation current, respectively. The outer surface of the 2FP was not biased, and therefore the floating potential was measured. With this setup it is possible to measure simultaneously either ion saturation current and floating potential or electron saturation current and floating potential.

The Ar pressure was 0.2 Pa, the pulse length was 200 µs, and the frequency 10 Hz. The choice of target materials (Al, Ti, Cr, Cu, Nb, and Mo) was based on the difference in the second ionization potential. It is 13.6 eV (Ti), 14.0 eV (Nb), 16.2 eV (Mo), 16.5 eV (Cr), 18.8 eV (Al) and 20.3 eV (Cu). The current density varied between 2 A cm⁻² (Mo, Cr, Cu) to 5 A cm⁻² (Al, Ti, Nb), depending on the target material.

When applying a positive bias it was observed that peaks of the electron current correspond in time, shape and amplitude to the occurrence of the negative peaks in the floating potential, shown in figure 2 (measured with a Ti target at current density of 5 A cm⁻²). The electron saturation current increases when there is an increase in the electron flux, i.e. increase in electron density and electron velocity. Therefore, a drop in the floating potential can be regarded as an indication of increased electron flux.

The results are presented in figure 3. The image of the spoke is shown on the left side of figure 3. On the right side of figure 3 the oscillations of light emission (top), floating potential (middle) and ion current signal (bottom) in time are shown. The light intensity around the racetrack is extracted from the spoke image, and plotted as a red line on top of
Figure 3. An image of the spoke (left side) and the oscillations of the light emission (top), floating potential (middle) and ion current signal (bottom) in time (right side) for six target materials. The number placed in bottom left part of the image is the value of second ionization potential of the element.

The results show that for elements having lower second ionization potential than the first Ar ionization potential (15.8 eV), such as Ti and Nb, the emission shape of the spoke is elongated and diffuse. While for elements having higher second ionization potential than Ar, such as Mo, Cr, Al and Cu, the emission shape of the spoke is triangular with a sharp edge. The light intensity oscillation of Ti and Nb is wide and symmetric, with the slope rising immediately after reaching the minimum. This corresponds well with the diffuse shape of the spoke of Ti and Nb. The light intensity oscillation of Mo, Cr, Al and Cu has an asymmetric optical signal, with three rising slopes ending with a sharp fall. The emission peak is narrow compared to the length of the spoke, as the spoke in the images of Mo, Cr, Al and Cu. The broad light intensity signal of Ti and Nb is accompanied with the broad signal of ion saturation current. Similarly, the narrow light intensity signal of Mo,
Cr, Al and Cu is accompanied with the localized signal of ion saturation current, during the spoke. The oscillation of the floating potential, with the negative peak indicating the presence of high electron fluxes, are more pronounced for Ti and Nb (amplitude between 5 and 10 V), and less pronounced for other elements (amplitude around 1 V). One common thing for all target elements is that the negative peak in the floating potential appears at the beginning of the spoke, during the rise time of the light emission. The fact that electron and ion fluxes are detected by the 2FP (positioned outside CMF lines volume), indicate enhanced outward diffusion of electrons and ions within the spoke.

The agreement of the width of the spoke with the width of the ion saturation current implies that the spoke is an ionization zone [5], which is broad in the case of Ti and Nb, and narrow in the case of Mo, Cr, Al and Cu. The ionization process with highest probability in the low pressure plasma is the electron impact ionization [12], thus a presence of ions implies a presence of electrons with energies above 6 eV (typical ionization energy of elements presented in this paper). The source of energetic electrons is most probably secondary electrons (SE), i.e. electrons ejected from the target by impinging ions, and accelerated by the target sheath. A recent publication, assuming homogeneous plasma, suggests that ohmic heating might be an alternative source of energy for electrons [13].

1. Model of the spoke

Based on the presented experimental results we postulate a model of the spokes. Localization of the ion saturation current signal implies localization of the discharge current and the sputtering process. Localization of the sputtering process signifies that SEs will be generated locally and the sputtering wind will locally rarefy Ar gas. We argue that localized dynamics of these two physical processes defines the shape of the spoke. The leading edge of any spoke, observed from the top, starts as a narrow trace expanding in width and intensity. Side-way images recorded by a streak camera, presented by Ni et al [11], of the triangular shape spoke, suggest that light emission from the leading edge is trailing away from the target. We are interpreting these images as electrons and ions diffusing away from the target obeying ambipolar diffusion. This interpretation would imply that an increase in electron and ion flux is expected during the leading edge of the spoke. Indeed, an increase in electron and ion flux is observed for all materials, shown in figure 3, simultaneously with the rise of the light intensity, representing the leading edge of the spoke. The electron flux is more pronounced for Ti and Nb, compared to other elements, which will be the focus of future studies. In most cases the peak of ion saturation current comes after the electron flux peak, which could be explained by heavy particle inertia and the influence of the cathode presheath repelling electrons and attracting ions to the target. Analysing further the side-way images from [11], it seems there is a reduced SE production since the emission is severely reduced, as if the SEs are solely created at the very beginning of the spoke. Assuming localization of discharge current and sputtering process, it is reasonable to expect that abundant sputtering would result in the depletion of Ar neutrals in the target vicinity, i.e. Ar gas rarefaction by sputtering wind [14, 15]. This leads to a scenario where the gas rarefaction locally changes the composition of the impinging flux from an Ar dominated to a metal dominated flux, which in turn will hinder the SE production. Energy conservation principles prevent singly charged metal ions ($M^+$) from generating SEs: the ionization energy of metal atom (5.98 eV (Al)–7.72 eV (Cu)) should be bigger than two times the electron work function of the sputtering target (4.16 eV (Al)–4.58 eV (Cu)), which is not the case [16]. This means that within the spoke SE generation is possible only by the doubly charged metal ions ($M^{2+}$), that have the ionization energy more than three times the electron work function of the sputtering target, e.g. more than $\approx 12$ eV.

The trailing edge of the spoke for Mo, Cr, Al, and Cu exhibits a sharp drop, both in optical (figure 3) and ion signal (figure 3). The abrupt end of the spoke parallel to the magnetic field lines, and the side-way images [11] allow us to speculate that electrons have diffused from CMF lines to open magnetic field lines. Instead, bouncing along the CMF lines, the electrons are then ‘free’ to diffuse towards the substrate (obeying ambipolar diffusion). This interpretation would require transition from abundant SE production to disruption of SE production, which is possible if impinging ions change from Ar dominated to metal dominated flux. Kadlec [14] showed that, due to the gas rarefaction, this transition is very probable for the HiPIMS discharge. As mentioned earlier, the ion flux with predominantly $M^+$ ions cannot generate SEs and disruption is expected. Now, if the impinging ions contain a sufficient amount of $M^{2+}$, the SE generation will continue. However, with lower efficiency, compared to an Ar SE production; indeed the energy required to produce $M^{2+}$ is the sum of the first and the second ionization potentials, and these $M^{2+}$ present a reduced SE yield. For instance, for a given discharge parameters $\gamma$ is 0.08 for Ti$^{2+}$ impinging on a Ti target, compared to 0.12 for Ar$. These SE will excite and ionize the species resulting in a continuous emission (i.e. without forming a sharp edge), but with diminishing intensity due to the already mentioned reduced SE generation efficiency. The critical factor could be an ionization potential of $M^{2+}$. If the second ionization potential is too high, the probability of generating a sufficient amount of $M^{2+}$ ions is reduced and SE generation will be disrupted; therefore, localizing the light intensity and ion production. If the second ionization potential is low enough, then there is a good probability to generate a sufficient amount of $M^{2+}$ ions that will contribute in the production of SEs, resulting in an elongated trailing edge of the light and prolonged ion production. The argument could be expanded by performing analysis based on the electron energy distribution function (EEDF) and electron-impact ionization cross sections of singly charged metal ions. Unfortunately, measurements of electron energies in the CMF region are not available since placing a Langmuir probe within CMF lines strongly disturbs the plasma.
2. Shape of the spoke

The triangular shape of the spokes is observed for the elements (Mo, Cr, Al, Cu) that have second ionization potential higher than the single ionization potential of Ar, figure 3. We postulate that the sharp edge is a consequence of gas rarefaction and suppressed SE production due to insufficient generation of $M^{2+}$ density to continue SE production. Excitation, resulting emission and ionization will diminish once the energetic electrons reach the open magnetic field lines, observed as the ‘jets’ in [11]. A transition to the open magnetic field lines and lack of additional energetic electrons due to an $M^+$ dominated impinging flux results in the sharp edge observed in the emission contour and reduced ion saturation current, figure 3.

The diffuse shape of the spoke is observed for the elements (Ti, Nb) that have a second ionization potential lower than the single ionization potential of Ar, figure 3. The leading edge of the spoke is universal, dominated by electrons generated at the beginning of the spoke. The diminishing trailing edge of elements with low second ionization potential is a result of prolonged SE production, due to sufficiently high $M^{2+}$ ion density, with reduced efficiency resulting in an elongated trailing edge and overall diffusive shape.

In this contribution we postulate a model to describe the observed light inhomogeneities in the HiPIMS discharge, based on the localization of the discharge current. This results in localized SE generation and gas rarefaction. A transition from the $Ar$ dominated to the metal dominated sputtering (due to gas rarefaction) results in reduced SE generation, due to the inability of $M^+$ ions to generate SE from the target. If sufficiently abundant, $M^{2+}$ can generate SE from the target. The critical factor could be the value of the second ionization potential. For elements with second ionization potential higher than the Ar ionization potential (Mo, Cr, Al, Cu), the spoke has a triangular shape with narrow light and ion current signal. For elements with second ionization potential lower than the Ar ionization potential (Ti and Nb), the spoke has a diffuse shape with wide light and ion current signal.

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